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Systematical law of spontaneous fission half-lives of heavy nuclei

Chang Xu[1] and Zhongzhou Ren[1,2,*]
[1] *Department of Physics, Nanjing University, Nanjing 210008, China*
[2] *Center of Theoretical Nuclear Physics, National Laboratory of Heavy-Ion Accelerator, Lanzhou 730000, China*

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Systematic calculations on spontaneous fission half-lives of heavy nuclei are carried out by Swiatecki's formula with new parameters and by its generalized form. A new formula with only four parameters is also proposed for spontaneous fission half-lives. Experimental half-lives are well reproduced by the three formulas. We have found from systematic analysis of available data that there is a long lifetime line of spontaneous fission $N = Z + 52$ for heavy elements with $Z \geqslant 90$. The new formula can be used to predict the spontaneous fission half-lives of heavy nuclei not far away from this long lifetime line. The small deviation between experimental half-lives and theoretical ones from the new formula is analyzed and discussed.

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Spontaneous fission of heavy nuclei was first predicted by Bohr and Wheeler in 1939 [1] and observed by Flerov and Petrjak in 1940 [2]. It is a prominent decay mode besides α and β decays for elements heavier than thorium. Since the discovery of spontaneous fission of ^{238}U [2], various theoretical approaches, both phenomenologically and microscopically, have been used to describe spontaneous fissions [3–6]. Microscopic calculation of spontaneous fission half-lives is very difficult due to both the complexity of the fission process and the uncertainty of the height and shape of the fission barrier [6]. Swiatecki and coworkers made much of the pioneering work in the liquid drop model of fission. In 1955, Swiatecki proposed a semiempirical formula for spontaneous fission half-lives [4,5]. By including the deviation of experimental ground state masses from a smooth liquid drop model reference surface, Swiatecki successfully reproduced the experimental data with a formula [4]. Although new experimental data of spontaneous fission have been accumulated in past years, systematic analysis of these data has not been completed. In this paper, we calculate spontaneous fission half-lives by Swiatecki's formula with renewed parameters and by its generalized form. Based on detailed analysis of experimental data, we have found that there is a long lifetime line of spontaneous fissions with $N = Z + 52$ for $Z \geqslant 90$ isotopes. A new formula for spontaneous fission half-lives with four parameters is proposed. The experimental data can be well reproduced by these formulas.

Swiatecki shows that there is a smooth trend in spontaneous fission half-lives with Z^2/A, and it can be written as [4]

$$\log_{10}(T_{1/2}) = c_1 + c_2\left(\frac{Z^2}{A}+k\right) + c_3\left(\frac{Z^2}{A}+k\right)^2 + c_4\left(\frac{Z^2}{A}+k\right)^3 + \left(c_5 + \left(\frac{Z^2}{A}+k\right)\right)\delta M, \tag{1}$$

where δM is the deviation between experimental ground state masses and theoretical ones from the smooth liquid drop model $\delta M = M_{\mathrm{exp}} - M_{\mathrm{the}}$.

In the above expression the unit of δM is mMU [4] where the mass of ^{16}O was used as the standard of mass. We now use the mass of ^{12}C as the standard of mass and therefore new parameters are needed in Swiatecki's formula. Another motivation to use new parameters is that much new information on spontaneous fission has been observed in recent years. It is also useful to test the validity of Swiatecki's formula for the new data and to search for a systematical law of the data.

In our calculations, the experimental ground state masses M_{exp} are taken from the 2003 atomic mass table by Audi and Wapstra [7]. The semiempirical mass surface is $M_{\mathrm{the}} = ZM(^1H) + NM(n) - B/c^2$ [8], where $M(^1H)$ and $M(n)$ are experimental masses of hydrogen and neutron, respectively. B is the binding energy from the liquid drop model [8]:

$$B = 15.56A - 17.23A^{\frac{2}{3}} - 0.7\frac{Z^2}{A^{\frac{1}{3}}} - 23.28\frac{(N-Z)^2}{A} + \delta\frac{12}{A^{\frac{1}{2}}}. \tag{2}$$

