Induced Radioactivity in Strontium and Yttrium

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Strontium and yttrium have been bombarded with 6.3 Mev deuterons, with 14–20 Mev neutrons, and with slow neutrons, and induced radioactivity obtained in both elements. The activity in strontium is readily obtained by the action of deuterons or neutrons on strontium and possibly, with very weak intensity, by the action of fast neutrons on yttrium with the emission of a proton. The periods observed in strontium are 3.0 ± 0.1 hours (negative active, maximum energy $=0.61$ Mev), and 55 ± 5 days (negative active, maximum energy $=1.9$ Mev). A gamma-ray is found associated with the 3-hour period. Evidence has been obtained showing that the two periods may be ascribed to isomeric forms of Sr^{89}. Activity in yttrium is obtained when strontium is bombarded with deuterons and when yttrium is bombarded with deuterons or neutrons. The periods observed are 120 ± 4 minutes (positive active, maximum energy $=1.2$ Mev) and 60.5 ± 2.0 hours (negative active, maximum energy $=2.6$ Mev). These periods are due, it is believed, to Y^{88} and Y^{90}, respectively. In the possible formation of Y^{90} from $Sr+H^2$, it is suggested that the process involves the complete capture of a deuteron, a reaction not previously observed.

INTRODUCTION

IN continuing the study of radioactivity induced in the heavier elements by deuteron, neutron, and alpha-particle bombardments, it was thought desirable to separate and identify the active isotopes produced in the bombardment of strontium. The known isotopes of elements in this region of the periodic system are given, together with their relative abundance, in Table I. The radioactive isotopes prepared in these experiments are shown in circles in their probable locations.

Fermi and his co-workers[1] exposed both strontium and yttrium to neutrons with negative results, although the yttrium is reported to have absorbed slow neutrons very intensively with the emission of gamma-rays. Somewhat later, Hevesy and Levi[2] reported a period of 70 hours in yttrium resulting from neutron bombardment. More recently, Pool, Cork, and Thornton,[3] in a general survey of radioactivity induced by very energetic neutrons, reported two periods from the bombardment of strontium and four from yttrium. No chemical separations were made in that survey in the case of yttrium, and only one period was found in the strontium precipitate from active strontium.

In order to clarify the matter, additional evidence has been obtained which now permits the positive identification of several isotopes of the two elements. Certain new active isotopes were found, together with revised values for the half-life periods of those previously reported. The experimental work included the bombardment of both strontium and yttrium with deuterons, high speed neutrons, and slow neutrons, together with the chemical separation of the active products. In addition, some work has been done on the related bombardment of zirconium with deuterons and neutrons, and of rubidium with alpha-particles and deuterons.

APPARATUS AND MATERIALS

The cyclotron was used as a source of high speed particles. Deuterons of 6.3 Mev were obtained and these either were used directly or were allowed to fall on a target of lithium metal

TABLE I. *Relative abundance of isotopes; active isotopes are shown with their half-life periods in circles in their suggested locations.*

At. Wt.	84	85	86	87	88	89	90	91	92	93	94	95	96
37 Rb		72.7		27.3									
38 Sr	0.5		9.8	7.5	82.4	3.0h 55d							
39 Y					2.0h	100	60.5h						
40 Zr							48	11.5	22		17		1.5

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[1] Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti, and Segrè, Proc. Roy. Soc. **149A**, 547 (1935).

[2] Hevesy and Levi, Kgl. Danske Videnskab. Selskab, Math.-fys. Medd. **14**, No. 5 (1936).

[3] Pool, Cork, and Thornton, Phys. Rev. **52**, 239 (1937).

to obtain fast neutrons, or on a target of beryllium in order to obtain neutrons of lower energy. The intensity of the deuteron beam was between 5 and 10 microamperes in most of the experiments here reported, and the length of exposure varied up to a maximum of about six hours. In the experiments involving fast neutrons, the sample was placed in a small cadmium cylinder to shield out slow neutrons, and the whole placed beneath a layer of lithium metal on which the deuterons were allowed to fall. For slow neutrons, the sample was covered with several centimeters of paraffin, and neutrons from a beryllium target were used. In the deuteron bombardment, the sample was covered with very thin aluminum, or with palladium foil in order to avoid the possibility of contaminating the yttrium separation with aluminum activity in case the aluminum foil adhered to the sample.

The activities obtained were measured either with a Lauritsen type electroscope or with a Wulf string electrometer using an ionization chamber filled with Freon (difluorodichloromethane).

