

radial parameters are involved in spin-other-orbit interaction and spin-spin interaction, they will probably have effects of the same order of magnitude on the interval. It should then be possible to get more consistent values of  $\zeta_d$  when  $C_1$  is corrected for spin-other-

orbit interaction, and this would be a check on the preceding analysis.

I wish to thank Dr. C. W. Ufford for suggesting this problem and for many helpful discussions during the course of the work.

## Half-Life of $I^{129}$ and the Age of the Elements\*

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The specific activities of several samples of methyl iodide containing  $I^{129}$  were measured with proportional counters, and the ratios of  $I^{129}$  to  $I^{127}$  were determined with a  $60^\circ$  sector type mass spectrometer. The half-life of  $I^{129}$  was found to be  $(1.72 \pm 0.09) \times 10^7$  years. The time interval between the formation of the elements and the formation of the earth calculated from this value is  $2.7 \times 10^8$  years. It was assumed that most of the  $Xe^{129}$  present on the earth at present originated from decay of  $I^{129}$  after the formation of the earth and that the original cosmic abundance of  $I^{129}$  was about equal to that of the stable  $I^{127}$ .

### I. HALF-LIFE DETERMINATION

THE half-life of  $I^{129}$  was found by measuring the absolute disintegration rates, isotopic compositions, and total iodine contents of several samples obtained from fission product iodine that was separated from a uranium slug which had received a 4-year irradiation in the Oak Ridge pile and had cooled for 21 months.

For the chemical separation the uranium was dissolved in concentrated hydrochloric acid to which a few mg of iodine carrier had been added. After oxidation with hydrogen peroxide the elementary iodine was removed by counter-current extraction into carbon tetrachloride. It was purified by six or eight (see below) chemical cycles consisting of reduction to iodide, extraction into water, re-oxidation to iodine, and re-extraction into carbon tetrachloride. The iodine was finally precipitated as  $PdI_2$ , which was washed and dried. Then it was decomposed by heating to  $350^\circ$  in an evacuated glass system; the evolved iodine vapor was collected in a tube containing methanol and red phosphorus. This mixture was heated at  $110^\circ$  for one hour to make methyl iodide, which was then passed over anhydrous calcium chloride to separate it from excess methanol. The methyl iodide was used for the activity measurements and the isotope ratio determinations.

The counters<sup>1</sup> were 2 cm in diameter and 30 cm long with silver cathodes and 2-mil wolfram center wires. They were operated in the proportional region with a 5000-volt power supply, a non-overloading pulse amplifier, and an Atomic Instrument Company scaler, model 101A. The counter gas was methane at one atmosphere. Preliminary experiments were performed

to study the effect of adding small amounts of methyl iodide to the methane. It was found that the counter plateau disappeared and that the counting rate of a  $Co^{60}$   $\gamma$ -ray standard started to decrease as the partial pressure of methyl iodide was increased above 50 microns. Therefore, the measurements were made with the radioactive methyl iodide samples at partial pressures between 17 and 38 microns and with the counters operating at 3800 volts on both voltage and gain plateaus. Each sample was counted at least twice for periods of 20 to 150 minutes for total recorded counts  $> 10^4$ . Then the counter was cooled to  $-195^\circ$  for one hour and the methane pumped off leaving the methyl iodide behind. This was then distilled, by warming the counter to room temperature, into a small tube containing a 30-mg piece of sodium and cooled with liquid nitrogen. This tube was set aside for the quantitative iodine determination. The counter was refilled with methane and the background counting rate determined. This was usually found to be somewhat higher than what it had been before the active methyl iodide had been in the counter. Apparently, anywhere, from 0 to 12 percent (average of 5 percent) of the methyl iodide remained in the counters. The backgrounds used in the calculations were those measured after the methyl iodide was distilled out. The operation of the counters was always checked by means of the  $Co^{60}$  standard. Its counting rate, corrected for  $I^{129}$  activity and background, remained the same within one percent, independent of the counter used and the amount of active methyl iodide therein. The total volumes and cathode volumes of the counters were measured at the conclusion of the experiments by adding water to them from a buret. It was shown by Bernstein and Ballentine<sup>1</sup> that the cathode volume is equal to the sensitive volume and that the observed counting rate is very nearly equal

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<sup>1</sup> W. Bernstein and R. Ballentine, *Rev. Sci. Instr.* **21**, 158 (1950).