Through a least-square fit to the available spontaneous fission data of 33 even-even nuclei with known experimental masses (^{232}Th $-^{264}$ Hs), we obtain a new set of parameters for Eq. (1). Their values are: $c_1 = 24.350359; c_2 = -7.839937; c_3 = 0.325838; c_4 = 0.0148211; c_5 = -8.875158; k = -33.749512$. These values are close to the original ones used by Swiatecki [4]. In order to use Swiatecki's formula to calculate the spontaneous fission half-lives of the nuclei with unknown masses, we consider a generalized form of Swiatecki's formula. We replace the deviation δM in the formula by the following expression associated with shell closures $Z = 82$ and $N = 126$: $\delta M = c_6(Z-82)^2 + c_7(N-126)^2 + c_8(N-Z)$. The simple expression approximately represents the influence of shell corrections on spontaneous fission half-lives. So a

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[*] Email address: zren@nju.edu.cn.

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TABLE I. Logarithm of spontaneous fission half-lives (in years) calculated by the three formulas.

Fission	$T_{\text{Expt.}}$	$T_{\text{Form.1}}$	$T_{\text{Form.2}}$	$T_{\text{Form.3}}$	Fission	$T_{\text{Expt.}}$	$T_{\text{Form.1}}$	$T_{\text{Form.2}}$	$T_{\text{Form.3}}$
^{232}Th	21.08	20.76	20.41	21.08	**^{248}Cf**	4.51	4.12	3.72	3.58
^{234}U	16.18	15.85	15.79	16.18	^{250}Cf	4.23	4.13	3.17	3.17
^{236}U	16.40	16.24	16.30	16.36	^{252}Cf	1.93	1.76	1.62	1.68
^{238}U	15.91	16.03	16.11	16.26	^{254}Cf	−0.78	−0.98	−0.79	−0.86
^{236}Pu	9.18	9.54	9.94	10.46	^{246}Fm	−6.60	−4.02	−6.39	−6.56
^{238}Pu	10.68	11.21	11.47	11.41	^{248}Fm	−2.94	−2.81	−2.65	−2.88
^{240}Pu	11.06	11.71	12.01	11.78	^{250}Fm	−0.10	−1.21	−0.55	−0.63
^{242}Pu	10.83	11.21	11.71	11.57	**^{252}Fm**	2.10	0.27	0.08	0.22
^{244}Pu	10.82	10.42	10.71	10.81	^{254}Fm	−0.20	−1.17	−0.59	−0.29
^{240}Cm	6.28	5.99	5.47	5.53	^{256}Fm	−3.48	−3.17	−2.43	−2.13
^{242}Cm	6.85	6.90	7.18	6.94	^{252}No	−6.54	−6.03	−5.93	−6.12
^{244}Cm	7.12	7.41	7.76	7.47	**^{254}No**	−3.04	−4.77	−3.63	−3.48
^{246}Cm	7.26	7.66	7.34	7.17	^{256}No	−4.77	−4.97	−2.96	−2.48
^{248}Cm	6.62	7.01	6.06	6.03	^{256}Rf	−9.71	−8.39	−8.47	−8.56
^{250}Cm	4.05	3.97	4.07	4.10	^{260}Sg	−9.65	−10.11	−10.08	−10.10
^{242}Cf	−1.33	0.07	−2.16	−1.95	^{264}Hs	−10.20	−10.42	−10.59	−10.64
^{246}Cf	3.26	2.78	3.11	2.88					

generalized form of Eq. (1) is written as

$$\log_{10}(T_{1/2}) = c_1 + c_2\left(\frac{Z^2}{A}+k\right) + c_3\left(\frac{Z^2}{A}+k\right)^2 + c_4\left(\frac{Z^2}{A}+k\right)^3 + \left(c_5 + \left(\frac{Z^2}{A}+k\right)\right) \times (c_6(Z-82)^2 + c_7(N-126)^2 + c_8(N-Z)). \tag{3}$$

The parameters of Eq. (3) are also obtained through a least-square fit to the experimental data: $c_1 = 31.196159$; $c_2 = -5.086737$; $c_3 = -0.0742314$; $c_4 = -0.161829$; $c_5 = 0.0398652$; $c_6 = 0.0585024$; $c_7 = -0.0124953$; $c_8 = 0.108390$; $k = -30.444904$.

