Occurrence of the \( ^{\text{ln}+1} \) Series in Nature

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A small quantity of \( ^{237}\mathrm{Np} \), the long-lived ancestor of the \( ^{\text{ln}+1} \) series, has been isolated from a natural source. The \( ^{232}\mathrm{Th} \) content of \( ^{230}\mathrm{Th} \) obtained from Belgian Congo pitchblende concentrate and of \( ^{232}\mathrm{Th} \) obtained from Brazilian monazite has been determined indirectly through isolation of the \( ^{235}\mathrm{Ac} \) daughter. The mass ratio of \( ^{237}\mathrm{Np} \) to \( ^{238}\mathrm{U} \) in the Belgian Congo pitchblende concentrate has been determined as \( (1.8 \pm 0.4) \times 10^{-12} \). The mass ratio of \( ^{233}\mathrm{U} \) to \( ^{235}\mathrm{U} \) in the same ore deposit is found to be \( (1.3 \pm 0.2) \times 10^{-13} \). The ratio of the respective neutron-capture cross sections of \( ^{238}\mathrm{U} \) in the ore body for the \((n,2n)\) and \((n,\gamma)\) reactions is calculated to be \((1.3 \pm 0.6) \times 10^{-3}\) on the basis of the relative \( ^{237}\mathrm{Np} \) and \( ^{239}\mathrm{Pu} \) contents.

Introduction.

Three radioactive series whose respective long-lived parent nuclides are \( ^{232}\mathrm{Th} \), \( ^{238}\mathrm{U} \) and \( ^{235}\mathrm{U} \) have long been known to exist in nature. In reference to the general formula for the mass numbers of the component nuclides these series are referred to respectively as the \( ^{\text{ln}} \), the \( ^{\text{ln}+2} \) and the \( ^{\text{ln}+3} \) series. The "missing" \( ^{\text{ln}+1} \) series was reported by Hagemann et al.\(^1\). These authors prepared \( ^{233}\mathrm{U} \), whose mass number conforms to the general formula \( ^{\text{ln}+1} \), by the \((\mathrm{n},\gamma)\) reaction. They determined the path of the decay chain between artificially produced \( ^{233}\mathrm{U} \) and stable \( ^{209}\mathrm{Bi} \) and reported the properties of the nuclides which are members of the chain. The results are in agreement with those obtained by English et al.\(^2\). Since \( ^{237}\mathrm{Np} \) decays by alpha-particle emission

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\(^1\) French Hagemann, L. I. Katzen, W. H. Studier, G. T. Seaborg and A. Ghiorso, Phys. Rev., 79, 435 (1950). The decay scheme of the \( ^{\text{ln}+1} \) series is given in this paper. The values for the half-lives, as well as the characteristic alpha energies, of the members were also taken from this reference.

to form beta-active \( \text{Pu}^{233} \) decaying in turn to \( \text{U}^{233} \), \( \text{Np}^{237} \) is considered to represent the long-lived genitor of the \( \text{Un} + 1 \) series, and the complete family of nuclides is referred to as the neptunium series.

Except for the stable terminal member of the chain, \( \text{Bi}^{209} \), this family of radioactive heavy nuclides corresponding to the formula \( \text{Un} + 1 \) has not been found in nature\(^{(1)} \). It has been pointed out by Seaborg\(^{(3)} \) that


uranium-bearing ores should contain trace quantities of \( \text{Np}^{237} \) formed by the \( \text{U}^{238}(n,2n)\text{U}^{237} \rightarrow \text{Np}^{237} \) reaction; and it has been suggested by Garner, Bonner and Seaborg\(^{(4)} \) that uranium-bearing ores containing an appreciable relative quantity of thorium should contain some long-lived \( \text{U}^{233} \) formed as the result of the \( \text{Th}^{232}(n,\gamma)\text{Th}^{233} \rightarrow \text{U}^{233} \) reaction. However, isolation of either \( \text{Np}^{237} \) or \( \text{U}^{233} \) from natural sources has not been reported.

In the work presently reported, \( \text{Np}^{237} \) was isolated from a Belgian Congo pitchblende concentrate and the ratios of \( \text{Np}^{237} : \text{U}^{238} \) and \( \text{U}^{233} : \text{U}^{238} \) in this ore source determined.