The strontium was bombarded in the form of the hydroxide, the chloride, and the nitrate. The nitrate was more satisfactory than the hydroxide since its use avoided the possibility of contamination by CO_2 which is absorbed from the air by the hydroxide. The yttrium was a preparation of $Y(NO_3)_3$ from C. A. F. Kahlbaum which was not further purified. It seems probable that this material contains as impurities small amounts of dysprosium and traces of thorium. The latter was suspected because of an extremely weak natural radioactivity in the yttrium, and the former because the dysprosium period was observed after slow neutron bombardments. Zirconium was bombarded in the form of the nitrate, and rubidium as the chloride.

Chemical Separations

A variety of methods were used in separating the activities in the bombarded strontium. First, yttrium hydroxide was precipitated by ammonium hydroxide from the solution of activated strontium in the presence of excess ammonium chloride and of potassium chloride. This was followed by the addition of ammonium oxalate and the removal of strontium oxalate. Finally, potassium and rubidium were precipitated together as the perchlorates by the addition of $HClO_4$ followed by evaporation and thorough washing with anhydrous ethyl acetate. The procedure is that commonly used in qualitative chemical analysis and was essentially the same as that followed by Walke[4] in his analogous separation of radioactive potassium, calcium, and scandium. From this procedure, it was found that the activity was in the strontium and yttrium precipitates, with no detectable activity (30 minutes after bombardment) in the rubidium residue.

Since the precipitation reactions for yttrium and strontium in this scheme are nonspecific and might well precipitate many other ions from solution, either directly or by adsorption on the gelatinous hydroxide precipitate, other reactions were necessary in order to avoid contamination. It was found that an isolation of yttrium could be obtained by dissolving the strontium salt in dilute acid, boiling to expel any F^{17} or N^{13}, then adding yttrium nitrate, excess ammonium oxalate, and ammonium hydroxide. The strontium was thus precipitated while most of the yttrium remained in solution as a complex oxalate. The yttrium was recovered from the filtrate by the addition of sodium phosphate. For the direct separation of strontium, the method of Willard and Goodspeed[5] was used. Yttrium and strontium were dissolved together in nitric acid and then crystalline strontium nitrate was precipitated by the addition of fuming nitric acid to make the final acid concentration 80 percent.

The same general methods were also used in separating strontium from bombarded yttrium and yttrium from zirconium after the removal of the latter as the phosphate.

Radioactive Isotopes of Strontium

From deuteron bombardment of strontium, two definite negative-active periods were obtained. These were found in every strontium precipitate, and were found to have half-lives of

[4] Walke, Phys. Rev. **51**, 439 (1937).
[5] Willard and Goodspeed, J. Ind. Eng. Chem., Anal. Ed. **8**, 414 (1936).

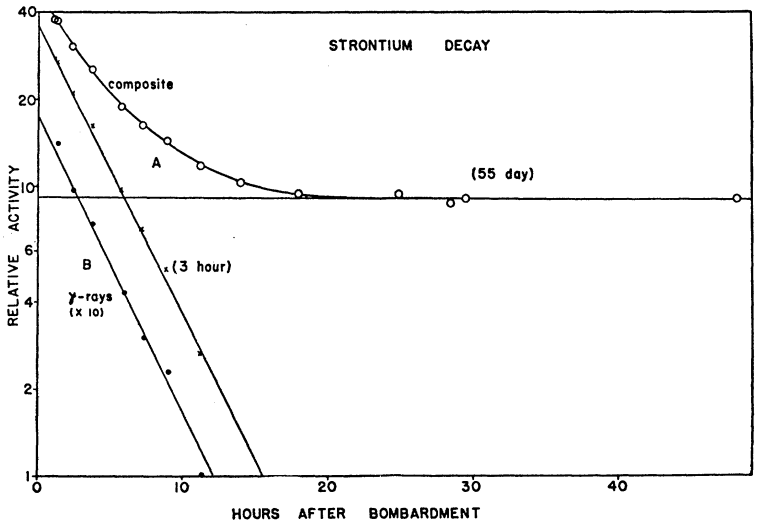

FIG. 1. Decay curves for Sr separated from $Sr+H^2$; curve *A*, total activity; curve *B*, gamma-activity only.

