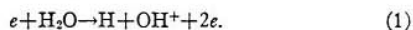


fragments in the ionization process



It has been found by Stevenson<sup>10</sup> that a necessary condition for zero kinetic energy in such a process is that  $I(\text{H}) > I(\text{OH})$ , a condition which is satisfied. On the assumption that the ionization fragments have zero kinetic energy we find  $D(\text{H}-\text{OH}) = 5.03 \pm 0.2 \text{ ev} = 116 \pm 5 \text{ kcal/mole}$  and combining this with thermochemical data<sup>11</sup> obtain  $D(\text{OH}) = 4.47 \pm 0.2 \text{ ev} = 103 \pm 5 \text{ kcal/mole}$ . This derived value for  $D(\text{OH})$  falls between the value of Dwyer and Oldenberg,<sup>12</sup>  $100.1 \pm 0.9 \text{ kcal/mole}$  and the recently determined value of Hornbeck,<sup>13</sup>  $106.71 \text{ kcal/mole}$ .<sup>14</sup> Both of these spectroscopically determined values fall within the limits of error of our measurement. It should be noted that if the ionization fragments in process (1) possessed any kinetic or excitation energy, the correction required in our value of  $D(\text{OH})$  would bring it closer to Hornbeck's value.

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## Radiochemical Measurements of the Natural Fission Rate of Uranium and the Natural Occurrence of the Short-Lived Iodine Isotopes

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AS early as 1939, Libby<sup>1</sup> made an attempt to measure the activity of the short-lived iodine isotopes in uranium salts, but the result was negative, and only a lower limit for the natural neutron emission rate of uranium, equivalent to a half-life of  $10^{14}$  yr, was given. Recent mass spectrometric analyses of the stable fission product gases in uranium minerals,<sup>2-4</sup> failed to give a reliable estimation of the total natural fission rate of uranium, due to the uncertainties concerning possible loss of gaseous products over the lifetime of the minerals. In an attempt to obtain the current total (spontaneous plus neutron-induced) fission rate in uranium salts and minerals, we have repeated the Libby experiment with larger amounts of starting materials.

Preliminary experiments were carried out with 1020 g of uranium extracted from a sample of Belgian Congo pitchblende and carefully purified. The uranium as nitrate was dissolved in 19 liters of water, and then 230 ml of concentrated sulfuric acid and 100 g of sodium nitrite were added to the uranium solution. The iodine isotopes were extracted with carbon tetrachloride (twice in 500 ml portions) together with iodine carrier (100 mg  $\text{I}_2$  in methanol). The carbon tetrachloride solution was washed with water, and shaken with a 100 ml portion of 2% sodium sulfite solution, in a separatory funnel. The sodium sulfite layer was transferred to another separatory funnel, acidified with 20 ml of

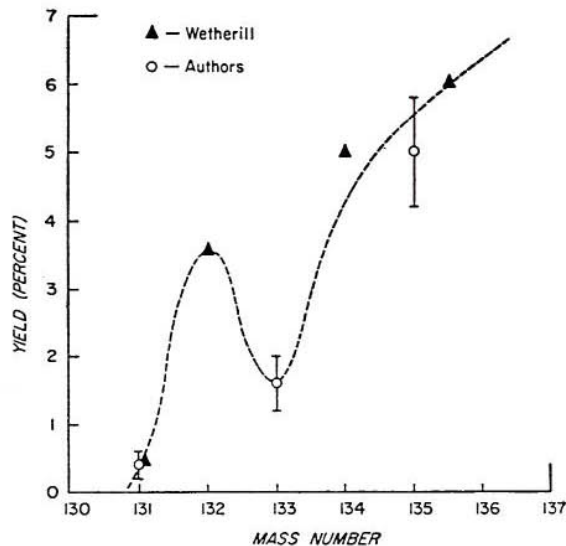


FIG. 1. Spontaneous fission yields of  $\text{U}^{238}$ .

6*N* sulfuric acid, 2 g of sodium nitrite were added to the solution, and again extracted with 200 ml of carbon tetrachloride.

The iodine isotopes were transferred into an aqueous solution of sodium sulfite, 10 mg of iron carrier was added, the solution was boiled, and the iron was precipitated with ammonia. The addition and removal of iron carrier were repeated several times, until the decontamination was complete. The solution was finally acidified with sulfuric acid, and the iodine was precipitated as silver iodide. The silver iodide precipitate was filtered, dried, and mounted on aluminum for measurement with an end-window Geiger tube. Recovery of the iodine carrier was about 75%. The spontaneous fission rate of uranium-238 can be obtained from the equation,

$$I/U(\text{curie/curie}) = T_{1/2}/T_{1/2}^f \cdot y, \quad (1)$$

where  $T_{1/2}$  is the 2-decay half-life,  $T_{1/2}^f$  is the spontaneous fission half-life of uranium-238, and  $y$  is the spontaneous fission yields of the iodine isotopes, provided the slow neutron-induced fission contribution is negligible. Since it is well known that the slope of the spontaneous fission yield curve is different from that of the slow neutron-induced fission yield,<sup>2-4</sup> only an approximate value for the spontaneous fission half-life for  $\text{U}^{238}$  can be obtained by assuming certain values for the spontaneous fission yields of iodine isotopes. Wetherill<sup>4</sup> gives the following yields for xenon isotopes;  $\text{Xe}^{131}$ , 0.455,  $\text{Xe}^{132}$ , 3.57,  $\text{Xe}^{134}$ , 4.99 relative to an assumed value (6.00%) for  $\text{Xe}^{136}$ . If an average fission yield for  $\text{I}^{131}$ ,  $\text{I}^{133}$ , and  $\text{I}^{135}$  is taken to be  $3.0 \pm 0.3\%$ , the spontaneous fission half-life of  $\text{U}^{238}$  may be calculated from Eq. (1) yielding  $(1.03 \pm 0.10) \times 10^{16}$  yr, which is higher than the value of Segrè,<sup>5</sup>  $(8.04 \pm 0.3) \times 10^{15}$  yr, but lower than the value of Perfilov,<sup>6</sup>  $(1.3 \pm 0.2) \times 10^{16}$  years.

The spontaneous fission yields for the chains of 131, 133, and 135 can be calculated, if Segrè's value is taken to be correct, with the further assumption that interchange between the fission iodine and the added iodine carrier was complete in the extraction procedure used. The results of these calculations are shown in Fig. 1 together with the mass-spectroscopic data obtained by Wetherill<sup>4</sup> for xenon isotopes.

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