The numerical results are given in Table I and Fig. 1. In Table I the first column denotes nuclides. The second column is the logarithm of experimental spontaneous fission half-lives [7,9]. The numerical results from Eq. (1) (Form.1 in Table I) and from Eq. (3) (Form.2) are given in column 3 and column 4. The results calculated by a new formula (Form.3) are listed in column 5. We discuss the new formula below. It can be seen from columns 3–5 that the half-lives from the formulas agree very well with the experimental values. The deviation between the data and the calculated values is less than 0.5 for many nuclei and this means that the calculated half-lives from the formula agree with the data within a factor of 3. The big deviation occurring for ^{252}Fm is mainly due to the subshell effect at $N = 152$. The average deviations between experiment and formula are $S = \sum |T_{\text{Expt.}} - T_{\text{Calc.}}|/33 = 0.5714$ for Swiatecki's formula and 0.6041 for its generalized form. The average deviation is 0.5535 for the new formula.

In order to show the level of agreement between experimental data and the formulas we also draw the variations of the theoretical half-lives and the experimental ones in Fig. 1. It can be seen from Fig. 1 that the theoretical curves are close to the experimental ones for many nuclei. So a quantitative description of the half-lives is obtained with the three formulas.

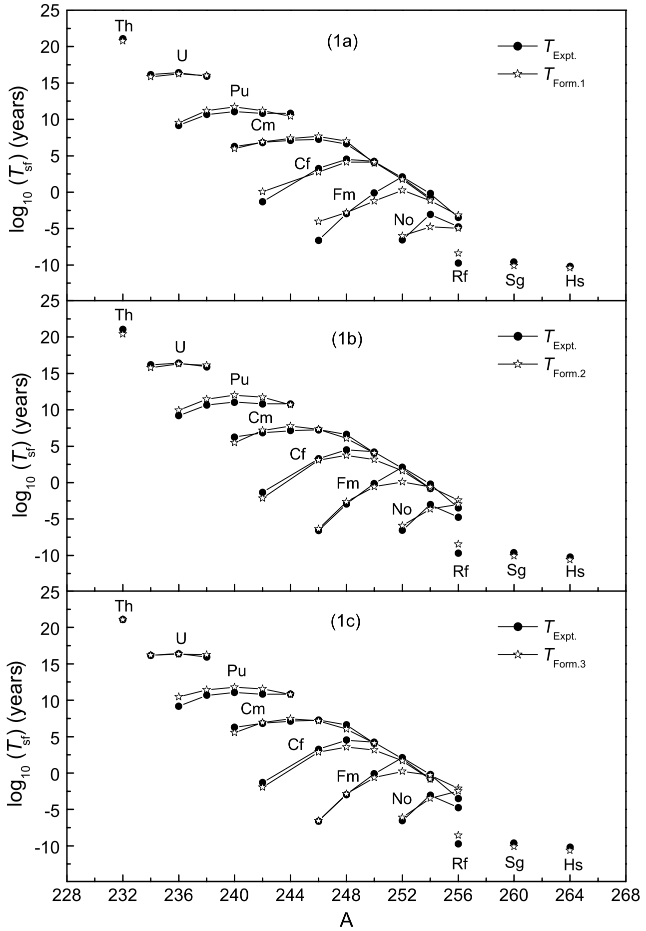

FIG. 1. Comparison of experimental spontaneous fission half-lives with theoretical ones, where three sets of numerical results are plotted (a, b, c).

After comparing the theoretical results of spontaneous fission half-lives with experimental ones, we now discuss the origin of the new formula of spontaneous fission half-lives. The new formula is

$$\log_{10}(T_{1/2}) = 21.08 + c_1 \frac{Z-90}{A} + c_2 \frac{(Z-90)^2}{A} + c_3 \frac{(Z-90)^3}{A} + c_4 \frac{Z-90}{A}(N-Z-52)^2, \tag{4}$$

where the value 21.08 is the logarithm of the experimental spontaneous fission half-life (in years) of ^{232}Th, and the number 52 in the last term is the neutron excess of ^{232}Th. The parameters in the above formula are also obtained through a least-square fit to the half-lives of 33 even-even nuclei in Table I: $c_1 = -548.825021; c_2 = -5.359139; c_3 = 0.767379; c_4 = -4.282220$.