Although in preliminary experiments \( \text{Np}^{237} \) was isolated from an aqueous waste resulting from the processing of Belgian Congo pitchblende concentrate, it was evident that establishing the \( \text{Np}^{237} : \text{U}^{238} \) ratio by a direct technique would involve the complete dissolution of approximately one hundred kilograms of ore concentrate. Consequently, an approach based on another member of the \( \text{Un} + 1 \) series was used.
The decay of \(^{237}\text{Np}\) leads to the formation of \(^{229}\text{Th}\) (alpha-active, \(7.38 \times 10^3\) years\(^{(1)}\)), and the decay of \(^{238}\text{U}\) leads to the formation of \(^{230}\text{Th}\) (alpha-active, \(8.0 \times 10^4\) years\(^{(5)}\)). Since the half-lives of these thorium isotopes are very long with respect to the time involved in processing an ore but very short with respect to the age of the ore deposit, it follows that the ratio of \(^{229}\text{Th}\) to \(^{230}\text{Th}\), by activity, should be identical to the ratio of \(^{237}\text{Np}\) to \(^{238}\text{U}\), by activity, assuming that \(^{229}\text{Th}\) has been formed by no means other than \(^{237}\text{Np}\) decay. However, it is to be noted that \(^{229}\text{Th}\) may be formed if neutrons are captured by the \(^{232}\text{Th}\) present in small quantities\(^{(1)}\). This correction will be shown to be negligible.

Therefore, a method was devised for measuring the ratio of \(^{229}\text{Th}\) to \(^{230}\text{Th}\) in a sample containing several grams of \(^{230}\text{Th}\) (ionium) which had been isolated, in this laboratory, from an aqueous waste resulting from the processing of Belgian Congo pitchblende concentrate. The first daughter of \(^{230}\text{Th}\) is \(^{226}\text{Ra}\) (alpha-active, 1590 years\(^{(5)}\)). The first daughter of \(^{229}\text{Th}\) is \(^{225}\text{Ra}\) (beta-active, 11.8 days\(^{(1)}\)) and the second is \(^{225}\text{Ac}\) (alpha-active, 10.0 days\(^{(1)}\)). Consequently, it was decided to determine the \(^{229}\text{Th}\) content of a \(^{230}\text{Th}\) sample by determining the \(^{225}\text{Ac}\) content. A procedure was therefore devised whereby \(^{225}\text{Ac}\) could be isolated and then identified by means of the characteristic alpha energies of it and its daughters\(^{(1)}\).

The requirements of the technique are stringent, since a virtually weightless sample of \(^{225}\text{Ac}\) is required for determination of alpha energies; and actinium, a trivalent element, must be separated from divalent radium.
and lead, trivalent bismuth, and tetravalent thorium. In order to obviate
the much more difficult problem of separating actinium from rare earths, the
Th²³⁰ was exhaustively purified from rare earths (and Ac²²⁵) and the Ac²²⁵
allowed to grow again.

By means of a solvent extraction technique, Ac²²⁵ of high purity was
isolated and an aliquot of the final product evaporated on a platinum
disc for alpha pulse analysis(6).

6. The energies of the alpha particles were determined by means of a multi-
channel differential pulse analyzer described by A. Ghiorso, A. H. Jaffey,
H. P. Robinson and B. B. Reissbourd, "National Nuclear Energy Series",

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For determination of chemical yield, a known quantity of Ac²²⁷ was then
added to the Th²³⁰ and the sample allowed to age. An actinium fraction was
isolated and the quantities of Ac²²⁵ and Ac²²⁷ present in the fraction
determined by alpha pulse analysis. From these data, the chemical yield for
the actinium isolation step and thereby the Ac²²⁵ content of the Th²³⁰ sample
may be calculated.

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Experimental.

Isolation of Np²³⁷ from a sample of Th²³⁰. — In the isolation of
approximately one microgram of naturally-occurring Pu²³⁹ from a uranium
process waste(7), the aqueous effluents from the first thienyltrifluoroacetone
extraction cycle were combined and set aside for further study. This composite
aqueous phase was subsequently evaporated to dryness. The residue was dis-
solved in 0.5 M HNO₃ and the resulting solution made 0.1 M in K₂Cr₂O₇. After
a heating period of several hours the solution was saturated with magnesium
nitrate and contacted with eight equal volume portions of diethyl ether (0.3 M in HNO₃) which were scrubbed successively with twelve half-volume portions of a solution 3.5 M in Mg(NO₃)₂ and 0.2 M in HNO₃. The combined ether extract was allowed to evaporate over water. This aqueous phase was evaporated to dryness.

