from a medium energy (e.g., 5 Mev), passes through the energy zero and is eventually captured in a $K$ orbit around some nucleus. Its lifetime in such an orbit is crudely estimated by taking for $N$ in the above formula the reciprocal volume of a sphere of radius equal to that of the $K$-shell, multiplied by the number of protons in the nucleus. For carbon, $N$ is about $10^{38}$ cm$^{-3}$ and the lifetime against annihilation after the negative proton is captured in the $K$-shell is of the order $10^{-20}$–$10^{-22}$ sec. Most of the negative protons reaching low energies would therefore be annihilated after capture in the $K$-shell, but the general features of the annihilation event as far as the products are concerned, would remain unchanged.

In summary, therefore, we would consider it to be useful to search for high energy annihilation events in material (sensitive emulsions for example) exposed to the cosmic radiation at 50 to 100 g/cm$^2$ below the top of the atmosphere, and to look for annihilation at rest at several hundred g/cm$^2$ below the top where the negative protons may be brought to a stop in an absorber.[15]

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# $U^{235}$ Fission Yields in the Rare Earth Region

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The $U^{235}$ fission yield curve for uranium fuel rods from mass number 143 to 160 has been measured, utilizing mass spectrometric techniques. Eight percentage yields are assumed in this region, and on the basis of measurements of the neodymium, samarium, europium, and gadolinium fission isotopes, nine other yields are obtained. The resulting yield curve is smooth. Based on the same assumptions which give the smooth yield curve the following cross sections and half-lives are obtained: Cross sections, $Sm^{149}$, 47,000 barns; $Sm^{151}$, 7200 barns; $Gd^{155}$, 41,000 barns; $Gd^{157}$, 59,500 barns; half-lives, $61^{147}$, 2.26 yr.; $Sm^{151}$, 122 yr.

## I. INTRODUCTION

ONE of the problems associated with the fission process is the determination of the fission yield curve for fissionable nuclei at various neutron energies. To date there is no satisfactory explanation of the commonly observed asymmetric fission. On the experimental side a great deal of work has been done in measuring the yields[1] in the fission of $U^{235}$. Almost all of these yields have been obtained by measuring, with radiochemical methods, the relative number of atoms in the various radioactive chains. To obtain relative yields in this manner the data must be corrected for the half-lives of the isotopes measured and for the relative counter efficiencies for radiations of different energies. In many cases the corrections are difficult or uncertain. An independent and more accurate method has been utilized by Thode and Graham[2] to measure the relative yields of krypton and xenon isotopes in $U^{235}$ fission. They measured the relative abundances of the stable fission products of these elements with a mass spectrometer.

This method is applicable to the entire fission yield curve if necessary corrections are made for neutron absorption in each chain and for long-lived isotopes earlier in that chain. The partial yield curves obtained for various elements can be fitted together by the method of isotopic dilution previously described by the authors.[3] In addition, if the $U^{235}$ depletion is measured, absolute fission yields can be obtained. The mass spectrometer method has been used here to obtain the $U^{235}$ fission yield curve for pile neutrons in the rare earth region. However, because the rare earth samples available to us had been partially depleted of lower atomic number components by resin column extraction, the method of isotopic dilution could not be used to fix the relative positions of the partial curves due to different elements. Thus the yield of one isotope of each element considered was assumed to be consistent with the curve of the previous element. For the first element considered, neodymium, the assumption of absolute yield was made on the basis of chemical data. Thus the relative values obtained, since they depend only on the mass spectrometer measurement, are considerably better than are the absolute values. A further difficulty in the experiment was that certain half-lives and cross sections necessary to deduce the relative yields were not known accurately. Here again certain yields were assumed on the basis of the rest of the curve and the unknown cross sections and half-lives were deduced. Table I gives the final data on fission yields. These data are compared with the fission yields from radio-chemical data in Fig. 1.

[1] Plutonium Project, "Nuclei formed in fission," Rev. Mod. Phys. **18**, 513 (1946).
[2] H. G. Thode and R. L. Graham, Can. J. Research, **A25**, 1 (1947).
[3] Hayden, Reynolds, and Inghram, Phys. Rev. **75**, 1500 (1949).

## II. NEODYMIUM

Fission neodymium was obtained from Parker and Lantz of Oak Ridge National Laboratory. It was in a sample of rare earth materials which had been separated from uranium and which had had the lighter rare earths removed by column extraction. The fission product mixture so obtained (neodymium and heavier rare earths) was analyzed by means of a filament source mass spectrometer using techniques previously described.[4] The results of this analysis are given in Table II.