It is well known that the occurrence of spontaneous fission of heavy nuclei is due to the increase of repulsive Coulomb interactions when the proton number increases. The spontaneous fission half-life of ^{232}Th is the longest in all known nuclei and it is used as a reference point of the new formula (the role of ^{232}Th in the new formula is like that of ^{208}Pb in the interaction boson model, the nuclear shell model, and the mean-field model). We expect that the decrease of spontaneous fission half-life from ^{232}Th to heavy nuclei (Z > 90) is directly related to the increase of the effective valence proton number $(Z-90)$. Available data on experimental half-lives show that there is a maximum of spontaneous fission half-lives for an isotopic chain (boldface in Table I). The maximums for different elements lie approximately on a straight line with $N = Z + 52$ where the number 52 is the neutron excess of ^{232}Th. We call the line $N = Z + 52$ the long lifetime line of heavy nuclei with $Z \geqslant 90$. The half-lives of nuclei on the line can be represented as a sum of the terms $(Z-90)$, $(Z-90)^2$, and $(Z-90)^3$ where the dominant term is $(Z-90)$. The terms $(Z-90)^2$ and $(Z-90)^3$ are introduced as the minor corrections to this linear relation. In order to describe the variation of the half-lives on an isotopic chain, we include a parabolic term $(N-Z-52)^2$. In this way the new formula can be obtained.

Now we try to derive the new formula of spontaneous fission half-lives. Because spontaneous fission is a pure quantum tunneling effect, we extend the formula of half-lives of α decay and of cluster radioactivity to that of spontaneous fission. There is the Viola-Seaborg formula for half-lives of α decay [10]. For the half-lives of cluster radioactivity a formula similar to the Viola-Seaborg formula is [11]

$$\log_{10}(T_{1/2}) = a\, Z_1 Z_2\, Q^{-1/2} + c Z_1 Z_2 + d + h. \tag{5}$$

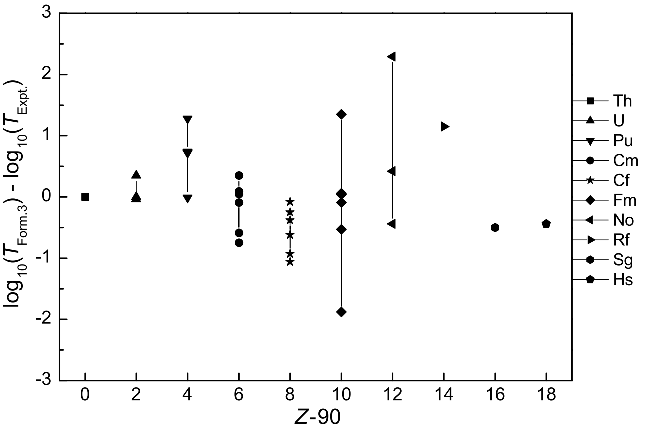

FIG. 2. The variation of the deviation between experimental values and calculated ones (Form.3) with Z-90.

We assume that the above equation is valid for half-lives of spontaneous fission, but the values of its parameters may be different from those of cluster radioactivity. For spontaneous fission it is difficult to define the released energy Q due to the existence of different fission channels (i.e., different kinds of fragments). So we assume $Z_1 \approx Z_2 \approx Z/2$ and try to express the energy Q with charge and mass numbers of fissionable nuclei. There is an expression for the kinetic energy released in fission of a nucleus: $Q = 0.1240\, Z^2/A^{1/3}$ [5]. Substituting this into Eq. (5), we have

$$\log_{10}(T_{1/2}) \approx C_1 Z + C_2 Z^2 + C_3, \tag{6}$$

where the quantity $A^{1/3}$ is approximately a constant for heavy nuclei in mass range $A = 232 - 300$. With the above equation we have

$$\log_{10}(T_{1/2}) - \log_{10}(T_{1/2}(^{232}\text{Th})) = C_4 (Z-90) + C_5 (Z-90)^2. \tag{7}$$

Considering the dependence of half-lives on mass number, we have added the terms $(Z-90)^3$ and $(N-Z-52)^2$. We approximately obtain the new formula [see Eq. (4)].

Equation (4) (i.e., Form.3) is a simple formula of half-lives for the very complex spontaneous fission which is a multidimensional tunneling effect of a quantum many-body system. It can be seen from Fig. 1 that the main correlations have been included in Eq. (4). However, there should be some residual correlations beyond this simple formula. In order to see these possible correlations clearly, we have plotted the variation of the deviations between experimental half-lives and theoretical ones ($\log_{10} T_{\text{Form.3}} - \log_{10} T_{\text{Expt.}}$) with the variables $Z-90$ and $N-Z-52$ in Figs. 2 and 3, respectively.