The resulting residue was dissolved in 5 M HCl and any plutonium present reduced to the trivalent state (leaving any neptunium present in the tetravalent state). The neptunium was then extracted into four successive portions of 0.1 M thioyltrifluoroacetone in benzene from the diluted solution(8). These organic phases were then scrubbed successively with small portions of 1.0 M HCl in order to decrease the content of Th²³⁰ and trivalent Pu²³⁹, until the total alpha activity of the composite organic phase (determined by measuring the alpha disintegration rate of an aliquot evaporated on a five ml platinum disc) was reduced to 110 counts per minute.

In order to simplify the interpretation of later alpha pulse data the absence of Th²³⁰ was proved. The proof was furnished by the removal of added Th²³².

Enough Th²³² tracer was added to the organic phase to produce a total beta counting rate, under the counting conditions used, of 12,700 counts per minute. Following several hours of agitation to permit thorium isotope exchange, the organic phase was scrubbed successively with six small portions of 1.0 M HCl in order to strip the thorium from the organic phase. The entire organic phase was then evaporated on a five ml platinum disc. The total alpha counting rate was 26 counts per minute and the total beta counting rate was less than 50 counts per minute.
Alpha pulse analysis showed 7 alpha counts per minute due to Hp²³⁷.

Most of the remaining activity was due to Pu²³⁹.

Isolation of Ac²²⁵ from a Sample of Th²³⁰, As tracer added.— A sample of thorium as the nitrate, containing 8.77 ± 0.09 g. of Th²³⁰ (and 7.4 g. of Th²³²), isolated from an aqueous waste resulting from the processing of Belgian Congo pitchblende concentrate for uranium, was extracted into approximately 10 liters of 0.6 M solution of thenoyltrifluoroacetone (9).

Isolation of Th²²⁵ from a Th²³⁰ Sample, Tracer Added. A composite aqueous phase, containing radium, bismuth and lead daughters of Th²³², of Th²³⁰, and of Th²²⁹ and relatively large quantities...
of Th\textsuperscript{230} and Th\textsuperscript{232} in addition to the desired Ac\textsuperscript{225}, was evaporated to dryness. The residue was dissolved in a small quantity of water and the solution converted to 10 ml of saturated ammonium nitrate, 0.3 M in HNO\textsubscript{3}.

This aqueous feed was then contacted successively with four 10 ml portions of dibutoxytetraethylenglycol\textsuperscript{(11)}, to extract thorium followed

11. Dibutoxytetraethylenglycol was obtained from Carbide and Carbon Chemical Corporation.

by two 10 ml portions of tributyl phosphate\textsuperscript{(12)}, to extract actinium.

12. Tributyl phosphate, the n-butyl orthoester, was obtained from Commercial Solvents Corporation.

leaving radium in the aqueous phase. (Each of these organic phases had previously been pre-equilibrated with a large volume of saturated ammonium nitrate, 0.3 M in HNO\textsubscript{3}). The aqueous feed was then followed, in the extraction cycle, by one 10 ml portion of ammonium nitrate scrub (saturated ammonium nitrate, 0.3 M HNO\textsubscript{3}).

The dibutoxytetraethylenglycol phases were then discarded. The two tributyl phosphate phases, containing actinium and traces of radium, were contacted successively with two five ml portions of ammonium nitrate scrub, in order to reduce the radium content, then contacted successively with three 10 ml portions of 4 M HNO\textsubscript{3} in order to transfer the actinium to an aqueous phase. The aqueous re-extract was scrubbed with carbon tetrachloride to remove traces of tributyl phosphate\textsuperscript{(13)} and then evaporated to dryness


with a heat lamp, taking care not to heat beyond dryness.
The residue was taken up in two milliliters of saturated ammonium nitrate and the above cycle of dibutoxytransethylenglycol and tributyl phosphate extractions repeated on a scale one-fifth of the foregoing.

The final water re-extract was evaporated to dryness and the residue allowed to age for 24 hours to permit Ac$^{228}$ and residual Pb and Bi isotopes, not of the Ac$^{225}$ series, to die out to a low value.

