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Ernest O. Lawrence

*Radiation
Laboratory*

RANGE AND RANGE STRAGGLING
OF HEAVY RECOIL ATOMS

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OF HEAVY RECOIL ATOMS

Ernest William Valyocsik
(Master's Thesis)

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ABSTRACT

The range and range straggling of 96.8-keV Ra^{224} recoil atoms produced by alpha decay of Th^{228} have been measured in H_2 , D_2 , He , N_2 , Ne , and A by a charged, parallel-plate, collection technique. The range and range straggling of 725-keV Th^{226} recoil atoms produced by bombardments of Ra^{226} with 41.6-MeV helium ions have been measured in D_2 , He , N_2 , and A by this same technique. Experimental results seem to indicate that the range-energy relation for the stopping of heavy recoils by nuclear collisions is not linear, as is theoretically predicted by Bohr. The distribution of ranges about the mean range appears to be Gaussian. Various factors contributing errors to the measured range straggling are discussed in detail...

Recoil efficiencies of the heavy recoils produced in the (d,n) , $(d,2n)$, $(d,3n)$, and (d,p) reactions of Bi^{209} have been measured at incident deuteron energies of 15.0 to 23.6 MeV for Bi^{209} targets of 0.6 to 42.5 $\mu\text{g}/\text{cm}^2$. The recoil efficiencies of the different reaction products have been compared on the basis of probable nuclear-reaction mechanisms by which they are produced. Mean ranges for Po^{208} in Bi^{209} have been calculated from the recoil efficiencies.

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I. INTRODUCTION

The recoil-range work described in this paper was begun with the object of testing the theoretical predictions made by Bohr¹ for the stopping of particles by nuclear collisions. Bohr considers stopping by nuclear collisions (elastic encounters between atoms as a whole) to dominate over electronic stopping, in which energy is transferred to individual electrons, in the region where the recoil velocities are near or less than v_0 , where $v_0 = e^2/\hbar^2$ and e is the electron charge.

When stopping is by nuclear collisions, Bohr predicts that the ranges of particles should be proportional to their energies. Lindhard and Scharff² find the range due to nuclear collisions to be given, to a close approximation, by

$$R = \frac{e^2}{\pi^2 a_0^2} \frac{M_1 + M_2}{2\mu N} \frac{v^2}{v_0^2} \frac{\sqrt{Z_1^{2/3} + Z_2^{2/3}}}{Z_1 Z_2} \quad (1)$$

Where M_1 , Z_1 , and v are the mass number, atomic number, and velocity, respectively, of the recoil atom; M_2 , Z_2 , and N are the mass, number, atomic

number, and density of the stopping medium, respectively, and we define $a_0 = \hbar^2/\mu e^2$, where μ is the mass and e is the base of natural logarithms.

Another form of the Lindhard and Scharff equation is given by³

$$R(\mu\text{g}/\text{cm}^2) = \frac{602EM_2(M_1 + M_2)^{1/2}}{M_1^{1/2} Z_1 Z_2} \sqrt{Z_1^{2/3} + Z_2^{2/3}}, \quad (2)$$

where E is the recoil energy in Mev. The probability that a particle in a group of particles with mean energy R_0 , will have a range between R and $R+dr$ is given by $W(R)dr$ where

$$W(R) = \frac{1}{\rho R_0 \sqrt{2\pi}} \exp \left[-\frac{(R - R_0)^2}{2\rho^2 R_0^2} \right], \quad (3)$$

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for which Lindhard and Scharff find the straggling parameter ρ to be, to a close approximation,

$$\rho^2 = \frac{2 M_1 M_2}{3(M_1 + M_2)^2} \quad (4)$$

The measurement of the ranges in different gases of heavy recoil atoms from natural alpha decay of various nuclides has been of interest for nearly half a century.⁴⁻¹³ Much of the earlier work has proven unsatisfactory and in wide disagreement.¹³ Some of the earlier workers collected the recoil atoms electrostatically by taking advantage of the fact that the recoils carry positive charges¹⁴⁻¹⁶ near the end of their ranges.

In this work, the recoil atoms were stopped in gases between two parallel metal plates and collected electrostatically upon an aluminum strip. This technique was first used in this laboratory by A. Ghiorso and T. Sikkeland.¹⁷ The advantage of this procedure is that it provides a convenient and sensitive method for measuring the range and range straggling. The ranges of heavy recoil atoms have been measured by stopping them in thin metal foils^{18,19} and in thin nonmetallic films.²⁰⁻²² The disadvantage of very thin films is that they are hard to make and difficult to handle.

Thorium-228 was selected as an essentially monoenergetic source of Ra^{224} recoil atoms in order to determine the magnitude of the range straggling produced by collisions with the stopping gas alone. With recoils produced by nuclear reactions there is an additional range straggling effect caused by emission of particles from the excited nuclei of the recoil atoms.²² The range and range straggling of Ra^{224} were measured in H_2 , D_2 , He , N_2 , Ne , and A .

