# DECAY CONSTANT FOR THE SPONTANEOUS-FISSION PROCESS IN $^{238}U$†

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A long-time (∿ 16 y) exposure experiment was carried out for re-determining the spontaneous-fission decay constant of $^{238}U$, $\lambda_f$. The method consisted of detecting fission fragments on glass plates covered with very adherent, thin (∿ 0.3 μm) $UO_3$ films. From the measured density of etched fission-tracks, the value $\lambda_f = (9.3 \pm 1.0) \times 10^{-17}$ $y^{-1}$ was obtained. Aiming to resolve the apparent disagreement among ∿ 40 $\lambda_f$-values published up to now, a systematic study covering 30 spontaneously fissioning even-even nuclei was made, and led to the following formula for the half-life: $T_{1/2} = 10^{\tau}$ y, with $\tau = (249.2 \pm 0.5) - (6.55 \pm 0.01)Z^2/A + [(1.35 \pm 0.01)Z^2/A - (45.1 \pm 0.3)]\delta M$, where $\delta M$ is the difference (expressed in MeV) between the calculated and experimental masses according to the Droplet Model of atomic nuclei by Myers and Swiatecki. This formula predicts $(8.8 \pm 2.0) \times 10^{-17}$ $y^{-1}$ for $\lambda_f$, which is, within the quoted errors, in good agreement with our experimental result. A discussion regarding the measured $\lambda_f$-values reported in literature is also presented, from which the value $\lambda_f = (8.49 \pm 0.14) \times 10^{-17}$ $y^{-1}$ emmerges as the recommended one for the spontaneous-fission decay constant of $^{238}U$.

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## 1. Introduction

The present work reports the results of an experiment which was carried out in order to re-determine the decay constant, $\lambda_f$, for the spontaneous fission of $^{238}U$. Due to the apparent disagreement which is observed among the experimentally determined $\lambda_f$-values by several research groups, an attempt to resolve such a disagreement, based on an estimated value for $\lambda_f$ which results from a systematic study of all known even-even spontaneously fissioning nuclei, is also presented.

The statistical nature of the spontaneous-fission process makes necessary to collect a sufficiently large number of fission events in order to obtain a satisfactory accurate result when measuring the decay constant $\lambda_f$. In the case under consideration, this requirement is satisfied when long-time exposure experiments, with a large number of $^{238}U$ atoms, are conducted, since previous measurements[1,2]) have indicated values for $\lambda_f$ as small as $10^{-16}$ $y^{-1}$.

We were successful in obtaining a new result for $\lambda_f$ from a long-exposure experiment by using thin films of uranyl oxide ($UO_3$) which were early in 1962 deposited by chemical methods on plane-glass supports. These films had been prepared with the aim of studying low-energy photofission of uranium by detecting the fission fragments with nuclear-track emulsion plates which would be kept close together with the uranium films. Such a study was not proceeded, however, and the uranium films remained stored up to 1978. Since it is well known that the passage of fission fragments through glasses (and other dielectric solids such as mica, makrofol, etc) produces trails of

radiation damage which can be observed with an optical microscope after chemical etching[3]), we decided, therefore, to make use of those glass plates covered with the uranium films for a new determination of $\lambda_f$. One of the main advantages in using glass for registration of fission fragments over other techniques (ionization chamber, nuclear-track emulsion, etc) is its natural ability to discriminate completely fission events against the high-intensity alpha-particle background.

A preliminary estimate of the uranium content of these films gave $\sim$ 0.2 mg-U/cm$^2$ which corresponds to a total exposure of about $10^3$ mg-d of uranium per square centimetre of detector surface. In recent determinations of $\lambda_f$ using glass as nuclear-track detector for fission fragments, equivalent total exposures of $1.3 \times 10^2$ mg-d/cm$^2$ (Emma and Lo Nigro[4])) and $9.5 \times 10^2$ mg-d/cm$^2$ (Thiel and Herr[1])) of uranium were available. Thus, the good statistics which would result in counting fission tracks from our glass plates has encouraged us to carry out a new determination of $\lambda_f$.

An additional motivation for conducting such an experiment was the observation of a remarkable disagreement among the results of the spontaneous-fission decay constant of $^{238}U$ reported up to now, as illustrated in Fig. 1. As we can see, three groups of results for $\lambda_f$ are predominant, corresponding to the approximate values of 7.0, 8.7 and 11.8 units of $10^{-17}$ y$^{-1}$. Although most of the results are distributed around the values $7.0 \times 10^{-17}$ y$^{-1}$ and $8.7 \times 10^{-17}$ y$^{-1}$, even in this case an uncertainty of 20 or 24% results for $\lambda_f$, depending on what value is chosen as the correct one. This uncertainty is of utmost impor-

tance, since it affects directly the results of dating of materials whenever the method of counting of fission tracks (due to fission events from uranium impurities) is used in many applications to several branches of nuclear science and technology[3]). From the phenomenological point of view, a better knowledge of the disintegration rate of $^{238}U$ by spontaneous fission is also important, since it makes feasible to test the validity of the current models for this not yet completely understood process.

## 2. Experimental

The films of uranyl oxide were prepared on hard melting, ordinary glass plates of 8 cm × 15 cm by means of the parlodion ignition method as described by Yagoda[9]). This method gives very stable, strongly adherent films, with the additional advantage of non-tendency to reticulate whenever thicknesses as small as 0.2 mg-U/cm$^2$ are to be obtained. A quite good overall thickness uniformity of the uranium films was ensured by floating the glass plates on a mercury bath. The samples prepared by the above procedure were stored up during $t_6$ = (15.75 ± 0.08) y under normal environmental conditions of temperature, pressure and air relative humidity. The annealing characteristics of fission fragment tracks in ordinary glass as reported by Fleischer *et al.*[3]) enabled us to neglect any thermal track-fading effects in our samples. Some test-samples were analysed in order to search for the percent abundances of the uranium isotopes. The alpha-particle energy-spectrum obtained by using a conventional alpha-spectrometry line (a surface-barrier detector

connected with a multi-channel pulse-height analyser) is shown in Fig. 2, from which the isotopic abundances have been deduced as (99.78 ± 0.03)% of $^{238}U$, (0.22 ± 0.03)% of $^{235}U$ and (0.0025 ± 0.0001)% of $^{234}U$. The indicated low $^{235}U$-isotopic abundance as compared with that of natural uranium led us to rule out the $^{235}U$ thermal-neutron induced-fission method for the determination of the uranium content in the samples. In the present experiment, the number of uranium atoms could be determined with sufficient accuracy from the measured mass-difference of the samples prior and after the removal of the uranium layer.