Another extraction cycle on the same scale was then performed. The final aqueous re-extract was then evaporated to dryness and the container heated with one milliliter of 0.02 M HNO$_3$, which was then evaporated to about 0.2 ml. This solution was then saturated with ammonium nitrate and contacted successively in a single centrifuge cone with two 0.10 ml portions of tributyl phosphate. The tributyl phosphate phases were combined and an aliquot evaporated on a five milliliter disc for alpha counting.$^{(11b)}$

14. Alpha counting was done with a methane proportional counter with an approximate 52% geometry factor. Beta counting was done on the first shelf of a Geiger-Muller counter, filled with neon-amyl acetate gas, with a window thickness of approximately 2 mg/cm$^2$, using appropriate aluminum absorbers to prevent counting of alpha particles.

and for measurement of alpha energies.$^{(5)}$ The data are listed in Table 1. The alpha pulse analysis curves are shown in Figure 1. The two curves pertain to the identical aliquot counted at different bias settings and for different counting times.

Isolation of Ac$^{225}$ from a Sample of Th$^{230}$, Tracer Added. -- Using a shortened version of the preceding procedure a sample of Ac$^{227}$ was separated from its daughter activities. This isotope of actinium has two modes of decay, approximately 1.2% of the decaying atoms emitting alpha particles and the remainder emitting beta particles.$^{(15)}$ The energy of the beta

particles is too low to permit the use of beta counting as an assay technique. However, standardisation of an $\text{Ac}^{227}$ solution by determination of the alpha disintegration rate associated with a certain aliquot is easily accomplished if the alpha particles emitted by the $\text{Ac}^{227}$ may be distinguished from the alpha particles emitted by the daughters of $\text{Ac}^{227}$. Consequently, a solution of this isotope was standardised, immediately after purification, by determining, through alpha pulse analysis, the number of alpha disintegrations per minute resulting from the alpha-branching of the $\text{Ac}^{227}$ contained in an aliquot evaporated on a platinum disc. (The energy of the particles considered is sufficiently different from that of any of the $\text{Ac}^{227}$ daughters that no difficulty is experienced if the sample is a freshly purified one.)

An aliquot of the standard $\text{Ac}^{227}$ solution corresponding to 420 c/m due solely to alpha branching was added to a portion of the benzene-
benzyll trifluoroacetone extract, described in the preceding section, containing $5.31 \pm 0.05$ g. of $\text{Th}^{230}$ (and $4.5$ g. of $\text{Th}^{232}$). (This portion of the extract had been mixed with 300 ml. of 0.1 M HNO$_3$ immediately following the isolation of an actinium fraction as described in the preceding section.) The mixture was agitated for approximately 30 min. each day to prevent the accumulation of $\text{Ac}^{225}$, as formed, on the walls of the container. The extract was allowed to age for 56 days.

An actinium fraction was then isolated by the method described in
the preceding section. The final tributyl phosphate extract was evaporated,
in approximately equal parts, on two five ml platinum discs. One of these samples was immediately subjected to alpha pulse analysis (to determine that fraction of the total alpha activity due to $\text{Ac}^{227}$ and that due to $\text{Ac}^{225}$ and its daughters) and the other was concurrently subjected to alpha counting in a counter calibrated at approximately 52% geometry(14). The data are included in Table 1. The alpha pulse analysis curve is shown in Figure 2. The bias setting and counting time differ from those used in obtaining the data of Figure 1.
Isolation of an Actinium Fraction from a Th$^{232}$ Sample, No Tracer Added.

A sample of thorium as the nitrate tetrahydrate (16), containing approxi- mately one kilogram of Th$^{232}$, was dissolved in approximately 14 liters of dibutoxytetraethyleneglycol by long stirring at room temperature. The clarified solution was evaporated with six 500 ml. portions of 6 M ammonium nitrate. The extract, containing 985 + 10 g. of Th$^{232}$, was then allowed to age for 56 days.

At the conclusion of the aging period, the extract was contacted with four 250 ml. portions of ammonium nitrate scrub (saturated ammonium nitrate, 0.3 M in HNO$_3$) in order to strip the actinium, leaving the thorium in the organic phase. These aqueous phases were then separately and successively contacted with two 250 ml. portions of tributyl phosphate, in order to separate actinium from radium, etc.

The dibutoxytetraethyleneglycol phases were combined with the large dibutoxytetraethyleneglycol extract. Tracer Ac$^{227}$ was added and the extract allowed to age for the experiment described in the section, Isolation of an Actinium Fraction from a Th$^{232}$ Sample, Tracer Added.