TABLE I. Data for determination of half-life.

No. of sample	No. of counter	Cathode vol.	Moles of CH <sub>3</sub> I in counter	Counter bkd. (c/m)	Activity of I <sup>129</sup> (c/m) (corr. for bkd.)	Half-life (years)
		Total vol.				
1a	7	0.859	$1.698 \times 10^{-7}$	127	467	$1.62 \times 10^7$
1b	8	0.867	$1.468 \times 10^{-7}$	117	376	$1.75 \times 10^7$
1c	7	0.859	$1.838 \times 10^{-7}$	183	470	$1.74 \times 10^7$
1d	8	0.867	$1.995 \times 10^{-7}$	137	506	$1.77 \times 10^7$
2a	6	0.864	$0.970 \times 10^{-7}$	169	250	$1.71 \times 10^7$
2b	7	0.859	$0.925 \times 10^{-7}$	148	238	$1.71 \times 10^7$
2c	8	0.867	$1.434 \times 10^{-7}$	119	370	$1.72 \times 10^7$
2d	10	0.864	$1.810 \times 10^{-7}$	95	462	$1.73 \times 10^7$
2e	8	0.867	$1.688 \times 10^{-7}$	131	427	$1.75 \times 10^7$

to the actual disintegration rate within this volume for C<sup>14</sup>. In the present work it was assumed that these conditions apply also.

The small tube containing the methyl iodide transferred from the counter was heated with the sodium to 250° for 20 minutes in order to form NaI. Then water vapor diluted with helium was passed in to react with the excess sodium. The mixture in the tube was dissolved in 3 ml of water and its iodine content determined quantitatively by a potentiometric titration with  $1.01 \times 10^{-4}$  molar silver nitrate solution against a calomel electrode. This analytical method was tested first by titrating eight 22- $\mu$ g samples of iodide from a stock solution. The mean deviation from the average was less than 2 percent, and the agreement with a gravimetric analysis of the stock solution in which two 13.5-mg samples of silver iodide were precipitated was better than 4 percent. The analytical method was also tested by filling counters with amounts of inactive methyl iodide measured with a manometer and expanded by a known amount, adding an atmosphere of methane, and then proceeding as in the final experiments. In five samples with methyl iodide pressures between 16 and 142 microns the average agreement was 7 percent, but the methyl iodide pressure determinations were not very accurate and no correction was made for the methyl iodide which remained behind in the counter. In one run when no methyl iodide was added a zero blank was obtained.

Two different isotopic analyses were performed. The first, (No. 1), was done with the CH<sub>3</sub>I which was prepared from the iodine that had been through six purification cycles and from which samples 1a-1d were taken. Isotopic analysis No. 2 and samples 2a-2e were taken from the iodine that had undergone eight purification cycles. The single collector 60° sector type mass spectrometer<sup>2</sup> that was used had 70-volt ionizing electrons, a 4000-volt ion accelerating potential, and magnetic scanning. Several preliminary runs were made with ordinary CH<sub>3</sub>I (only I<sup>127</sup>). For analysis No. 1, the ratios of the ion current at mass 129 to that at mass 127 (I<sup>+</sup> peaks) in six successive runs were: 0.0707,

0.0703, 0.0701, 0.0701, 0.0713, and 0.0702. The average was 0.0704. In analysis No. 2, the ratios in four runs were: 0.0714, 0.0691, 0.0685, 0.0682; average: 0.0694. Since a compensating hairpin-type leak was used, a correction for fractionation had to be made: The ratios were multiplied by (mass of CH<sub>3</sub>I<sup>127</sup>/mass of CH<sub>3</sub>I<sup>129</sup>)<sup>1/2</sup>. Thus, the corrected ratios for analyses No. 1 and No. 2, respectively, are: 0.0699 and 0.0689; or 6.53 and 6.45 atomic percent I<sup>129</sup>. For some of the runs the complete spectrum was computed using these ratios and the pattern for ordinary CH<sub>3</sub>I. The maximum deviation was less than 2 percent for any mass. The isotopic ratios found are probably accurate to  $\pm 2$  percent even considering the possible systematic errors.