The $UO_3$ film was dissolved off by immersing the glass plate (∿ 6.2 cm × 2.5 cm × 0.2 cm) into a 10% nitric acid solution at (60.0 ± 0.5)°C. Preliminary tests showed that a 30-min immersion time was adequate to dissolve off completely the uranyl oxide layer (see Fig. 3). The number of $^{238}U$ atoms per unit area, $N$, is related to the measured mass of $UO_3$ removed, $\Delta M$, by

$$N = C \frac{N_o}{M_o} \frac{\Delta M}{S} = (2.1009 \pm 0.0006) \times 10^{21} \frac{\Delta M}{S} \qquad (1)$$

where $C$ is the isotopic abundance of $^{238}U$, $N_o$ the Avogadro's number, $M_o$ the molecular weight of $UO_3$, and $S$ the area over which the uranium film was distributed. A precise measurement of $S$ gave (15.55 ± 0.08) $cm^2$. We were able to determine $\Delta M$ accurately by using a microbalance and careful manipulation of the sample. From a set of measurements we obtained $\Delta M = (3.439 \pm 0.020) \times 10^{-3}$ g. In this way, eq. (1) gives $N = (4.646 \pm 0.052) \times 10^{17}$ $cm^{-2}$, which corresponds to a uranium thickness of (0.1840 ± 0.0021) $mg/cm^2$.

In order to determine the spontaneous-fission decay

constant of $^{238}U$ by the fission-track method it is necessary to relate the number of fission tracks observed per unit area, $\rho_f$, with the number of spontaneous-fission-like events from the $^{238}U$ atoms in the film, which took place during the exposure time $t_f$. Such a relation gives, for $t_f \ll 10^9$ y, the following expression for the decay constant

$$\lambda_f = \frac{1}{N t_f} \left[ \frac{\rho_f - \rho_i}{\varepsilon \, \varepsilon_o} - (N_r + N_n) \right] \quad , \qquad (2)$$

where $\rho_i$ is the number of tracks observed per unit area due to fission events from uranium and thorium impurities of the glass itself, $\varepsilon$ is the detection efficiency for fission fragments coming from a thin extended source lying on the glass surface ($2\pi$-geometry), $\varepsilon_o$ is the counting efficiency, and $N_r$ and $N_n$ are the numbers of fission events per unit area induced in the uranium isotopes by cosmic-ray neutrons and laboratory neutrons ($^{252}Cf$-source), respectively. From data available in literature, it can be deduced that the contributions of $^{234}U$ and $^{235}U$ isotopes to the total number of spontaneous-fission events could be taken negligible at all ($\simeq 0.001\%$ and $\simeq 0.005\%$, respectively).

Optimum conditions for observation of etched tracks at the optical microscope (measurable track-diameter and, at the same time, good distinguishableness from spurious defects of the glass itself) were achieved by etching the glass in a 4.9% HF solution at $(26.0 \pm 0.5)^{\circ}C$ during 15 min. After stopping in a 20% ammonia solution for a few minutes the glass plate was rinsed in distilled water and put to dry. Measurements of etched-track densities ($\rho_f$ and $\rho_i$) and track-diameter were carried out by

using total optical magnifications of 270 × and 560 ×, respectively. Calibrated eyepieces enabled us to obtain accurately the scanning areas for $\rho_f$ and $\rho_i$. The scanning over $(3.175 \pm 0.021)\,cm^2$ uniformly distributed on the surface which was covered with the uranium film yielded a total number of $(853 \pm 51)$ fission tracks, thus corresponding to $\rho_f = (269 \pm 18)\ cm^{-2}$. On the other hand, the scanning over the opposite side gave $\rho_i = (38 \pm 3)\ cm^{-2}$.

Supplementary experiments were conducted in order to determine the angle $\phi_c$ (measured from the glass surface) below which fission fragment tracks fail to register, from which the detection efficiency $\varepsilon$ to be used in eq. (2) could be deduced. Small samples of the glass plate were irradiated with fission fragments from an external $^{252}Cf$-point-source. By using identical etching and scanning conditions as before we inferred the critical angle of etching for fission fragment tracks in ordinary glass as being $\phi_c = (33.5^o \pm 0.9^o)$. This result agrees reasonably well, within that indicated uncertainty, with the obtained by Khan and Durrani[10]). This value of $\phi_c$, together with the measured $UO_3$ thickness, is also indicative of no absorption effects of fission fragments (even in the extreme case of the shortest-range fission fragment travelling through the maximum distance possible) within the uranium film itself, as can be deduced from energy--loss processes of heavy-charged particles in matter[11]). In this way, the quantity $\varepsilon$ will depend only upon the registration efficiency of the glass for fission fragments. But, in this case, $\varepsilon$ is given by $1-\text{sen}\phi_c$ and, therefore, the detection efficiency results to be $\varepsilon = (44.8 \pm 1.3)\%$.

Knowledge of the counting efficiency $\varepsilon_o$ was necessary to infer the "true" track-densities $\rho_f/\varepsilon_o$ and $\rho_i/\varepsilon_o$ due to tracks

occurring in a certain area on each side of the glass. By using the double-scan-coincidence technique[12]) the value $\varepsilon_o$ = (74 ± 4)% could be estimated from the analysis over a given area by two observers under the same criteria of identification of tracks.

Finally, for the sample we have worked with, an estimate of the total number of fissions induced in $^{238}U$ by the fast component of cosmic-ray neutrons and in $^{235}U$ by both the slow and fast components gave ≃ 2 fissions per mg of uranium. On the other hand, the contributions to fission events originating from laboratory-neutron induced-fission in the uranium isotopes amounted to ∿ 98 fissions per mg of uranium. These figures will correspond, in our case, to $N_r \simeq 0.3\ cm^{-2}$ and $N_n \simeq 18\ cm^{-2}$.