Figures 2 and 3 show the variation of the deviations between experimental half-lives and the values from the new formula with $Z-90$ and $N-Z-52$. It can be seen again that the theoretical values are close to experimental ones. The mean-square deviation with the new formula is 0.6070, and this corresponds to an average deviation of experimental half-life with a factor of 4. The experimental half-lives are reasonably reproduced by the new formula. The biggest deviation between experiment and formula is approximately a factor of 10^2 near ^{252}Fm, which is still acceptable due to the influence of the deformed subshell effect at $N = 152$. It is concluded from Figs. 2 and 3 that many points lie near the line $\log_{10} T_{\text{Form.3}} - \log_{10} T_{\text{Expt.}} = 0$ (i.e., $T_{\text{Form.3}} = T_{\text{Expt.}}$). The distributions of the points are scattered. We have tried to find systematic behavior for the deviations in Figs. 2 and 3, but we have not found it with the available

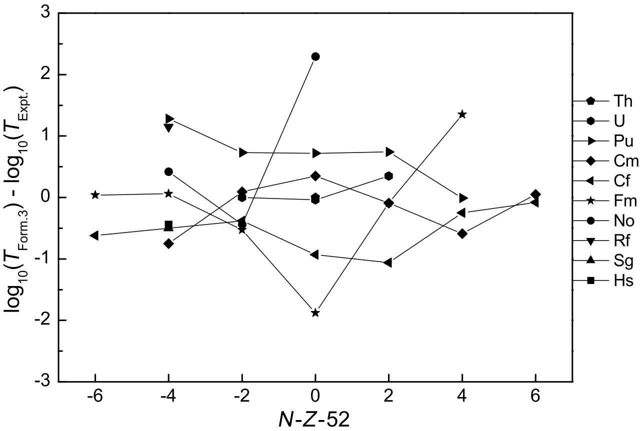

FIG. 3. The variation of the deviation between experimental values and calculated ones (Form.3) with N-Z-52.

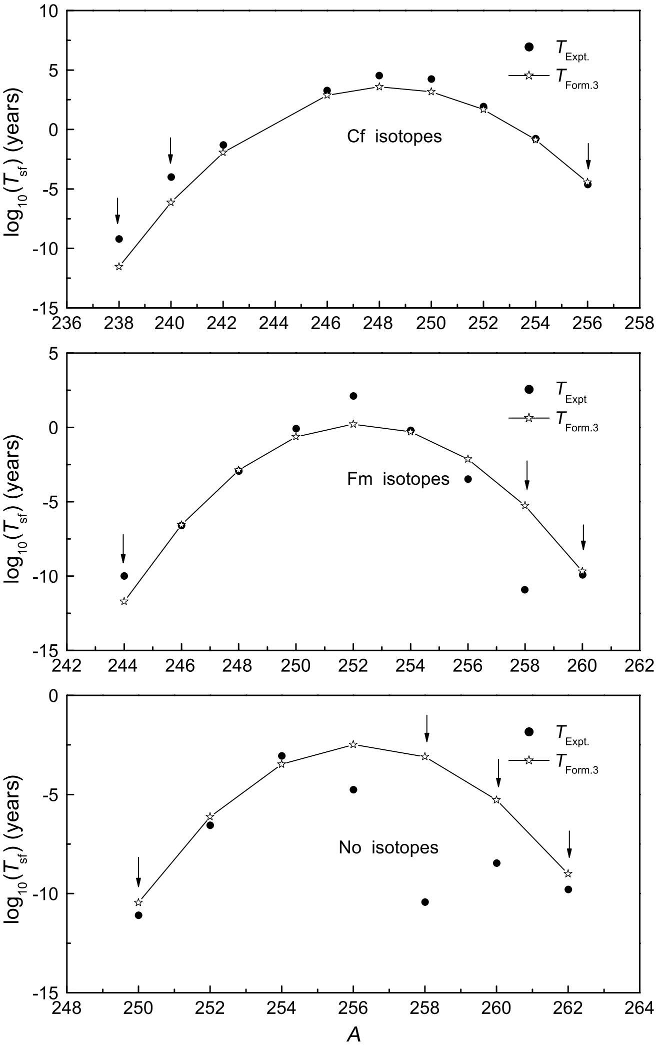

FIG. 4. Experimental and calculated fission half-lives for Cf, Fm, and No isotopic chains.

experimental data. With the accumulation of more and more experimental data in the future, it will be very interesting to investigate a possible law of the deviations between formula and data. We believe that the systematic behavior of the deviations is very useful for further improvement of the above formula and for more reliable predictions for new experiments.