Table I gives the data and results for each sample. The average of the nine values for the half-life of I<sup>129</sup> is  $1.72 \times 10^7$  years with an over-all estimated probable error of 5 percent. A half-life of  $(3 \pm 1) \times 10^7$  years based on experiments in which the activity of solid samples of AgI containing I<sup>129</sup> was measured has been reported by Parker, *et al.*<sup>3</sup> They also report a maximum  $\beta$ -ray energy of 100-135 kev in good agreement with the value of 130 kev found in this work by observing that the range of the  $\beta$ -rays in aluminum is about 22 mg/cm<sup>2</sup>.

## II. AGE OF THE ELEMENTS

It was first pointed out by Suess<sup>4</sup> that a lower limit for the time interval,  $t$ , between the formation of the elements and the formation of the earth's atmosphere could be calculated if the half-life of I<sup>129</sup> were known. Any I<sup>129</sup> still in existence when the earth was first able to retain its atmosphere must have decayed to Xe<sup>129</sup> and therefore increased the abundance of this isotope over its primeval value. By making the plausible assumption that the original abundance of I<sup>129</sup> was approximately equal<sup>4-6</sup> to that of I<sup>127</sup>, it is only necessary to compare the present abundance in the earth's crust of I<sup>127</sup> with that of Xe<sup>129</sup>. Suess used values of 1 g/cm<sup>2</sup> and  $10^{-4}$  g/cm<sup>2</sup> for these abundances, respec-

<sup>3</sup> Parker, Creek, Herbert, Lantz, and Martin, Oak Ridge National Laboratory Report No. 286, p. 53, 1949 (unpublished).

<sup>4</sup> H. E. Suess, *Z. Physik* 125, 386 (1948).

<sup>5</sup> H. E. Suess, *Z. Naturforsch.* 2a, 311 (1947).

<sup>6</sup> J. Mattauch and S. Flüge, *Nuclear Science Tables* (Interscience Publishers, Inc., New York), p. 104.

<sup>2</sup> O. A. Schaeffer and J. M. Hastings, *J. Chem. Phys.* 18, 1048 (1950).

tively, and calculated a lower limit for  $t$  of 10  $I^{129}$  half-lives. From the systematics of nuclear abundance data, he thought that  $Xe^{129}$  did not have an abnormally high abundance compared to the other Xe isotopes.

Aten,<sup>7</sup> however, has pointed out that in all cases, where an odd isotope occurs between two even ones, the odd one is always less abundant than the sum of the abundances of the other two. The only exception is  $Xe^{129}$  whose abundance is 26.23 percent while  $Xe^{128}$  and  $Xe^{130}$  have abundances of 1.90 percent and 4.07 percent, respectively. Therefore, most of the  $Xe^{129}$  present in the atmosphere today originated by decay of  $I^{129}$  after the formation of the earth just as most of the  $A^{40}$  originated from decay of  $K^{40}$ .

It was suggested by Dr. M. Goldhaber of this Laboratory that  $t$  probably could be calculated more accurately by comparing the  $I^{127}/K^{40}$  abundance ratio with the  $Xe^{129}/A^{40}$  abundance ratio. In this way the terrestrial abundance data given by Goldschmidt<sup>8</sup> can be applied directly; and as a first approximation it can be assumed that the atmosphere contains the same fraction of the earth's  $Xe^{129}$  that it does of the earth's  $A^{40}$ . The following equation was used:

$$(I^{127}e^{-\lambda t}/K^{40})_T = (Xe^{129}/A^{40}).$$

$I^{127}$  refers to an abundance<sup>8</sup> of 0.24 atom per  $10^6$  atoms

<sup>7</sup> A. H. W. Aten, Jr., Phys. Rev. **73**, 1206 (1948).

