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Radioactive Isotopes of Lanthanum

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The radioactive isotope La^{140} , known to be produced by deuteron and slow neutron bombardments of lanthanum is found to have a half-life of 40.0 ± 0.3 hours. This isotope has now been produced by the reaction $\text{Ce}^{140}(n, p)\text{La}^{140}$. Evidence for the occurrence of a d, γ reaction has been obtained through the formation of La^{140} by the reaction $\text{Ba}^{138}(d, \gamma)\text{La}^{140}$. The 40-hour isotope decays with the emission of 1.41 ± 0.05 -Mev electrons and 2.00 ± 0.05 -Mev gamma-rays. Deuteron and also proton bombardments of

barium produce a new radioactive isotope of lanthanum of half-life 17.5 ± 0.5 hours. This activity decays by the process of K -electron capture with the emission of x-rays identified as the characteristic K radiation of barium. The presence of a 0.88 ± 0.1 -Mev gamma-ray of low intensity indicates that the resulting barium nucleus may also be left in an excited state. This 17.5-hour activity is assigned to La^{137} .

INTRODUCTION

ARTIFICIAL radioactivity in lanthanum was first induced by a slow neutron activation of the oxide, from which a 45.6 ± 4.8 -hour activity, assigned to La^{140} , was obtained.¹ The determinations of the half-life of the lanthanum activity produced by different nuclear processes and described by several investigators vary to a rather large extent, namely, from 31 hours to 46 hours.²⁻⁶

It is the purpose of this paper to investigate more accurately the La^{140} period and in addition to describe a new 17.5-hour period found in lanthanum after activation of barium with deuterons and protons.

¹ J. K. Marsh and S. Sugden, *Nature* **136**, 102 (1935).

² M. L. Pool and L. L. Quill, *Phys. Rev.* **53**, 437 (1938).

³ O. Hahn and F. Strassmann, *Naturwiss.* **27**, 11 (1939).

⁴ O. Hahn and F. Strassmann, *Naturwiss.* **28**, 54 (1940).

⁵ W. G. Chlopin, M. A. Passwik-Chlopin, and N. F. Wolkov, *Nature* **144**, 595 (1936).

⁶ G. N. Glasoe and J. Steigman, *Phys. Rev.* **58**, 1 (1940).

EXPERIMENTAL

All bombardments were carried out in the 42-inch cyclotron of The Ohio State University. The bombarding particles used were approximately 20-Mev alpha-particles, 10-Mev deuterons, 5-Mev protons, slow neutrons produced by the $\text{Be}+d$ reaction, and fast neutrons from the $\text{Li}+d$ reaction. Chemical separations were made after all activations except those with slow neutrons.

In all bombardments of lanthanum and cerium, Hilger H.S. brand oxides were used while, in the barium activations, Hilger "Specpure" barium chloride was used. A spectroscopic analysis accompanying the material revealed the following maximum impurities:

Lanthanum oxide, 99.9 percent, Lab No. 6404
—very slight traces of Na, Mg, Ca, Bi, Sm;
Gd, 0.015 percent; Nd, 0.1 percent.

Cerium oxide, 99.99 percent, Lab No. 6401—
traces of Mg and Gd.

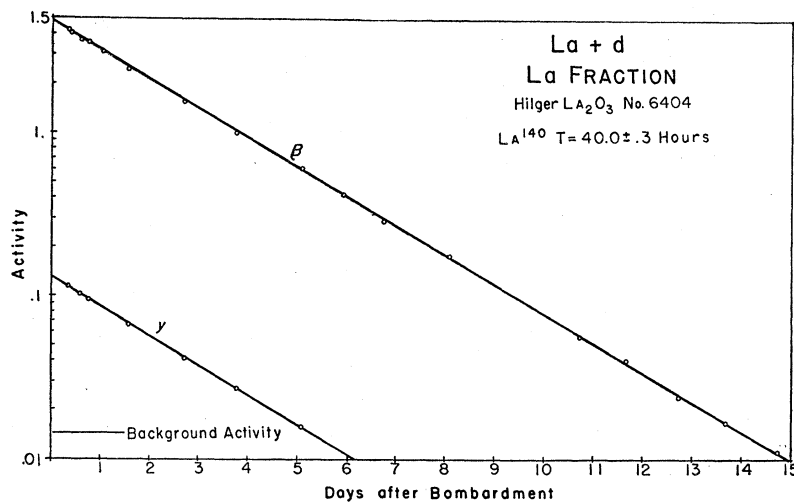


FIG. 1. Decay curve for lanthanum fraction from $La+d$.