## 3. Results and Discussion

Using eq. (2) along with the values of $N$, $T_f$, $\rho_f$, $\rho_i$, $\varepsilon$, $\varepsilon_o$, $N_r$ and $N_n$ from last section, one has $\lambda_f = (9.3 \pm 1.0)\times 10^{-17} y^{-1}$. The quoted uncertainty was estimated taking into account only statistical errors, since the possibility of occurrence of systematic errors at each stage of the experiment was kept as small as possible. Under the conditions of the present experiment the main source of error comes from the uncertainty on the $\rho_f$-determination, although the uncertainties on the quantities $\varepsilon_o$ and $\varepsilon$ be also significant sources of errors.

Our result for the spontaneous-fission decay constant of $^{238}U$, $(9.3 \pm 1.0) \times 10^{-17}\ y^{-1}$, agrees within the experimental errors with those obtained by Thiel and Herr[1]), $(8.57 \pm 0.42) \times 10^{-17} y^{-1}$,

Wagner *et al.*[13]), $(8.7 \pm 0.6) \times 10^{-17}$ $y^{-1}$, Galliker *et al.*[8]), $(8.46 \pm 0.06) \times 10^{-17}$ $y^{-1}$, and also with the recent measurement carried out by de Castro Rizzo[5]), $(10.24 \pm 0.22) \times 10^{-17}$ $y^{-1}$. Within the referred error, the result obtained in the present experiment belongs certainly to that group of measurements which defines the central peak in Fig. 1 (see also Table 1). But in this case there still remains a 25-30% difference between the present result and those centred on the extreme peaks of Fig. 1, as it can be seen by comparing, for instance, with the result reported by Fleischer and Price[14]) ($(6.6 \pm 0.8) \times 10^{-17}$ $y^{-1}$) and a previous measurement carried out in our laboratory[2]) by using the nuclear-track emulsion technique ($(11.6 \pm 0.8) \times 10^{-17}$ $y^{-1}$).

The result obtained in the present experiment does not decide on what value (or values) of $\lambda_f$, among the reported values in literature, can be taken as the most representative one for the decay constant. We were led, therefore, to conduct a systematic study of even-even spontaneously fissioning nuclei in order to obtain an estimated value for $\lambda_f$. The quantity $\tau = \log_{10} T_{1/2}$, where $T_{1/2}$ denotes the spontaneous fission half-life expressed in years, has been plotted against $Z^2/A$ (Z and A standing for the atomic number and the mass number, respectively) for all known even-even isotopes of $Z \geq 90$ (filled circles in Fig. 4). Early in 1955, Swiatecki[15]) has shown that the irregularities from a smooth trend of $\tau_{exp}$ versus $Z^2/A$ could be interpreted as irregularities in the ground-state masses associated with shell structures in the ground-state configurations. We followed this result and the Swiatecki's analysis as a guide to perform a similar systematics of spontaneous

fission half-lives for the even-even isotopes of the elements beyond thorium. The present analysis, however, differs basically from that of Swiatecki[15]) in that the deviations $\delta M = M_{calc} - M_{exp}$ are here taken as the differences between the calculated masses of the nuclei according to the mass-formula by Myers and Swiatecki[16]), including the contribution to the nuclear mass due to the shell effects, and the experimental masses. These later are referred to as the 1971 Atomic Mass Table by Wapstra and Gove[17]). A plot of $\tau_{exp}$ against $\delta M$ defines a series of straight lines, one for each pair of nuclei having approximately the same $Z^2/A$ value, according to the equation

$$\tau_{exp} = \tau_{\delta} + \kappa \delta M \quad , \qquad (3)$$

where both the parameters $\tau_{\delta}$ and $\kappa$ are to be known as functions of $Z^2/A$. In doing so, experimental half-lives were taken from the compilations by Vandenbosh and Huizenga[18]), Randrup *et al.*[19]) and from data quoted in Ref. [20]), whereas the values of $\delta M$ are those reported by Myers[21]). The straight lines obtained in this way indicate that, for the isotopes of a given $Z^2/A$, small $\delta M$ is invariably associated with shorter half-life. In addition, a plot of $\tau_{\delta}$ versus $Z^2/A$ has shown $\tau_{\delta}$ decreasing quite linearly with increasing $Z^2/A$. By means of a least-squares method, a preliminary function $\tau_{\delta} = \delta(Z^2/A)$ was obtained and then inserted into eq. (3), from which the value of the parameter $\kappa$ for each nucleus could be obtained. The results showed a quite linear increase of $\kappa$ with increasing $Z^2/A$. The above conclusions have suggested us to look for a four-parameter empirical formula of the type

$$\tau = a - b\theta + c\theta\delta M \quad , \quad \theta = Z^2/A - d \tag{4}$$

in order to reproduce the experimentally determined spontaneous fission half-lives for the heaviest even-even nuclei. The set of the best values of the parameters $a$, $b$, $c$ and $d$ with which eq. (4) would fit the observed half-lives was determined by means of a least-squares treatment over a total of nearly 80 data on $\tau_{exp}$ and $\delta M$, extending from uranium up to element-104 isotopes. The wide spectrum of the observed half-life $\tau_{exp}$ for $^{238}U$, as reported in Fig. 1, was not included in the analysis, for it might affect to some extent the final result for the calculated half-life of this isotope. Our fitting procedure was carried out by using the method of successive iterations with data rejection, i.e. after each iteration all those $\tau_{exp}$ values whose deviations from the calculated values $\tau_c$ were larger than twice the standard deviation were rejected. Four iterations were needed, resulting in 12% of the experimental data being rejected. The final values of the parameters, when $\delta M$ was expressed in MeV, were obtained as follows:

$$\begin{aligned} a &= 30.48 \pm 0.07 \qquad & c &= 1.35 \pm 0.01 \\ b &= 6.55 \pm 0.01 \qquad & d &= 33.39 \pm 0.01 \end{aligned} \tag{5}$$

from which we have

$$\tau_\delta = (249.2 \pm 0.5) - (6.55 \pm 0.01)Z^2/A \tag{6}$$

$$\kappa = (1.35 \pm 0.01)Z^2/A - (45.1 \pm 0.3) \quad . \tag{7}$$

The results of the present systematic study are summarized in Fig. 4. The straight line (eq. (6)) represents the smooth