<sup>8</sup> V. M. Goldschmidt, Geochem. Verteilungsgesetze IX, Videnskapsakademien, Oslo (1937).

of silicon;  $\lambda$  is the disintegration constant of  $I^{129}$ ;  $K^{40}$  refers to the number of atoms (per  $10^6$  atoms of Si) which have decayed to  $A^{40}$  in time  $T$ , the age of the earth,  $3.35 \times 10^9$  years.<sup>9</sup> Its present abundance was taken as 0.0119 percent of the total potassium abundance of 44,200 atoms<sup>8</sup> per  $10^6$  Si atoms; its half-life  $-1.27 \times 10^9$  years<sup>10</sup> with 12 percent decaying to  $A^{40}$ . The  $A^{40}$  in the equation refers to its abundance in the atmosphere by volume, 0.93 percent; and  $Xe^{129}$  refers to the radiogenic  $Xe^{129}$  which is assumed to be 20 percent of the total Xe abundance in the atmosphere,  $8 \times 10^{-6}$  percent Xe by volume.<sup>11</sup> This leaves 6.23 percent  $Xe^{129}$  which is not radiogenic. Thus, the rule of Aten is approximately satisfied, and the rule which states that two odd isotopes of an element have roughly the same abundance<sup>5,6</sup> is not seriously violated either (abundance of  $Xe^{131}$  is 21.2 percent).

Solving the equation for  $t$  gives 15.4 half-lives of  $I^{129}$  or  $2.7 \times 10^8$  years for the time between the formation of the elements and the formation of the earth. Thus, the age of the elements ( $T+t$ ) is  $3.6 \times 10^9$  years. This calculation of  $t$  is very insensitive to the accuracy of the assumptions and approximations that were used. For example, if the amount taken for radiogenic  $Xe^{129}$  is wrong by a factor of 10 the error in  $t$  is only 22 percent.

<sup>9</sup> A. Holmes, Nature **159**, 127 (1947).

<sup>10</sup> G. A. Sawyer and M. L. Wiedenbeck, Phys. Rev. **79**, 480 (1950).

<sup>11</sup> G. Damköhler, Z. Elektrochem. **41**, 74 (1935).

## Conservation of Angular Momentum in the Statistical Theory of Nuclear Reactions

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The conservation of total angular momentum  $J$ ,  $z$ -component of angular momentum  $m$ , and parity  $p$  are considered in connection with the statistical theory of nuclear reactions. Results are obtained for the angular distribution of a single group of outgoing particles and for the cross section as a function of the spin of the residual nucleus. The energy and angular distributions are also considered for the case in which many groups are observed simultaneously. The density of nuclear states as a function of spin affects results appreciably.

### INTRODUCTION

WHEN a particle of energy less than 50 Mev strikes a nucleus it is usually pictured as forming a compound nucleus.<sup>1</sup> In this process the incident energy is distributed among the many particles of the nucleus with the result that the incident particle can no longer be distinguished from the others. The outgoing product particles of the collision are the products of the decay of the compound nucleus. The state of the compound nucleus and, consequently, its decay products are often said to be independent of the way in which the compound nucleus is produced. This statement must be

<sup>1</sup> N. Bohr, Nature **137**, 344 (1936).

qualified, however, by the condition that any compound nuclear state must have the same total angular momentum  $J$ ,  $z$ -component of total angular momentum  $m$ , and parity  $p$  as the initial state.<sup>2</sup> The conservation of  $J$  and  $p$ , as has been noted a number of times,<sup>3</sup> results in selection rules which forbid transitions between certain compound nuclear states and particular initial or final states. Little mention, however, has been made of the conservation of  $m$ , which may be said to have as its

<sup>2</sup> It is assumed here and throughout that the initial state is resolved into a sum of products of spin and orbital functions which in turn can be expressed as a sum of states with particular values of  $J$ ,  $m$ , and  $p$ .

<sup>3</sup> For example, H. A. Bethe, Revs. Modern Phys. **9**, 108 (1937).