Barium chloride, 99.98 percent, Lab No. 8442
—Ca, 0.0015 percent; Fe, 0.005 percent;
Mg, less than 0.001 percent; Na, about 0.001
percent; Pb, 0.01 percent; and Sr, absent.

The other source of barium used was a special grade of the nitrate prepared by a modification of the method used in preparing barium for atomic weight determinations. Spectroscopic analysis of this material revealed that it was of a high degree of purity. Hilger cesium chloride was also bombarded in the course of the work.

Measurements of the radioactivity were made on a Wulf unifilar electrometer with an ionization chamber containing two atmospheres of freon, or, in some cases, on another Wulf unifilar electrometer with an ionization chamber having a 0.25-mil aluminum window and containing air at atmospheric pressure. The latter chamber was used in the detection of soft beta-radiation.

For the study of the emitted radiations, a Wilson cloud chamber of 15-cm diameter and a 180° focusing type magnetic spectrometer with a radius of curvature of 16 cm were available.

THE 40-HOUR PERIOD

A very intense 40.0-hour lanthanum activity emitting electrons and gamma-rays, was produced by the deuteron bombardment of the lanthanum oxide. The procedure used for the purification of lanthanum after activation was

identical with that described for praseodymium.⁷ Since the decay curve of the deuteron-activated lanthanum, shown in Fig. 1, was linear through 11 half-lives, a very reliable evaluation of the period could be made. From an average of three such determinations, the half-life was found to be 40.0 ± 0.3 hours.

Activation of lanthanum with slow neutrons gave a value of 39.8 hours for the period of La^{140} . Although the activity produced in this case was weak compared to that produced by the deuteron bombardment, it yet was easily followed through several half-lives.

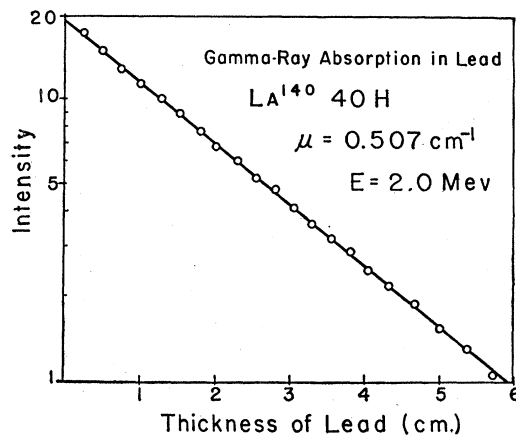


FIG. 2. Gamma-ray absorption in lead of 40-hour lanthanum.

⁷J. W. DeWire, M. L. Pool, and J. D. Kurbatov, *Phys. Rev.* **61**, 564 (1942).

After cerium was bombarded with fast neutrons a lanthanum fraction was separated from the cerium. The activity of the lanthanum fraction also decayed with a 40-hour period. This activity was likewise weak compared to that obtained by deuteron bombardment. The new reaction, $Ce^{140}(n, p)La^{140}$ seemed therefore to be established.

An energy of 2.0 Mev for the gamma-rays emitted by the 40-hour period has been determined by absorption in lead. Measurements, as shown in Fig. 2, made with a very strong source gave a single absorption coefficient of $0.507 \pm 0.005 \text{ cm}^{-1}$, corresponding to an energy of $2.00 \pm 0.05 \text{ Mev}$.

From aluminum absorption measurements, shown in Fig. 3, and from a beta-ray spectrum, shown in Fig. 4, taken with the magnetic spectrometer, the maximum energy of the beta-particles was estimated to be $1.41 \pm 0.05 \text{ Mev}$. A tail of high energy Compton recoil electrons produced by the 2.0-Mev gamma-rays was observed in both cases.

In Fig. 5, a section of the periodic table is reproduced showing the assignments of and the reactions producing the various radioactive isotopes in the lanthanum region.