The tributyl phosphate phases were then contacted successively with three 125 ml. portions of 4 M HNO$_3$. The combined aqueous re-extract, containing actinium and contaminants, was Borsoled with carbon tetrachloride to remove traces of tributyl phosphate and then evaporated to dryness with a heat lamp, taking care not to heat beyond dryness.

The residue was dissolved in a small quantity of water and the solution converted to 30 ml. of saturated ammonium nitrate, 0.3 M in HNO$_3$. The technique used for the isolation of an actinium fraction from a Th$^{230}$ sample was then used, beginning at the first dibutoxytetraethyleneglycol and tributyl phosphate cycle. The data are listed in Table 2.
Isolation of an Actinium Fraction from a Th\(^{232}\) Sample, Tracer Added.

An aliquot of the standard Ac\(^{227}\) solution corresponding to 430 c/m due solely to alpha branching was added to the dibutoxytetraethyleneglycol extract, containing 962 ± 10 g. of Th\(^{232}\), described in the immediately preceding section. Following a 73 day ageing period, an actinium fraction was isolated according to the procedure of the preceding section, beginning with the saturated ammonium nitrate scrub. The data are included in Table 2.

Results and Discussion.

From the data of Table 1 and the value of the half-life of Ac\(^{225}\), 10.0 days\(^{(1)}\), the ratio of Ac\(^{225}\) to Th\(^{230}\) by mass at secular equilibrium is calculated to be 9.9 x 10\(^{-16}\). This figure should probably be set at (9.9 ± 0.8) x 10\(^{-16}\). From this ratio, values for the following mass ratios, at secular equilibrium in the ore body, may be derived: Th\(^{229}\) to Th\(^{230}\), (3.1 ± 0.3) x 10\(^{-10}\) and U\(^{233}\) to U\(^{238}\), (1.3 ± 0.2) x 10\(^{-16}\).

Table 1

<table>
<thead>
<tr>
<th>Expt.*</th>
<th>Th(^{230}) (g)</th>
<th>Ageing (days)</th>
<th>Ac(^{225}) isolated (c/m)</th>
<th>Yield (%)</th>
<th>Ac(^{225}) content (c/m)</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>8.77</td>
<td>80</td>
<td>227</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>5.31</td>
<td>56</td>
<td>153</td>
<td>44.5</td>
<td>344**</td>
</tr>
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</table>

* Expt. 1 involved no Ac\(^{227}\) tracer. In Expt. 2, Ac\(^{227}\) tracer was added so that the chemical yield could be determined.

** Since the feed solution of Th\(^{230}\) had aged 56 days and the period between isolation of the Ac\(^{225}\) sample from the Th\(^{230}\) and alpha pulse analysis of the aliquot was 2h hrs., the "Ac\(^{225}\) content" represents 76.4% of the maximum value.
However, in order to calculate the $\text{Np}^{237}$ to $\text{U}^{238}$ ratio the contribution
to the $\text{U}^{233}$ content resulting from the $\text{Th}(n, \gamma \gamma \gamma)$ reaction must be
evaluated. It is thought that this contribution may be calculated with
reasonable accuracy from the experimental value for the ratio of $\text{Th}^{229}$
to $\text{Th}^{232}$ in a $\text{Th}^{232}$ ore.

From the data of Table 2 the ratio of $\text{Ac}^{225}$ to $\text{Th}^{232}$ at secular
equilibrium is calculated to be less than $1.1 \times 10^{-19}$ and may certainly
be assumed to be less than $2 \times 10^{-19}$. From this latter figure the mass
ratio of $\text{Th}^{229}$ to $\text{Th}^{232}$, at secular equilibrium in the ore body, is calculated
to be less than $3 \times 10^{-11}$ corresponding to an upper limit for the $\text{U}^{233}$
to $\text{Th}^{232}$ mass ratio of $7 \times 10^{-13}$.

Table 2

<table>
<thead>
<tr>
<th>Expt.</th>
<th>$\text{Th}^{232}$ (g)</th>
<th>Ageing (days)</th>
<th>$\text{Ac}^{225}$ isolated (c/m)</th>
<th>Yield (%)</th>
<th>$\text{Ac}^{225}$ content (c/m)</th>
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<tr>
<td>1</td>
<td>985</td>
<td>55</td>
<td>$&lt;5$</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>962</td>
<td>73</td>
<td>$&lt;3$</td>
<td>51.5</td>
<td>$&lt;6^a$</td>
</tr>
</tbody>
</table>

* Expt. 1 involved no $\text{Ac}^{227}$ tracer. In Expt. 2, $\text{Ac}^{227}$ tracer was added
so that the chemical yield could be determined.