As has been shown by the authors[5] the only appreciable absorber in normal neodymium is $Nd^{143}$ which has an isotopic cross section of 240 barns. Thus the 143-peak here observed was lower, and the 144-peak was higher, than the pure fission yield would predict. The number of atoms of $Nd^{143}$ and $Nd^{144}$ present at time $t$ are

$$N_{143}(t)=\frac{N_u\sigma_u f_{143}\Phi t}{\sigma_{143}\Phi t}[1-\exp(-\sigma_{143}\Phi t)], \quad (1)$$

$$N_{144}(t)=N_u\sigma_u f_{144}\Phi t+\frac{N_u\sigma_u f_{143}\Phi t}{\sigma_{143}\Phi t}\times[\sigma_{143}\Phi t-1+\exp(-\sigma_{143}\Phi t)], \quad (2)$$

where $N_u$ is the number of uranium atoms and $\sigma_u$ is the fission cross section of uranium. When ratios of the abundance equations are taken these quantities always cancel out. The quantities $f$ are the fractional fission yields, $\sigma_{143}$ is cross section of $Nd^{143}$ and $\Phi$ is the neutron flux. Other yields are given by

$$N_j(t)=N_u\sigma_u f_j\Phi t, \quad j=145,\ 146,\ 148,\ 150. \quad (3)$$

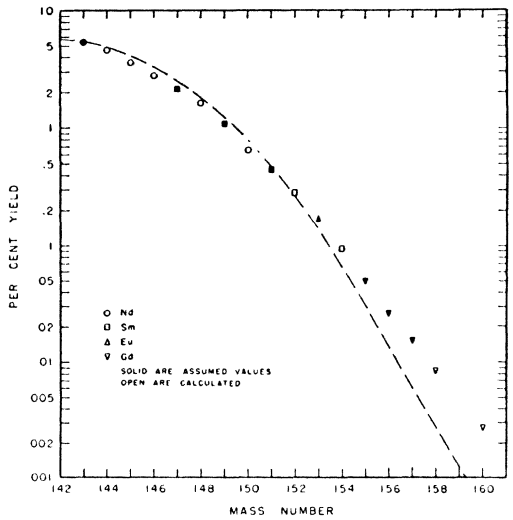


FIG. 1. The fission yield curve in the mass range 143→160 as determined by the mass spectrometer. For comparison the radiochemically determined fission yields (reference 1) are shown as the dashed curve, which is fitted to the data at mass 143.

[4] Inghram, Hayden, and Hess, Phys. Rev. **72**, 967 (1947).
[5] D. C. Hess, Jr. and M. G. Inghram, Phys. Rev. **76**, 300 (1949).

TABLE I. Fission yields in percent (the underlined values are those assumed to be known).

| Mass | Nd | Sm | Eu | Gd |
|---|---|---|---|---|
| 143 | 5.40 | | | |
| 144 | 4.64 | | | |
| 145 | 3.62 | | | |
| 146 | 2.81 | | | |
| 147 | | 2.15 | | |
| 148 | 1.64 | | | |
| 149 | | 1.10 | | |
| 150 | 0.658 | | | |
| 151 | | 0.445 | | |
| 152 | | 0.279 | | |
| 153 | | | 0.170 | |
| 154 | | 0.0908 | | |
| 155 | | | | 0.0500 |
| 156 | | | | 0.0260 |
| 157 | | | | 0.0150 |
| 158 | | | | 0.0084 |
| 159 | | | | |
| 160 | | | | 0.0027 |

From these and the two previous equations the relative values of the fission yields can be obtained, since the $N(t)$'s are proportional to the observed isotopic peak heights. Assuming a value 5.40 for the percentage yield at 143 the values tabulated in Table I and shown in Fig. 1 were obtained. The accuracy of measurement was such that ratios of these values are good to 1 percent.

## III. SAMARIUM

The fission samarium and higher rare earths used in this investigation were obtained from Parker and Lantz of the Oak Ridge National Laboratory. The samples consisted of resin column separated heavy rare earths formed from uranium which had been submitted to a flux of $7.45\times10^{11}$ neutrons/cm² sec. for 920 days in the Clinton pile. Three hundred ten days after removal from the pile the lower rare earths (through Pm) were removed from the sample by a resin column extraction. Five hundred five days after removal from the pile the sample was analyzed by a filament source mass spectrometer. The results of this mass analysis are given in Table III.