THE 17.5-HOUR PERIOD

A new radioactive isotope of lanthanum with a half-life of 17.5 hours has been found to be pro-

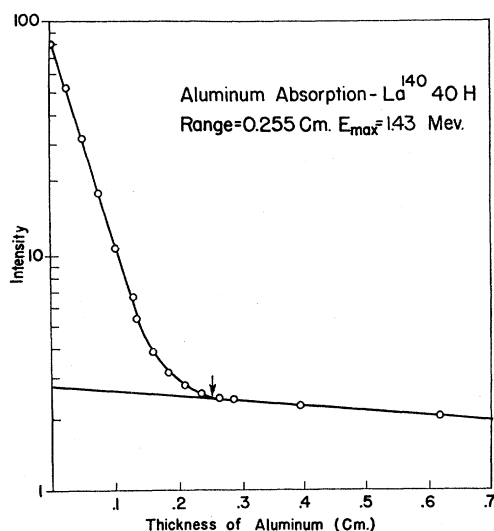


FIG. 3. Beta-ray absorption in aluminum of 40-hour lanthanum.

duced by bombardments of barium with 10-Mev deuterons. From the lanthanum fractions of these bombardments, complex decay curves were obtained; a portion of one is shown in Fig. 6. The longer period was observed to be 40 hours. Upon subtraction of this activity from the original curve, a period of 17.5 ± 0.5 hours resulted. Proton bombardment of barium gave only a 17.5-hour activity in the lanthanum fraction.

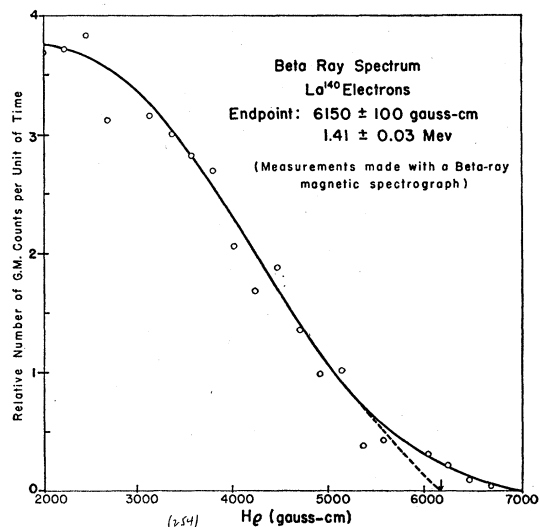


FIG. 4. Beta-ray spectrum of 40-hour lanthanum.

In order to observe the 17.5-hour period a careful separation of barium and lanthanum is necessary because of the very strong beta-radiation emitted by Ba^{139} of 85.6-minute period. It has been found that even 0.01 percent of this activity immediately after bombardment, if left in the lanthanum fraction, is sufficient to mask the correct evaluation of the 17.5-hour period.

The chemical procedures were arranged in the following manner: Bombarded barium chloride, which was contained in a platinum holder, was dissolved in nitric acid with the addition of lanthanum oxide. The radioactive chlorine was mainly removed by evaporation and the nitrate solution was converted into a chloride solution. Added platinic acid was reduced and platinum was filtered off.

Separation of barium and lanthanum was first made with ammonium hydroxide. Precipitated lanthanum was dissolved and reprecipitated as an oxalate in presence of stabilizers for active

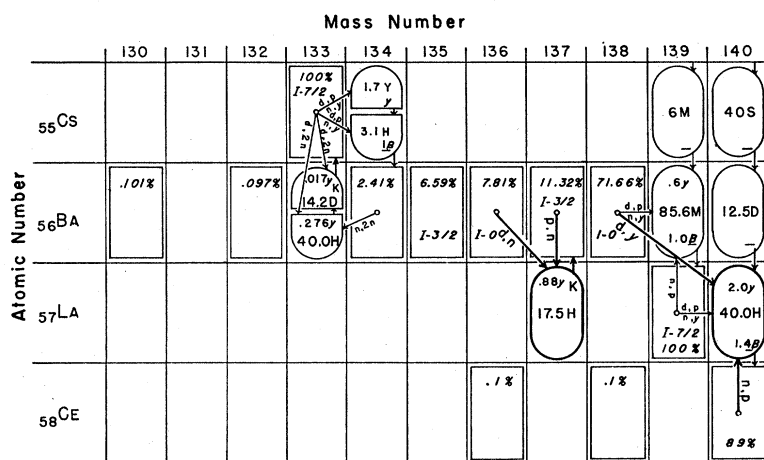


FIG. 5. Isotopic chart of the lanthanum region.

impurities other than those of the rare-earth group. Separate purification of lanthanum was performed from phosphoric acid to remove radioactive phosphorus, which was present because of the $\text{Cl}^{35}(n, \alpha)\text{P}^{32}$ reaction.