trend of the half-lives $\tau_\delta$ versus $Z^2/A$ which are obtained after subtracting the quantity $\kappa\delta M$ from the observed $\tau_{exp}$. The "corrected" values $\tau_\delta = \tau_{exp} - \kappa\delta M$ of the half-lives themselves are represented by open circles which are seen to be fairly well distributed around the straight line, thus indicating that the observed half-lives are well reproduced by formula (4) with the values of the parameters given by (5). The goodness of this adjustment of data can be inferred from the distribution of the ratio $R = T^c_{1/2}/T^e_{1/2}$ between calculated and experimental half-lives (see inserted diagram in Fig. 4). It was deduced that almost 90% of the observed half-lives are reproduced within a factor of five, and 70% within a factor of three. When several $\tau_{exp}$ values were available for a given nucleus, we considered that measured half-life that better agrees with the calculated $\tau_c$ from formula (4). These, together with the measured single-value of the half-life for the other nuclei, were chosen to be shown in Fig. 4. For the above mentioned set of data, it is verified that almost 80% of the experimental half-lives are reproduced within a factor of four. This level of reproducibility might be considered very satisfactory if one looks at the large interval (28 orders of magnitude !) covered by the observed half-lives, the errors involved in individual measurements, fluctuations of the measured half-lives for the same nucleus under consideration and, perhaps to a less extent, the uncertainties associated with the experimental masses. Thus, if one takes $Z^2/A$ as an appropriate parameter for representing the spontaneous-fission half-lives of the even-even nuclei, then the variations in the observed half-lives can adequately be described in terms of the deviations $\delta M$ between calculated and experimental nuclear masses. For instance,

an excess of 1-MeV extra on $\delta M$ causes, in the region of the U-Pu isotopes, the half-life to be increased by about four orders of magnitude, whereas it results in a variation of about ten orders of magnitude for the Fm-No region. As a concluding remark, we point out that although the quantity $\delta M$ defined in the present systematics includes semi-empirical shell corrections to the nuclear masses[21]), its remaining fluctuations are still strongly correlated to the fission half-lives when they are analysed in terms of $Z^2/A$(*).

Formula (4) can now be applied to estimate (within an average uncertainty of about 25%) the spontaneous-fission half-life of any even-even nucleus. In the case of $^{238}U$, for which $\delta M = -0.12$ MeV, it immediately gives $T_{1/2} = (7.9 \pm 1.8) \times 10^{15}$ y, from which one gets $\lambda_\delta = (8.8 \pm 2.0) \times 10^{-17}$ $y^{-1}$. This result compares quite favourably with that obtained in our present experiment in which ordinary glass has been used for detecting fission fragments from an external, thin layer of uranyl oxide. Table 1 shows a comparison between representative values of $\lambda_\delta$ corresponding to the peaks of the $\lambda_\delta$-distribution showed in Fig. 1 and the estimated value as described above. As can be seen there is strong indications for accepting the values of $\lambda_\delta$ belonging to the second group of measurements, for they are the experimental determinations of $\lambda_\delta$ which are in best agreement with

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(*) Of course, this result must be strictly related to the *mechanism* of fission (nuclear deformations, fission barriers, etc), and how the mass deviations $\delta M$ can influence the probability of spontaneous fission through $Z^2/A$ is a matter not easy to understand for the time being. An insight on this subject will be left to a future communication.

the estimated value of the decay constant. Among these, the results reported by Segrè[28]), Spadavecchia and Hahn[31]), von Gunten[32]), Galliker *et al.*[8]) and Thiel and Herr[1]) can, with a high degree of confidence, be taken as the most recommended values, from which the weighted mean $\lambda_f = (8.49 \pm 0.14) \times 10^{-17}\ y^{-1}$ has been derived for the spontaneous-fission decay constant of $^{238}U$.

A much more refined treatment, based on theoretical considerations about the mechanism of spontaneous fission, will clearly be needed in order to decide in a more phenomenological basis on the discrepancies met when measuring the decay constant of long-lived spontaneously fissioning nuclei, of which $^{238}U$ is just an illustrative, but fruitful and important example of such a situation.

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References

1) K. Thiel and W. Herr, Earth. Planet. Sci. Lett. 30(1976)50. See also references quoted therein.

2) H.G. de Carvalho, J.B. Martins, I.O. de Souza and O.A.P. Tavares, An. Acad. brasil. Ciênc. 48 (1976) 205. See also references quoted therein.

3) R.L. Fleischer, P.B. Price and R.M. Walker, *Nuclear Tracks in Solids: Principles and Applications* (University of California Press 1975).

4) V. Emma and S. Lo Nigro, Nucl. Instr. Meth. 128 (1975) 355.

5) D.M. de Castro Rizzo, An. Acad. brasil. Ciênc. 50(1978)303.

6) E.R.V. Spaggiari, *Proceedings of the 31$^{th}$ Annual Meeting of the Sociedade Brasileira para o Progresso da Ciência*, Fortaleza-CE (July, 1979) Summary 355-D.1.

7) Hoff Lu and Hsuan-Ling Tsao, Acta sci. sinica 1 (1952) 77.

8) D. Galliker, E. Hugentobler and B. Hahn, Helv. Phys. Acta 43 (1970) 593.

9) H. Yagoda, *Radioactive Measurements with Nuclear Emulsions* (John Wiley, N. York, 1949) 53; see also H.G. de Carvalho and A.G. da Silva, *Proc. Fourth Inter. Colloq. on Corpuscular Photography*, Munich (1963), Korpuscularphotographie IV (1963) 654.

10) H.A. Khan and S.A. Durrani, Nucl. Instr. Meth. 98(1972)229.

11) See, for instance, L.T. Chadderton and I. McC. Torrens, *Fission Damage in Crystals* (Methuen, London, 1969) 15.

12) S.N. Sokolov and K.D. Tolstov, *Proc. Fourth Inter. Colloq. on Corpuscular Photography*, Munich (1963), Korpuscularphotographie IV (1963) 468.

13) G.A. Wagner, G.M. Reiner, B.S. Carpenter, H. Faul, R. van der Linden and R. Gijbels, Geochim. Cosmochim. Acta 39 (1975) 1279.

14) R.L. Fleischer and P.B. Price, Phys. Rev. 133 (1964) B63.

15) W.J. Swiatecki, Phys. Rev. 100 (1955) 937.

16) W.D. Myers and W.J. Swiatecki, Ann. Phys. (N.Y.) 84(1974)186.

17) A.H. Wapstra and N.B. Gove, Nuclear Data Tables 9 (1971) 265.