Several factors complicate the analysis of these data. In the first place the hold-up at mass 147, due to the incomplete decay of $Pm^{147}$ at the time of the column separation, is unknown because of the uncertain half-life[1] of this isotope (2.2 yr. to 4 yr.). Further, although the cross section of $Sm^{149}$ is well known as a function of energy[6] its mean cross section to the neutrons inside a

TABLE II. Fission neodymium, percentage composition.

| Mass | 143 | 144 | 145 | 146 | 148 | 150 |
|---|---|---|---|---|---|---|
| Percentage abundance | 27.9 | 25.6 | 19.3 | 15.0 | 8.72 | 3.51 |

[6] W. J. Sturm, Phys. Rev. **71**, 757 (1947).

uranium slug in a pile has not been measured. Finally, the cross section of the long-lived $Sm^{151}$ is entirely unknown and its half-life has only been crudely measured.[7]

The equations for the numbers of atoms of the various samarium isotopes are,

$$N_{147}(t_3)=\frac{N_u\sigma_u f_{147}\Phi}{\lambda_{147}^*}[\lambda_{147}^* t_1-\exp[-\lambda_{147}^*(t_2-t_1)]+\exp[-\lambda_{147}^* t_2]], \quad (4)$$

$$N_{149}(t_3)=\frac{N_u\sigma_u f_{149}\Phi}{\sigma_{149}\Phi}[1-\exp[-\sigma_{149}\Phi t_1]], \quad (5)$$

$$N_{150}(t_3)=\frac{N_u\sigma_u f_{149}\Phi}{\sigma_{149}\Phi}[\sigma_{149}\Phi t_1-1+\exp[-\sigma_{149}\Phi t_1]], \quad (6)$$

$$N_{151}(t_3)=\frac{N_u\sigma_u f_{151}\Phi}{\lambda_{151}+\sigma_{151}\Phi}[1-\exp[-(\lambda_{151}+\sigma_{151}\Phi)t_1]]\times\exp[-\lambda_{151}(t_3-t_1)], \quad (7)$$

$$N_{152}(t_3)=N_\mu\sigma_\mu f_{152}t_1+\frac{N_u\sigma_u f_{151}\sigma_{151}\Phi^2}{(\lambda_{151}+\sigma_{151}\Phi)^2}[(\lambda_{151}+\sigma_{151}\Phi)t_1-1+\exp[-(\lambda_{151}+\sigma_{151}\Phi)t_1]], \quad (8)$$

$$N_{154}(t_3)=N_u\sigma_u f_{154}\Phi t_1. \quad (9)$$

In these equations $t_1$ is the time of neutron irradiation, $t_2$ is the time from the beginning of neutron irradiation until column separation, $t_3$ is the time from the beginning of irradiation until isotopic analysis, and $\lambda_{147}^*$ is the decay constant of $Pm^{147}$. Division of (5) by (6) yields an equation in $\sigma_{149}$, independent of the fission yield, of which a numerical solution then gives for the cross section of $Sm^{149}$, $\sigma_{149}=47{,}000$ barns. On the basis of the neodymium yield curve, the values 2.15, 1.10, and 0.445 percent are assumed for the yields at 147, 149, and 151 respectively. By combining (4) and (5), $\lambda_{147}^*$, and hence the half-life of $Pm^{147}$, are obtained. Calculation gives $T_{\frac{1}{2}}^{147}=2.26$ years. The assumption is now made that $\lambda_{151}$ is small relative to $\sigma_{151}\Phi$ and $t_3^{-1}$. This assumption will later be justified. Then adding (7) and (8) we obtain

$$N_{151}(t_3)+N_{152}(t_3)=N_u\sigma_u\Phi t_1(f_{151}+f_{153}). \quad (10)$$

Division of (10) by (5) yields an equation in $f_{152}$. Solving this gives $f_{152}=0.279$. Similarly, combining (9) and (10) gives $f_{154}=0.0908$. Solving (7) divided by (9) for $\sigma_{151}$ gives $\sigma_{151}=7200$ barns. The samarium fission yield data are summarized in Table I and in Fig. 1. The half-life of $Sm^{151}$ cannot be calculated until the europium data are considered.

[7] Inghram, Hayden, and Hess, Phys. Rev. **71**, 643 (1947).