3.0±0.1 hours and 55±5 days.[6] In certain of the strontium precipitates a third, positive-active period of 10.5 minutes was observed with considerable intensity and at first it was believed due to Sr^{85} which could then decay to stable Rb^{85}. That this is not the case, and that the 10.5 minute period is probably extraneous, was shown by more thorough chemical separations in which the period was not observed when strontium chloride or nitrate was bombarded instead of strontium hydroxide. In addition, if the period had been that of Sr^{85}, it should also have been made from Sr^{86} by the ejection of a neutron. Fast neutron bombardment of strontium in any form, however, gave only a weak activity of the longer negative periods. Attempts to produce Sr^{85} by the reaction:

$$Rb^{85}+H^1 \rightarrow Sr^{85}+n^1 \qquad (1)$$

also gave no activity in the strontium. We can thus conclude either that reactions tending to produce Sr^{85} are very improbable, or that any Sr^{85} formed has a very short half-life. Slow neutron bombardment of strontium also gave no evidence of a 10-minute period. It seems most likely that the positive activity observed was due to an impurity (possibly N^{13} formed from carbon) which was not completely separated in some of the chemical processes.

If we assume that all stable isotopes of strontium are known, we must conclude that the two negative periods are due to isomers of Sr^{89}, formed by neutron capture:

$$Sr^{88}+H^2 \rightarrow Sr^{89}+H^1 \qquad (2)$$

$$Sr^{89} \rightarrow Y^{89}+e^- . \qquad (3)$$

It was hoped that these two periods might also be obtained from the bombardment of yttrium with fast neutrons, according to the reaction:

$$Y^{89}+n^1 \rightarrow Sr^{89}+H^1 . \qquad (4)$$

Experiment has shown, however, that the activity obtained in strontium by prolonged fast neutron bombardment of yttrium is too small to be significant. It is impossible, with such very weak activity, to state what periods are present or to be sure that the activity is not due to an impurity in the yttrium, but the evidence suggests the possibility of obtaining the expected periods by more intense irradiation.

The ratio of the initial intensities of the 3-hour to the 55-day period was measured after bombarding strontium with deuterons, and from this was calculated the ratio between the rates of formation of the two isomers, corrected for infinite exposure. This ratio was found to vary between 1 : 94 and 1 : 130.

When the strontium was bombarded with slow neutrons, the periods observed were the same as those obtained from deuteron bombardment but the 55-day period was relatively much

[6] The 18-minute period previously observed (reference 3) was not obtained, and the sign of the 3-hour activity was found to be negative instead of positive as there reported.

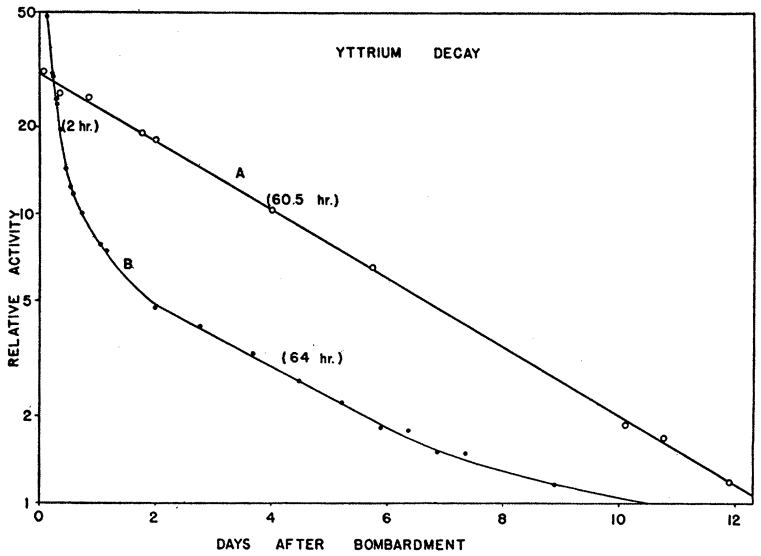

FIG. 2. (*A*) Decay of Y^{90} as produced from $Y+H^2$; (*B*) decay curve for Y separated from $Sr+H^2$.

weaker. In this case the ratio of the yields was approximately 1 : 1. All attempts to alter greatly the ratio of initial intensities of the two periods either by bombarding strontium in different chemical forms or by chemical methods after bombardment were unsuccessful. Likewise the possibility that one of the two negative periods belongs to a strontium isotope of mass greater than 89 (formed, hypothetically, by neutron capture from an unknown stable strontium, as Sr^{90}) was eliminated in part by chemical separations. These showed that the strontium activity did not decay through an yttrium activity of appreciable half-life before reaching a stable zirconium isotope. This was further confirmed by the observed purity of the beta-ray spectra, which would be likely to show the presence of any very short lived secondary yttrium activity.

It was observed that a gamma-ray activity was connected with the decay of the 3-hour period but no such activity was found with the longer period. The gamma-decay was measured by interposing a $\frac{1}{8}''$ thick aluminum sheet between the sample and the ionization chamber of the electroscope. Curves showing the decay of the gamma-activity and of the total strontium activity obtained by bombarding strontium with deuterons are shown in Fig. 1.