18) R. Vandenbosh and J.R. Huizenga, *Nuclear Fission* (Academic Press, N. York, 1973) 46.

19) J. Randrup, S.E. Larsson, P. Möller, S.G. Nilsson, K. Pomorski and A. Sobiczewski, Phys. Rev. C13 (1976) 229.

20) M.R. Schmorak, Nuclear Data Sheets 17 (1976) 391; Y.A. Ellis, Nuclear Data Sheets 21 (1977) 549; Y.A. Ellis and R.L. Haese, Nuclear Data Sheets 21 (1977) 615.

21) W.D. Myers, *Droplet Model of Atomic Nuclei* (Plenum, N. York, 1977).

22) A.H. Wapstra and K. Bos, Atomic Data and Nuclear Data Tables 19 (1977) 185.

23) P.K. Kuroda, R.R. Edwards and F.T. Ashizawa, J. Chem. Phys. 25 (1956) 603.

24) J.H. Roberts, R. Gold and R.J. Armani, Phys. Rev. 174 (1968) 1482.

25) J.D. Kleeman and J.F. Lovering, Geochim. Cosmochim. Acta 35 (1971) 637.

26) H.A. Khan and S.A. Durrani, Radiation Effects 17 (1973) 133.

27) K.N. Ivanov and K.A. Petrzhak, Sov. Atom. Energy 36 (1975) 514 [Engl. Transl. from Atomnaya Energiya 36 (1974) 403].

28) E. Segrè, Phys. Rev. 86 (1952) 21.

29) P.L. Parker and P.K. Kuroda, Journ. Inorg. Nucl. Chem. 5 (1958) 153.

30) S.M.C. de Barros, H.G. de Carvalho and A.G. da Silva, *Proceeding of the Study Group Meeting on the Utilization of Research Reactors*, São Paulo (November, 1973) Session VII, 1-13.

31) A. Spadavecchia and B. Hahn, Helv. Phys. Acta 40 (1967) 1063.

32) H.R. von Gunten, Actinides Rev. 1 (1969) 275.

33) D. Storzer, Thesis, Universität Heidelberg (1970).

34) P.K. Kuroda and R.R. Edwards, Journ. Inorg. Nucl. Chem. 3 (1957) 345.

35) B.D. Kuz'Minov, L.S. Kutsaeva, V.G. Nesterov, L.I. Prokhorova and G.P. Smirenkin, Soviet Phys. JEPT 10 (1960) 290.

36) E.K. Gerling, Yu. A. Shukwhyukov and B.A. Makarodikin, Radiokhimiya 1 (1959) 223.

TABLE 1

Some results concerning the spontaneous-fission decay constant of $^{238}U$(*).

| | Author | Year | Method | decay constant $\lambda_f(10^{-17}y^{-1})$ | Ref. |
|---|---|---|---|---|---|
| Group I(**) | Kuroda *et al.* | 1956 | I/$^{238}$U-equilibrium ratio | 6.7 ± 0.7 | 23 |
| | Fleischer and Price | 1964 | Mica-uranium sandwich | 6.6 ± 0.8 | 14 |
| | Roberts *et al.* | 1968 | Mica-uranium-sandwich | 7.03 ± 0.11 | 24 |
| | Kleeman and Lovering | 1971 | Lexan-uranium-sandwich | 6.8 ± 0.6 | 25 |
| | Khan and Durrani | 1973 | Mica-uranium-sandwich | 6.82 ± 0.55 | 26 |
| | Ivanov and Petrzhak | 1975 | Mica-uranium-sandwich | 7.12 ± 0.32 | 27 |
| | Emma and Lo Nigro | 1975 | Glass-uranium-sandwich | 7.2 ± 0.2 | 4 |
| Group II | Segrē | 1952 | Ionization chamber | 8.60 ± 0.29 | 28 |
| | Parker and Kuroda | 1958 | $^{99}$Mo/$^{238}$U-equilibrium ratio | 8.7 ± 0.5 | 29 |
| | de Barros *et al.* | 1963 | Fission tracks in nuclear-track emulsion | 8.2 ± 0.5 | 30 |
| | Spadavecchia and Hahn | 1967 | Rotating bubble chamber | 8.42 ± 0.10 | 31 |
| | von Gunten | 1969 | Fission products from $^{238}$U | 8.66 ± 0.22 | 32 |
| | Galliker *et al.* | 1970 | Rotating bubble chamber | 8.46 ± 0.06 | 8 |
| | Storzer | 1970 | Fission tracks in dated U-glass | 8.49 ± 0.76 | 33 |
| | Wagner *et al.* | 1975 | Fission tracks in dated U-glass | 8.7 ± 0.6 | 13 |
| | Thiel and Herr | 1976 | Fission tracks in dated U-glass | 8.57 ± 0.42 | 1 |
| | de Carvalho *et al.* | 1980 | Fission tracks in ordinary glass | 9.3 ± 1.0 | this work |
| Group III | Kuroda and Edwards | 1957 | $^{140}$Ba/$^{238}$U-equilibrium ratio | 11.7 ± 0.8 | 34 |
| | Kuz'Minov *et al.* | 1959 | $BF_3$-U-paraffin counter | 10.7 ± 0.5 | 35 |
| | Gerling *et al.* | 1959 | Xe-content of dated U-containing minerals | 11.9 ± 1.0 | 36 |
| | de Carvalho *et al.* | 1975 | Fission tracks in nuclear-track emulsion | 11.6 ± 0.8 | 2 |
| Estimated value | de Carvalho *et al.* | 1980 | Systematics of spontaneous-fission half-lives covering 30 even-even nuclei | 8.8 ± 2.0 | this work |

(*) For a complete list of experimentally determined $\lambda_f$-values see Refs. (1-3,5,6).

(**) The $\lambda_f$-values grouped in this table are in correspondence with the distribution depicted in Fig. 1.