K-Electron Capture

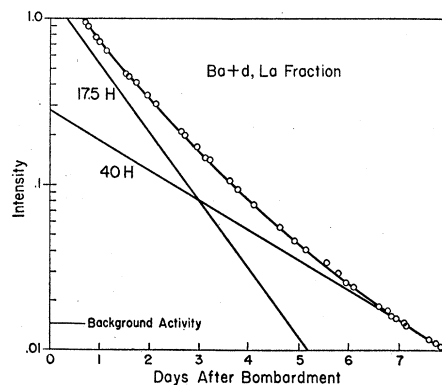
The radiation from the 17.5-hour period consisted entirely of x-rays and gamma-rays as evidenced in the aluminum absorption curve of Fig. 7 and in the copper absorption curve of Fig. 8. Because of the presence of a low intensity gamma-ray, a gradual bending of these curves was observed. Upon subtraction of the activity of the gamma-ray in each case, new curves were obtained, from which were determined mass absorption coefficients of $0.86 \text{ cm}^2/\text{g}$ in aluminum and of $9.11 \text{ cm}^2/\text{g}$ in copper. These coefficients corresponded, respectively, to x-ray wave-lengths of 0.367A and 0.389A , or to an average wave-length of $0.378 \pm 0.011\text{A}$. The close agreement of this wave-length with the estimated mean wave-length, 0.376A , of the K series for barium, suggested at once that the radioactive lanthanum might be decaying by the process of K -electron capture.

The deuteron bombardment of barium could, however, give rise to a radioactive isotope of the stable La^{139} , which would then decay to the ground state by the emission of a gamma-ray. A possibility therefore exists that the x-rays observed might arise from the internal conversion

of this gamma-ray. As the K -radiation produced in this case would be that of lanthanum while in the case of K capture it would be that of barium, a more refined measurement of the wave-length of these x-rays should make it possible to distinguish between the two processes.

Critical Absorption

The most widely used method for the determination of the wave-length of the x-rays emitted by radioactive substances is provided by critical absorption. From the data in Table I, it may be seen that tellurium should absorb the x-radiation from both barium and lanthanum strongly, but that iodine should absorb only about 20 percent of the barium x-rays as compared with 74 percent of the lanthanum x-rays. In addition, if the K radiation were that of lanthanum, the iodine

FIG. 6. Decay curve for lanthanum fraction from $\text{Ba}+d$.

absorption curve should reveal the presence of the two principal lines of the K series, one of which ($K\alpha_1$) would be absorbed and the other ($K\alpha_2$), transmitted.

Filters of powdered tellurium metal and of sodium iodide were therefore prepared for the purpose of determining the mass absorption coefficients of the x-rays in the two substances. The main source of error in measurements of this kind lies in the difficulty in preparing thin filters of uniform thickness.⁸ An estimate of this error, for an extreme case, was made by measuring the absorption coefficients for the x-rays emitted by the 17.5-hour isotope with tin foils of 9.8 mg/cm² and with filters of 30-mesh tin powder. The coefficient obtained with the foils was 28.9 cm²/g, which is very nearly the true value, while with the powder, it was found to be only 12.6 cm²/g.

The x-rays from the 17.5-hour activity had a mass absorption coefficient, in tellurium, of 18 cm²/g. A correction, which was estimated from the tin absorption measurements just described, was made necessary by the fact that the thin filters of tellurium metal used were not homogeneous. The value of the absorption coefficient in tellurium then obtained was approximately 30 cm²/g, showing that the x-rays were strongly absorbed by the metal. This strong absorption was to be expected for either barium or lanthanum x-rays.

In iodine, a single mass absorption coefficient

TABLE I. Data on K series lines of barium and lanthanum

K series line	$K\alpha_2$	$K\alpha_1$	$K\beta_1$	$K\beta_2$
Fraction of total intensity	0.26	0.53	0.16	0.05
Ba wave-length (Å)	0.38899	0.38443	0.34022	0.33222
La wave-length (Å)	0.37466	0.37004	0.32726	0.31966

Critical absorption limits: Te, 0.38926Å; I, 0.37244Å.

of 8.5 cm²/g was found for the x-rays from the 17.5-hour period. Correction for inhomogeneity was relatively small in this case. Barium x-rays would give a simple absorption curve in iodine, corresponding to a mass absorption coefficient estimated to be approximately 11.7 cm²/g. The observed value, 8.5 cm²/g, is in satisfactory

⁸ A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), second edition, p. 517.