** Since the feed solution had aged 73 days and the period between iso-
lation of the $\text{Ac}^{225}$ sample from the $\text{Th}^{232}$ and alpha pulse analysis of
the aliquot was 2h hrs., the "$\text{Ac}^{225}$ content" represents 85.1% of the
maximum value.

In order to correct for the contribution from the $\text{Th}^{232}(n, \gamma \gamma \gamma \gamma)$
reaction to the $\text{Th}^{229}$ content of the $\text{Th}^{230}$ sample of Table 1, use was made
of the data of Levine and Seaborg\footnote{C. A. Levine and G. T. Seaborg, J. Am. Chem. Soc., \textbf{73}, 3278 (1951).} who found that the $\text{Pu}^{239}$ to $\text{U}^{238}$

lower relative Pu$^{239}$ content of the latter mineral is a result of neutron capture by rare earths. On this supposition, it appears logical to assume that for the pitchblende and monazite samples the respective rates of conversion of Th$^{232}$ to U$^{233}$ should equal the ratio of the respective rates of conversion of U$^{238}$ to Pu$^{239}$.

Consequently, the maximum contribution to the Th$^{229}$ to Th$^{230}$ ratio in the pitchblende sample should be equal to the triple product: (Th$^{229}$ to Th$^{232}$ ratio in the monazite sample) times (the factor 1.5 mentioned above) times (the Th$^{232}$ to Th$^{230}$ ratio in the pitchblende sample). In the work being reported, the Th$^{230}$ used had been isolated from Belgian Congo pitchblende concentrate, and the Th$^{232}$ used had been isolated from Brazilian monazite. Therefore, it is felt that by the introduction of a factor of ten (to cover sample differences within a deposit of Brazilian monazite and within a deposit of Belgian Congo pitchblende) the contribution of the Th$^{232}$ (n, gamma) reaction to the Th$^{229}$ to Th$^{230}$ ratio in the pitchblende concentrate used in the present work may be calculated by the above method.

Since the mass ratio of Th$^{232}$ to Th$^{230}$ in the sample studied is 8.40 the maximum contribution to the Th$^{229}$ to Th$^{230}$ mass ratio by the Th$^{232}$ (n, gamma) reaction is calculated, by this method, to be 4 x 10$^{-12}$. This value is considerably below the experimental error involved in the determination of the Th$^{229}$ to Th$^{230}$ ratio.

It is concluded, therefore, that the source of the Th$^{229}$ is Np$^{237}$ formed by the $^{238}$Np (n, 2n) reaction. The Np$^{237}$ to $^{236}$U mass ratio(5) is then calculated to be (1.8 ± 0.4) x 10$^{-12}$.

This ratio may be compared with the Pu$^{239}$ to $^{238}$U mass ratio of (1.5 ± 0.2) x 10$^{-11}$ for a sample of pitchblende concentrate from the same deposit.(7) Since the ratio of the respective rates of formation of
$\text{Np}^{237}$ and $\text{Pu}^{239}$ is the same as the ratio of their respective activities as measured in the present ore body, assuming no appreciable variation in the neutron flux over past ages, it follows that the ratio of the respective neutron capture cross sections of $\text{U}^{238}$ in the ore body for the $(n, 2n)$ and $(n, \text{gamma})$ reactions may be calculated to be $(1.3 \pm 0.6) \times 10^{-3}$.

The neutron flux is assumed to be contributed to by neutrons resulting from: spontaneous fission of $\text{U}^{238}$, the $\text{U}_1(a, n)\text{U}_2$ reaction where $\text{U}_1$ is a light element such as Li, Si, Mg etc., induced fission of $\text{U}^{235}$ and cosmic rays. The importance of these various neutron sources in ore bodies has been discussed by Seaborg et al (3,18) Garner et al (14), Levine et al (17).

FIG. 1. ALPHA-SPECTRUM OF NATURALLY OCCURRING AC\textsuperscript{229}.
FIG. 2. ALPHA-SPECTRUM OF NATURALLY OCCURRING AC$^{227}$ WITH AC$^{227}$ TRACER.