RADIOACTIVE ISOTOPES OF YTTRIUM

In the yttrium separated from activated strontium, a composite activity shown in curve *B*, Fig. 2, was observed. This is resolvable into a strong initial period of 120 ± 4 minutes (positive active) and certain longer periods, negative active. The strongest of these, shown as a 64 hour period, resolves into a period of about 60 hours when the very weak residual activity probably due to a contaminant is subtracted. Bombardment of yttrium itself with deuterons gave a very strong negative yttrium activity of single period equal to 60.5 ± 2.0 hours. See Fig. 2(*A*). No shorter periods due to impurities could be observed. The 60.5-hour period was therefore assigned to Y^{90} produced in the reaction:

$$Y^{89}+H^2\rightarrow Y^{90}+H^1. \quad (5)$$

$$Y^{90}\rightarrow Zr^{90}+e^-. \quad (6)$$

This is undoubtedly the 70-hour activity as measured by Hevesy and Levi.[2] When bombarded with fast neutrons, yttrium showed both the 120-minute and the 60-hour periods, with evidence possibly for another, shorter period.[7] Slow neutrons gave the 60-hour period and also some shorter activity which has been attributed to Dy or Th in the original yttrium, a difficulty also encountered by Hevesy.[2] From this evidence, the 120-minute substance must be Y^{88}

[7] The period of 6.5 hours previously reported (reference 3) was not observed. This was undoubtedly due to a contaminant and was responsible, by subtraction, for reporting the 120-minute period, here observed, as 1.2 hours.

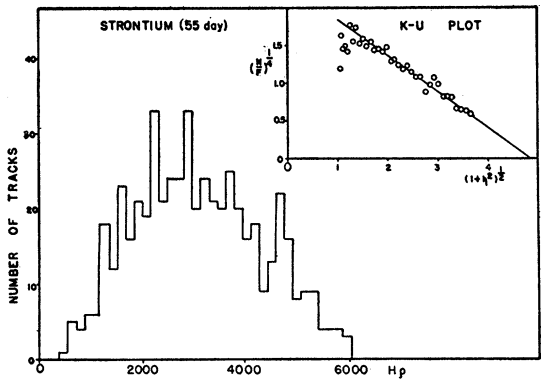

FIG. 3. Beta-ray spectrum and K-U plot for Sr^{89} (55-day period).

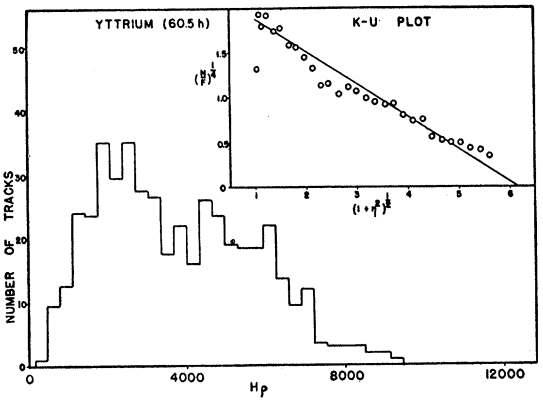

FIG. 5. Beta-ray spectrum and K-U plot for Y^{90} (60.5-hour period).

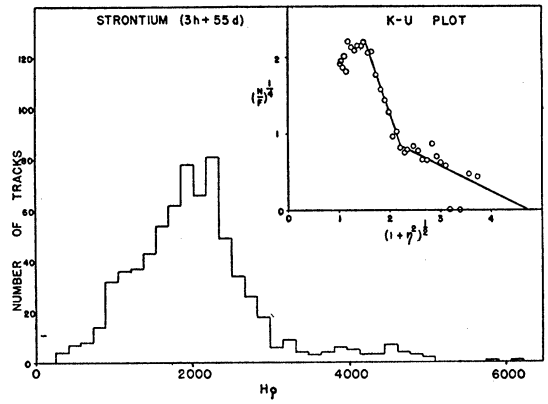

FIG. 4. Beta-ray spectrum and K-U plot for initial activity of Sr^{89} (3-hour and 55-day composite).