TABLE III. Fission samarium, percentage composition.

| Mass | 147 | 149 | 150 | 151 | 152 | 154 |
|---|---|---|---|---|---|---|
| Percentage abundance | 34.2 | 12.7 | 25.2 | 12.7 | 12.1 | 3.11 |

## IV. EUROPIUM

The europium peaks in the first-mentioned fission product sample were also observed. Unfortunately, the amount of fission $La^{139}$ present in the sample, even after column purification, was so large that the mass 155 $LaO^+$ peak, for which no correction could be made, precluded measurement of the $Eu^+$ peak at 155. Since europium does not emit appreciably as $EuO^+$, no measurement of $Eu^{155}$ could be made. Although the $Eu^{151}$ daughter was overlapped by its $Sm^{151}$ parent at the 151 position, utilization of the time fractionation of the emission of these two elements in the manner previously described[3] gave the $Eu^{151}/Eu^{153}$ ratio. Assumption of the fission yield at 153 then allowed the half-life of $Sm^{151}$ to be calculated. The equation which determines $\lambda_{151}$ is,

$$\frac{N_{151}'(t_3)}{N_{153}'(t_3)}=\frac{f_{151}}{f_{153}}\frac{\lambda_{151}\sigma_{153}'\Phi}{(1-\exp[-\sigma_{153}'\Phi t_1])}\left[\frac{1}{\sigma_{151}'\sigma_{151}\Phi^2}+\frac{\exp[-\sigma_{151}'\Phi t_1]}{\sigma_{151}'(\sigma_{151}'-\sigma_{151})\Phi^2}+\frac{\exp[-\sigma_{151}\Phi t_1]}{\sigma_{151}(\sigma_{151}-\sigma_{151}')\Phi^2}+\frac{(1-\exp[-\sigma_{151}\Phi t_1])(t_3-t_1)}{\sigma_{151}\Phi}\right], \quad (11)$$

where primed quantities refer to isotopes of europium, where unprimed quantities refer to isotopes of samarium, and where, as before, $\lambda_{151}$ has been assumed small relative to $t_3^{-1}$ and to $\sigma_{151}\Phi$. On the basis of the samarium portion of the yield curve we assume $f_{153}=0.170$. Taking the value of $\sigma_{151}=7200$ barns obtained above, and the values $\sigma_{151}'=5200$ barns and $\sigma_{153}'=240$ barns from previous work,[3] Eq. (11) gives $T_{\frac{1}{2}}^{151}=122$ years.
This half-life makes $\lambda_{151}$ small relative to $t_3^{-1}$ and $\sigma_{151}\Phi$. This new value of $T_{\frac{1}{2}}^{151}$ is certainly more reliable than our previous estimate of 20 years, which was based on photographic darkening.

## V. GADOLINIUM

The gadolinium fission products present in the second sample were investigated with the filament source mass spectrometer. The percentage abundances shown in Table IV were obtained.

Owing to the uncertainty in the values of the large cross sections of $Gd^{155}$ and $Gd^{157}$ the fission yields at masses 155, 156, and 157 were assumed. Then the cross sections of $Gd^{155}$ and $Gd^{157}$ and also the fission yields at masses 158 and 160 were computed. The equations used

TABLE IV. Fission gadolinium, percentage abundance.

| Mass | 155 | 156 | 157 | 158 | 160 |
|---|---|---|---|---|---|
| Percentage abundance | 26.2 | 44.8 | 4.4 | 22.0 | 2.5 |

in the evaluation of these quantities are,

$$N_{155}(t_3)=\lambda_{155}'N_u\sigma_u\Phi f_{155}\Big[\frac{1}{\sigma_{155}\Phi(\lambda_{155}'+\sigma_{155}'\Phi)}+\frac{\exp[-\sigma_{155}\Phi t_1]}{\sigma_{155}\Phi(-\lambda_{155}'-\sigma_{155}'\Phi+\sigma_{155}\Phi)}+\frac{\exp[-(\sigma_{155}'\Phi+\lambda_{155}')t_1]}{(\sigma_{155}'\Phi+\lambda_{155}')(\lambda_{155}'+\sigma_{155}'\Phi-\sigma_{155}\Phi)}\Big]+\frac{N_u\sigma_u f_{155}\Phi}{\lambda_{155}'+\sigma_{155}'\Phi}[1-\exp[-\lambda_{155}'(t_3-t_1)]]\times[1-\exp[-(\lambda_{155}'+\sigma_{155}'\Phi)t_1]], \quad (12)$$