Figure Captions

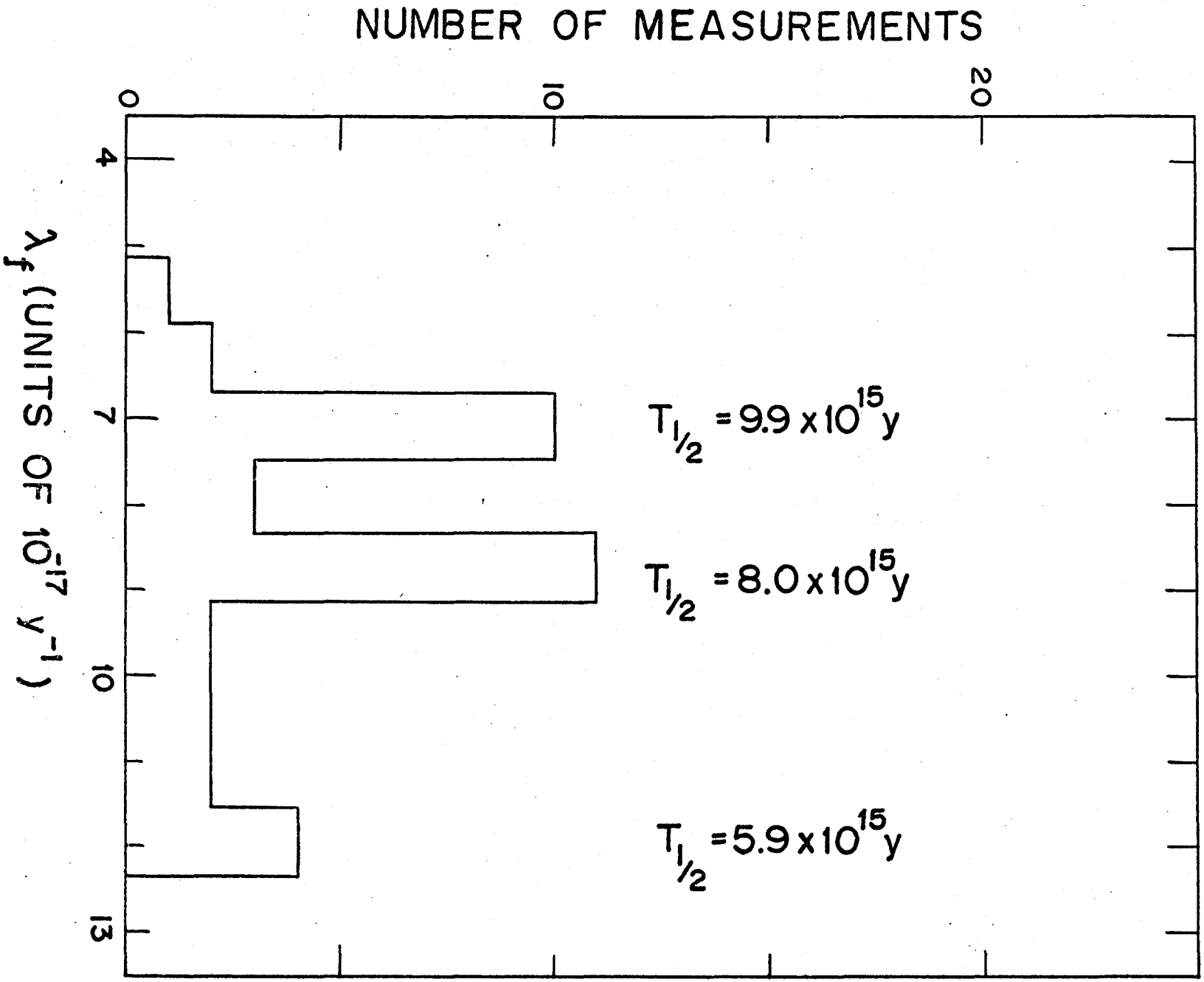
Fig. 1. Frequency distribution of experimentally determined spontaneous-fission decay constant of $^{238}U$, $\lambda_f$, up to now. The histogram was constructed from data taken from the compilations by Fleischer *et al.*[3]), de Carvalho *et al.*[2]) and Thiel and Herr[1]), and the most recent publications by de Castro Rizzo[5]) and Spaggiari[6]). Data published before 1947, as well as the $\lambda_f$-value by Hoff Lu and Hsuan-Ling Tsao[7]) have not been considered since they are far from most of the results by a factor greater than two. In addition to the wide interval covered by the $\lambda_f$-values, three groups of measurements are predominant: ≃ 34% around $7.0\times10^{-17}y^{-1}$, ≃ 36% around $8.7\times10^{-17}\ y^{-1}$ and ≃ 14% around $11.8\times10^{-17}y^{-1}$. The corresponding half-lives $T_{1/2}$ are also indicated. At present, the most accurate value $\lambda_f = (8.46\pm0.06)\times10^{-17}y^{-1}$ reported by Galliker *et al.*[8]) is the recommended one for fission track dating work[1]).