Since the new formula is only dependent on mass and charge numbers, it can be used to predict the half-lives of nuclei not far away from the long lifetime line even if their masses are unknown. At present we limit our calculations of spontaneous fission half-lives to nuclei not far away from the long lifetime line [11–13]. Figure 4 shows the variations of the experimental half-lives and the theoretical ones for Cf, Fm, and No isotopic chains where the arrows denote the nuclides where experimental masses are still unknown. It is seen from Fig. 4 that the theoretical half-lives of Cf isotopes agree very well with experimental ones even for nuclei for which experimental masses are not available now. For Fm and No isotopes, the trends of the experimental data are reasonably reproduced, although large deviations between experiment and formula exist for two nuclides (^{258}Fm and ^{258}No). We also use the generalized form of Swiatecki's formula [Eq. (3)] to calculate the half-lives of the above isotopic chains and reach reasonable agreement.

Spontaneous fission half-lives of even-even $Z = 104$-110 isotopes are calculated by the new formula and listed in Table II. There are seven experimental data points for these nuclei. The results from the formula agree reasonably with five data points within a factor of 10^2. For example, the experimental spontaneous fission half-life of ^{264}Hs is 1 ms in the mass table [7] or 2 ms in Ref. [9]. The theoretical half-life of ^{264}Hs is 0.7 ms, and this agrees well with the data. But for ^{258}Rf and ^{260}Rf the deviations between the data and the formula are as high as a factor of 10^4, and this is similar to those of ^{258}Fm and ^{258}No in Fig. 4. Möller *et al.* [6] consider that a deviation such as 10^{4-5} is still acceptable for spontaneous fission half-lives due to the uncertainty of various factors. In the future it will also be interesting to search for new physics of this sudden deviation from both the experimental and theoretical sides.

In summary, we systematically test the validity of Swiatecki's formula with new parameters for new experimen-

TABLE II. Half-lives of spontaneous fission from the new formula.

Fission	$T_{\mathrm{Expt.}}$	$T_{\mathrm{Form.3}}$	Fission	$T_{\mathrm{Expt.}}$	$T_{\mathrm{Form.3}}$
^{256}Rf	6.2 ms	87.1 ms	^{260}Sg	7 ms	2.5 ms
^{258}Rf	14 ms	1.5 min	^{262}Sg		6.0 s
^{260}Rf	20 ms	20.3 min	^{264}Sg		1.8 min
^{262}Rf	2.1 s	3.9 min	^{266}Sg	31.8 s	15.7 s
^{264}Rf		0.7 s	^{268}Sg		21.5 ms
^{264}Hs	2 ms	0.7 ms	^{268}Ds		2.6 ms
^{266}Hs		3.8 s	^{270}Ds		28.4 s
^{268}Hs		1.4 min	^{272}Ds		13.6 min
^{270}Hs		9.8 s	^{274}Ds		1.2 min
^{272}Hs		6.2 ms	^{276}Ds		20.9 ms

tal data of spontaneous fission. We also obtain a generalized form of Swiatecki's formula with the deviation δM replaced by an expression associated with shell closures $Z = 82$ and $N = 126$. We have found a long lifetime line of spontaneous fissions with $N = Z + 52$ for $Z \geqslant 90$ isotopes based on systematic analysis of experimental data, and a new formula of spontaneous fission half-lives with four parameters is proposed. Experimental data are well reproduced by the three formulas. The new formula is only dependent on mass and charge numbers. It can be used to predict the half-lives of nuclei not far away from the long lifetime line even if their masses are unknown. Spontaneous fission half-lives of the isotopes of $Z = 104$-110 (Rf-Ds) are predicted, and these values are consistent with current experimental facts.

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