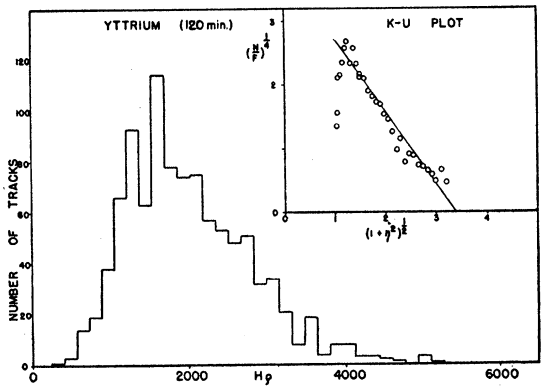

FIG. 6. Beta-ray spectrum and K-U plot for Y^{88} (120-minute period).

formed, by fast neutrons, according to the reaction:

$$Y^{89}+n^1\rightarrow Y^{88}+n^1+n^1 \quad (7)$$

$$Y^{88}\rightarrow Sr^{88}+e^+. \quad (8)$$

If these activities are correctly placed, then it is obvious that the 120-minute period can be formed from strontium only by the following process:

$$Sr^{87}+H^2\rightarrow Y^{88}+n^1. \quad (9)$$

If the 60-hour period in yttrium from strontium bombarded by deuterons is to be identified with Y^{90}, then its production from strontium can be explained only by assuming the complete capture of a deuteron according to the reaction:

$$Sr^{88}+H^2\rightarrow Y^{90}+h\nu \quad (10)$$

or by assuming that the strontium contained yttrium as a major impurity. This latter assumption is highly improbable since the strontium salts used were of the highest obtainable purity. Although deuteron capture has not previously been observed, it is known to be a possible, although not a probable process. In many respects it would be analogous to the capture of a high speed proton—a reaction known to occur in several cases. Bethe and Placzek[8] in discussing the possibility of capture of charged particles by heavy nuclei conclude that deuteron capture should be quite possible but that cases in which it might be observed would probably be rare. Accepting the tentative identification of the 60-hour period as Y^{90}, the present example offers the possibility of calculating the probability of proton capture (from deuteron bombardment) as compared with deuteron capture for isotopes of strontium. (Eqs. (9)–(10).) The ratio of the

[8] Bethe and Placzek, Phys. Rev. **51**, 479 (1937).

rates of formation of the two activities is about 1 : 3. Correcting this ratio for the difference in abundance of the two parent isotopes, Sr^{87} and Sr^{88}, it would appear that the ratio of proton captures by Sr^{87} to deuteron captures by Sr^{88} is about 7 : 2.

Other possibilities for the production of active isotopes of yttrium would be expected from the bombardment of zirconium with deuterons or fast neutrons, followed by the emission of an alpha-particle or a proton, respectively. Preliminary experiments showed that isotopes of yttrium are actually produced in these bombardments and there is indication of the two yttrium periods just reported, but as yet the work of complete identification has not been attempted. Similarly, bombardment of rubidium with alpha-particles of 12–13 Mev has yielded yttrium activities too weak for exact analysis.

Beta-Ray Spectra

It has been possible to obtain tentative values for the upper limits of the energy of the beta-ray spectra of the two isomeric forms of Sr^{89}, for Y^{88} and for Y^{90} by photographing the tracks in a cloud chamber. More than 600 tracks of each kind were measured in a magnetic field of 330 gauss. In Figs. 3 to 6 are shown curves obtained from these data. In each case is given the distribution histogram actually obtained and the Konopinski-Uhlenbeck plot. In Fig. 4, where the tracks photographed were from both the 3-hour and the 55-day periods, the K-U plot is fitted best by two straight lines. The line yielding a higher energy limit is in agreement with the results for the 55-day period, shown alone in Fig. 3. The extrapolated K-U upper energy limits may be summarized as follows:

Sr^{89} (3-hour period) 0.61 Mev;
Sr^{89} (55-day period) 1.9 Mev;
Y^{88} (120-min. period) 1.2 Mev;
Y^{90} (60.5-hour period) 2.6 Mev.

It will be noted that the energy limit for the strontium isomer with the longer half-life is considerably greater than that found for the 3-hour activity. Present concepts of isomeric nuclei place the two excited states very close together. In view of the beta-ray energies found above, this would require the gamma-ray activity associated with the three-hour period to have an energy of about 1.3 Mev, i.e., the difference in energy of the beta-activity from the two isomers. Measurements of the observed gamma-ray energy are yet to be made.

Acknowledgments

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Field Theory of Nuclear Interaction

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The problem of explaining the recent results on nuclear interaction by means of a field theory is studied. Fermi's theory of the electron-neutrino field is used as a model which is sufficient to account for the symmetries of the problem, although it fails to explain the order of magnitude of the forces. The equality of forces between like and unlike particles is exactly accounted for by introducing interaction terms involving the emission of electron pairs or neutrino pairs. The interaction law may be stated very simply with the aid of an isotopic spin variable for light as well as for heavy particles. The ratio of force constants obtainable from the theory of mass defects may be accounted for in detail by a suitable choice of the light particle field. However, it is difficult to explain any law involving more than one potential function $J(r)$.