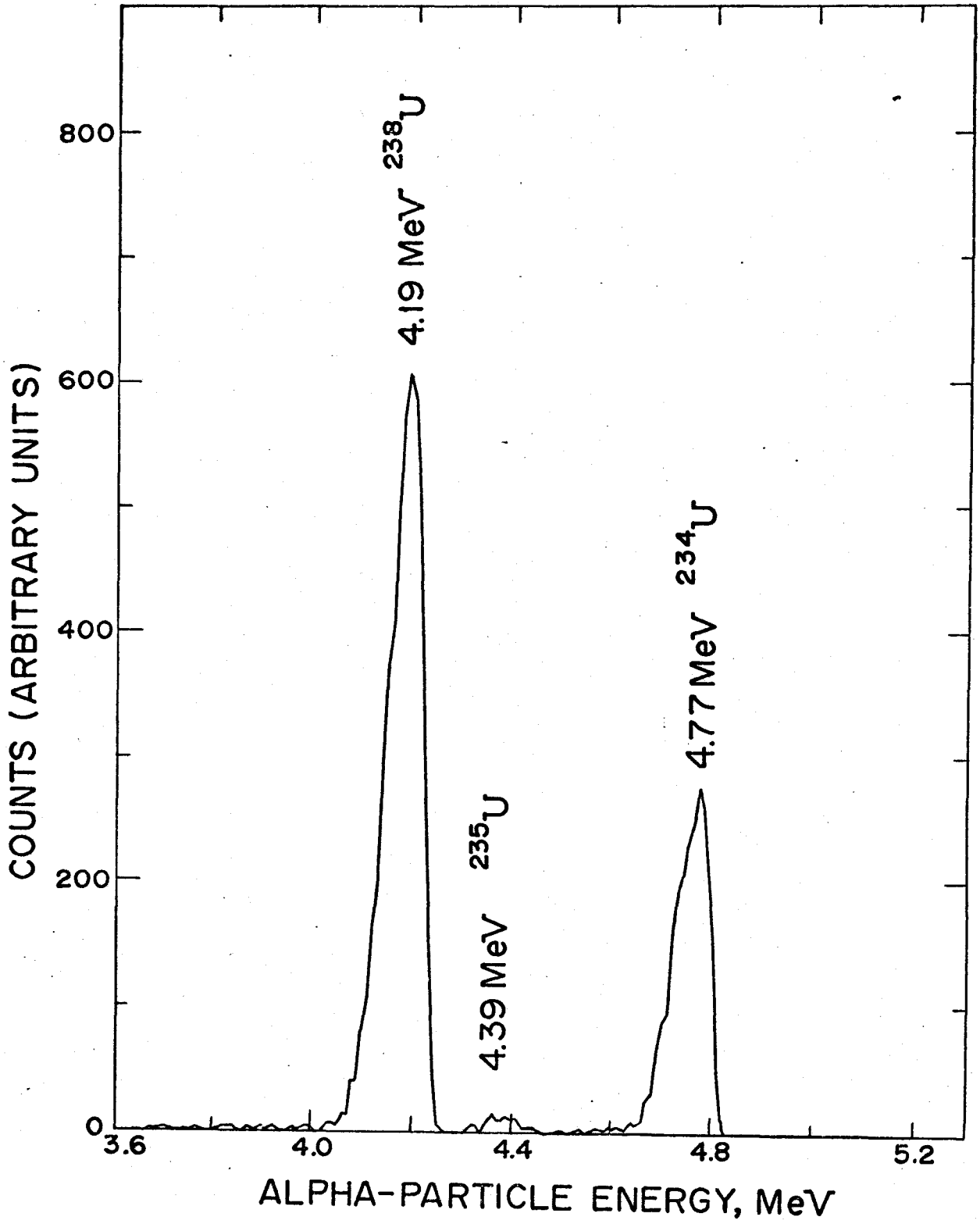
Fig. 2. The energy spectrum of alpha particles resulting from the decay of the uranium isotopes contained in our samples. Separation between the lines belonging to the uranium isotopes is clearly seen. The corresponding peak energies are also indicated.

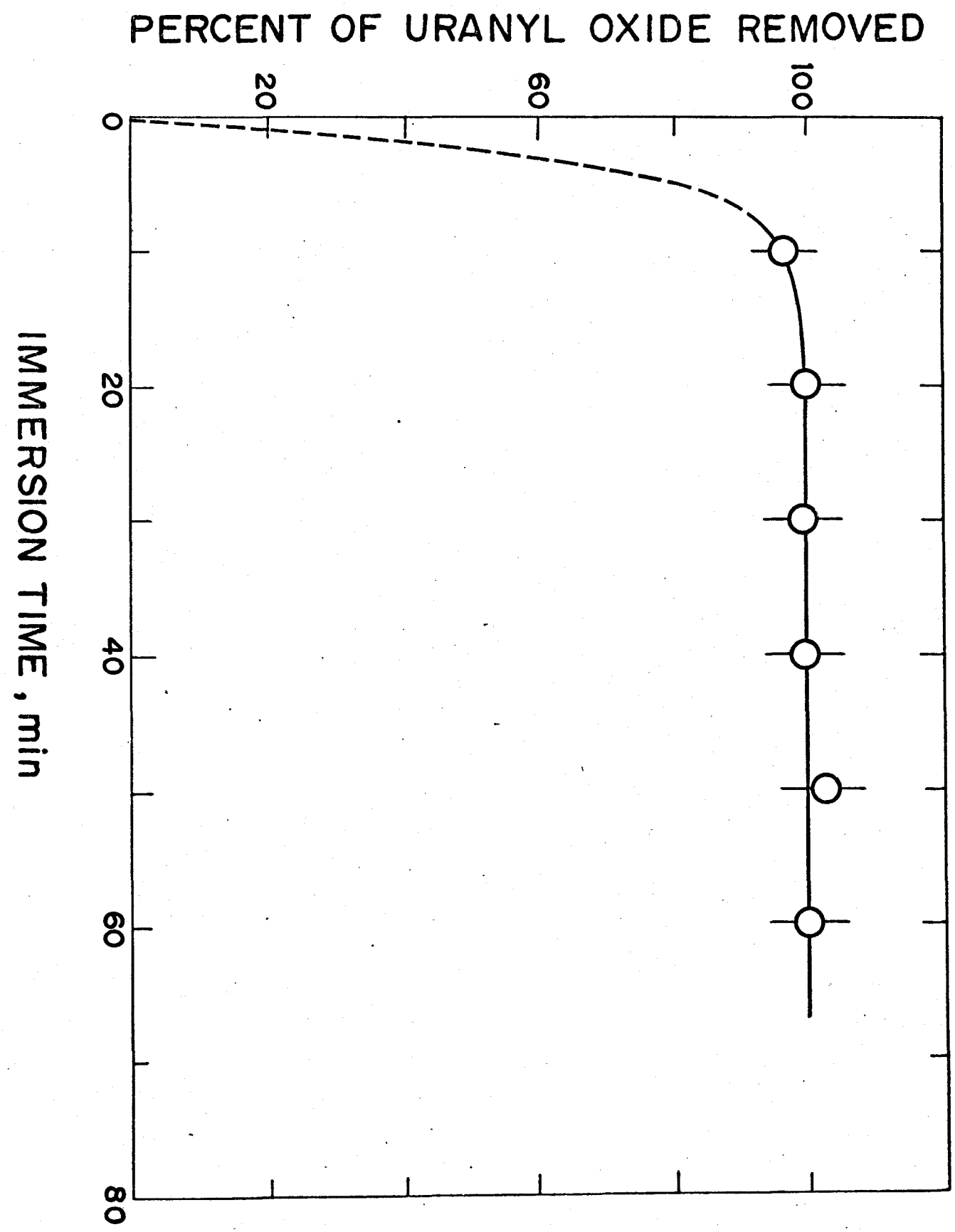
Fig. 3. Fraction (expressed in percent) of the $UO_3$ film removed as a function of the immersion time of the glass plate into a 10% nitric acid solution at 60°C. Points were