$$N_{156}(t_3)=N_u\sigma_u f_{156}\Phi t+\frac{N_u\sigma_u f_{155}\sigma_{155}'\Phi^2}{(\lambda_{155}'+\sigma_{155}'\Phi)^2}[(\lambda_{155}'+\sigma_{155}'\Phi)t_1-1+\exp[-(\lambda_{155}'+\sigma_{155}'\Phi)t_1]]+N_u\sigma_u f_{155}\sigma_{155}\lambda_{155}'\Phi^2\Big[\frac{t_1}{\sigma_{155}\Phi(\lambda_{155}'+\sigma_{155}'\Phi)}+\frac{1-\exp[-\sigma_{155}\Phi t_1]}{(\sigma_{155}\Phi)^2(\sigma_{155}\Phi-\lambda_{155}'-\sigma_{155}'\Phi)}+\frac{1-\exp[-(\sigma_{155}'\Phi+\lambda_{155}')t_1]}{(\sigma_{155}'\Phi+\lambda_{155}')^2(\lambda_{155}'+\sigma_{155}'\Phi-\sigma_{155}\Phi)}\Big], \quad (13)$$

$$N_{157}(t_3)=\frac{N_u\sigma_u f_{157}\Phi}{\sigma_{157}\Phi}[1-\exp[-\sigma_{157}\Phi t_1]], \quad (14)$$

$$N_{158}(t_3)=N_u\sigma_u f_{158}\Phi t_1+N_u\sigma_u f_{157}\Phi\times\Big[\frac{\sigma_{157}\Phi t_1-1+\exp[-\sigma_{157}\Phi t_1]}{\sigma_{157}\Phi}\Big], \quad (15)$$

$$N_{160}(t_3)=N_u\sigma_u f_{160}\Phi t_1, \quad (16)$$

where primed quantities refer to isotopes of europium, and unprimed quantities refer to isotopes of gadolinium. For the calculation it was assumed that $Eu^{155}$ has a half-life of 1.7 years and a cross section of 7900 barns.[3] Values of $f_{155}$, $f_{156}$, and $f_{157}$ of 0.0500, 0.0260, and 0.0150 percent respectively were assumed. Dividing (13) by (12) an equation in $\sigma_{155}$ was obtained. Numerical solution then gave the value of 41,000 barns for the cross section of $Gd^{155}$. Then combining (13) and (14) the cross section of $Gd^{157}$ was obtained as 59,500 barns. The variations of these cross sections from those presently accepted are discussed in the conclusion. Finally, (15) and (16) combined with any of the earlier equations give 0.0084 and 0.0027 percent for the fission yields at masses 158 and 160, respectively. These fission yield data are summarized in Table I and in Fig. 1.

## VI. CONCLUSIONS AND COMMENTS

Certain of the results here reported differ widely from accepted results, and to a considerably greater extent than would be expected from the uncertainties of measurement or from possible slightly incorrect assumptions for various of the fission yields. The first of these discrepancies is the short half-life obtained for $Pm^{147}$. Although published values for this half-life vary from 2.2 years to 4 years, a 3.7 year value has been most generally accepted. Another discrepancy is that the cross section of $Gd^{157}$ obtained is considerably lower than the 200,000 barn value reported in the literature.[6,8] We have attempted to explain this result by the assumption that part of the $Gd^{157}$ peak observed is caused by a contamination of the sample with normal gadolinium. However, even if this contamination is assumed to be so large that all the $Gd^{160}$ peak arises from this cause, the effect on the $Gd^{157}$ is not enough to bring its cross section much over 100,000 barns. Thus this explanation of the low cross section appears untenable. Although contamination at the mass 173 position, where the $Gd^{157}$ was measured, due to some other element cannot be entirely ruled out, the low cross section is probably real and due to the high neutron temperature in the uranium. A question might be raised as to the validity of the method of assuming yields to produce a smooth yield curve. In the opinion of the authors this procedure is justified because of the smoothness of the neodymium partial curve and because some simple connection must exist between the yields if the choice of eight yields on a particular smooth curve forces nine other yields to lie on the curve. In conclusion, it should be explicitly pointed out that our fission yields and cross sections are for the neutron energy distribution found in a thick uranium slug. These results would not be expected to be in exact agreement with those obtained by irradiation of thin uranium foils with moderated pile neutrons.

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[8] Lapp, Van Horn, and Dempster, Phys. Rev. **71**, 745 (1947).