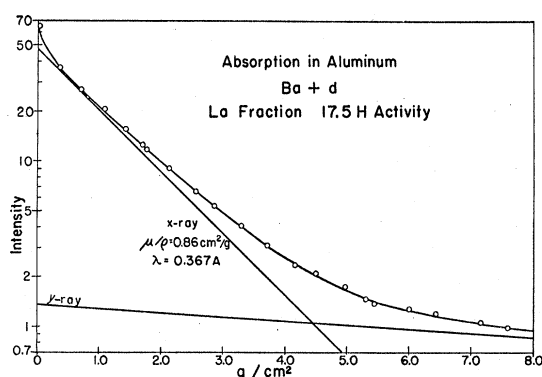


FIG. 7. Aluminum absorption curve for 17.5-hour lanthanum.

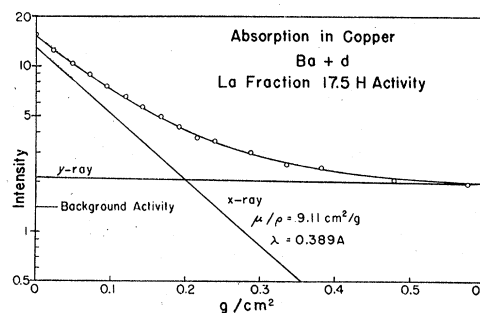


FIG. 8. Copper absorption curve for 17.5-hour lanthanum.

agreement with this estimated coefficient. As the iodine was relatively transparent to the x-rays, the possibility that the K radiation was that of lanthanum seemed definitely excluded.

The evidence from these critical absorption measurements, therefore, leads to the conclusion that the x-rays are those of barium and that, consequently the 17.5-hour isotope decays by the process of K -electron capture.

The 0.88-Mev Gamma-Ray

The gamma-ray observed in the aluminum and copper absorption measurements was also found to decay with a half-life of 17.5 hours. From lead absorption measurements, the energy of this penetrating radiation was determined to be 0.88 ± 0.1 Mev. Should this gamma-ray be internally converted, electrons of approximately 0.84 Mev would be produced. As no such electrons were observed, the possibility that the x-rays might arise from the internal conversion of this gamma-ray has been excluded independently of

the identification of the K x-rays as those of barium.

No positrons were observed in any of the cloud-chamber photographs taken of the 17.5-hour activity. This would indicate that K -electron capture is a very much more probable type of disintegration for this isotope than positron emission. The absence of annihilation radiation also confirmed the failure to observe positrons.

The number of x-rays relative to the number of gamma-rays is about 50 which means that in about two percent of the capture processes the barium nucleus is left in an excited state.

Assignment

The assignment of the 17.5-hour period has been made to La^{137} as illustrated in Fig. 5. Bombardments of cesium with alpha-particles and of lanthanum with fast neutrons failed to produce any activity in the lanthanum fractions other than a small amount of the 40-hour activity in the latter case. It seems therefore unlikely that the 17.5-hour activity can be due to either La^{136} or La^{138} . It is improbable that the lighter and less abundant isotopes of barium would produce an activity as intense as was obtained. In view of these considerations, the activity was assigned to La^{137} , which is probably produced by the reactions $\text{Ba}^{136}(d, n)\text{La}^{137}$ and $\text{Ba}^{137}(p, n)\text{La}^{137}$.

EVIDENCE FOR A d, y REACTION

A weak 40-hour period was observed in the lanthanum fractions from the deuteron bombardments of barium. The decay curve for one

such fraction, shown in Fig. 6, has already been considered. It was found that the ratio of the intensity of the total radiation for this 40-hour activity to that of the gamma-radiation alone was of the same order of magnitude as that obtained previously for the 40-hour activity of La^{140} .

The activity of the 17.5-hour lanthanum calculated for an infinite bombardment, relative to that calculated for this 40-hour period remained practically constant in each of the several deuteron activations of barium. As the spectroscopic analysis for both sources of barium used in the bombardments showed the complete absence of lanthanum, there was no possibility of producing the 40-hour activity by a d, p reaction from a lanthanum impurity. It therefore seems reasonably certain that the reaction $\text{Ba}^{138}(d, y)\text{La}^{140}$ does take place.

There are only a few cases where an activity can be produced unequivocally by a d, y reaction. In general, the distribution of the stable isotopes is such that the more probable d, n or $d, 2n$ reactions will occur with a consequent masking of any contribution made by a d, y reaction.

The probability that the d, y reaction will occur in this particular case, relative to the occurrence of the d, n reaction was calculated to be 1 to 1300.

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