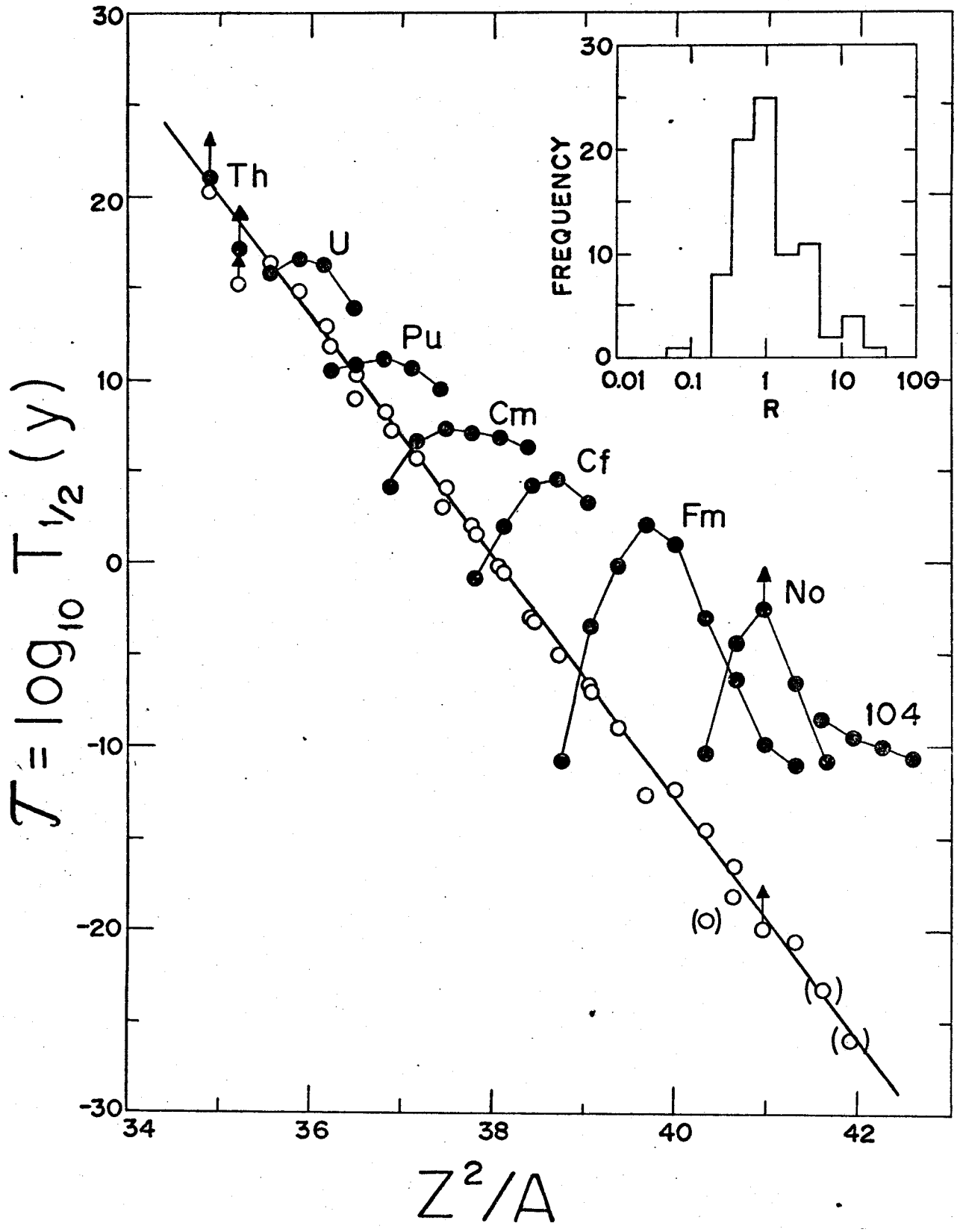
obtained from the measured mass-differences of the glass plate prior and after the immersion by using a microbalance. The curve is an eye-fit through experimental points. It is clearly seen that a 30-min immersion time was adequate to remove completely the uranyl oxide layer from the glass surface.

Fig. 4. Spontaneous fission half-lives of even-even nuclei plotted against $Z^2/A$. The straight line, whose equation is given by (6) in the text, represents the smooth trend of the "corrected" half-lives $\tau_{exp} - \kappa\delta M$ (open circles) which are obtained by subtracting the quantity $\kappa\delta M$ from the observed $\tau_{exp}$ (filled circles). The parameter $\kappa$ was found to vary with $Z^2/A$ according to eq. (7). The mass-differences $\delta M = M_{calc} - M_{exp}$ are those reported by Myers[21]) (last column of his Table of Masses). In the case of nuclei for which experimental masses were not available in Ref.[21]) (points enclosed in parentheses), they were taken from the 1977 Atomic Mass Table by Wapstra and Bos[22]). Experimental half-lives, joined by straight lines for the isotopes of a given element, were taken from Refs.[18-20]). When several $\tau_{exp}$ values were available for the same nucleus, we chose that value corresponding to the lowest deviation from the calculated half-life $\tau_c$ (eq. (4)). The inserted figure shows the frequency distribution of the ratio $R = T^c_{1/2} / T^e_{1/2}$ between the calculated and experimental half-lives for the whole set of input data used in the present analysis.


NUMBER OF MEASUREMENTS
0
10
20
4
7
10
13
$\lambda_f$ (UNITS OF $10^{-17}$ $y^{-1}$)
$T_{1/2} = 9.9 \times 10^{15}$ y
$T_{1/2} = 8.0 \times 10^{15}$ y
$T_{1/2} = 5.9 \times 10^{15}$ y


800
600
400
200
0
COUNTS (ARBITRARY UNITS)
4.19 MeV $^{238}U$
4.39 MeV $^{235}U$
4.77 MeV $^{234}U$
3.6
4.0
4.4
4.8
5.2
ALPHA-PARTICLE ENERGY, MeV


PERCENT OF URANYL OXIDE REMOVED
20
60
100
0
20
40
60
80
IMMERSION TIME, min


30
20
10
0
-10
-20
-30
34
36
38
40
42
$Z^2/A$
$T = \log_{10} T_{1/2}\,(y)$
Th
U
Pu
Cm
Cf
Fm
No
104
(0)
(0)
(0)
FREQUENCY
30
20
10
0
0.01
0.1
1
10
100
R