Thorium-226 was selected as a recoil atom in the range work carried out at the 60-in. cyclotron for the following reasons:

- (a) Data obtained by R. Vandenbosch²³ for the $\text{Ra}^{226}(\alpha, 4n)\text{Th}^{226}$ reaction indicated a large cross-section at the peak of the excitation function where one could still expect the reaction to be proceeding by a compound-nucleus mechanism.²⁴ The entire

momentum of the incident particle is transferred to the nucleus penetrated when compound-nucleus formation takes place. One can then easily calculate the recoil energy of the heavy recoil atom. Short helium-ion bombardments were sufficient to produce adequate quantities of Th^{226} .

- (b) The collection efficiency was ~90%. It was found that polonium and astatine recoils could not be collected with this technique.
- (c) Th^{226} is a short-lived alpha emitter which permits measurement of the Th^{226} collected by gross alpha counting and which produces a high disintegration rate during decay.

The range and range straggling of Th^{226} were measured in D_2 , He, N_2 , and A.

Deuteron induced reactions were studied at incident energies of 15.0 to 23.6 Mev with Bi^{209} targets of thickness 0.6 to $42.5 \mu\text{g}/\text{cm}^2$. Bismuth-209 was selected as a target because of the availability of good cross-section data,²⁵ which were very useful in determining the activity to be expected in experiments so that the recoil collection efficiencies could be measured for the different reaction products. The recoil efficiency was determined by measuring the fraction of the recoils that escaped from targets of different thicknesses. It was of interest to obtain quantitative results for the yields expected of recoil atoms escaping from Bi^{209} targets under different experimental conditions, because since the recoil technique previously has been used in this laboratory^{22,26} in cross-section measurements. In the cross-section work, recoil efficiencies of product atoms were assumed to be high for thin targets. The recoil efficiencies of the different reaction products were compared on the basis of probable nuclear-reaction mechanisms instrumental in their formation. Mean ranges in Bi^{209} for the (d,3n) reaction product, Po^{208} , were calculated from the recoil efficiencies by the method employed by Chetham-Strode.²⁶

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II. Ra^{224} RECOIL ATOMS FROM ALPHA-DECAY OF Th^{228}

A. Production and Separation of U^{232}

It was necessary to produce the U^{232} , from which the Th^{228} sources were derived, by neutron irradiation in the Materials Testing Reactor (M.T.R.) at Arco, Idaho of 2.3 mg of Pa^{231} sealed in a quartz tube. Sufficient time was allowed between the removal of the quartz tube from the M.T.R. and the chemical separation of the U^{232} for most of the Pa^{232} (1.31 d, β^-)²⁷ to have decayed.

The quartz tube was scratched and carefully broken open. The solid mass was slurried into a platinum dish with a mixture of 12 M HCl and ca. 1.0 M HF. The platinum dish was slowly heated to accelerate dissolution of the solid. The protactinium resisted dissolution; therefore, it was necessary to leach the U^{232} from the undissolved solid by repeated treatments with the HCl-HF solution.

Protactinium²⁸ and uranium are known to adsorb strongly on anion resins in greater than 8 M HCl, but with as much as 0.1 M HF present in the HCl protactinium does not adsorb.²⁹ Uranium adsorption remains unchanged with HF present.³⁰ The use of HF required that polyethylene transfer pipettes and test tubes be used. A column was constructed from a piece of lucite 2 cm by 2 cm by 8.5 cm by drilling a 1/8-in. diam. hole down the center with a 1/16-in.-diam. hole at the lower end. The lucite column was filled with about 4 cm of Dowex A-1 anion resin with shreds of polyethylene forming the plug at the bottom of the column. The anion resin was washed repeatedly with conc. HCl and distilled water in order to remove as much as possible of any residues and impurities which might pass through the column during the separation procedure.

The separation of U^{232} from other undesirable elements was accomplished by transferring the HCl-HF solution on to the anion-resin bed. Protactinium and various fission products did not adsorb on the resin and were collected in a polyethylene cone. An aliquot of this liquid was placed on a platinum disc, evaporated, flamed, and counted in an alpha-particle pulse-height analyzer³¹ to determine whether or not U^{232}

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had passed through the column and whether Pa^{231} was present. The resin was washed several times with 12 M HCl to remove fluoride ion. Aliquots of the wash were analyzed, also. After thorough washing with a 12 M HCl, a clean test tube was placed under the resin column, and the U^{232} was eluted with 0.5 M HCl. Pulse-height analysis of aliquots of this solution revealed that $\sim 36 \mu\text{g}$ of U^{232} free of other alpha-emitting nuclides had been separated from the protactinium. The 0.5 M HCl solution containing the U^{232} was then evaporated to dryness and the HCl driven off. Then to these dried salts was added 6 M HCl in preparation for the electrodeposition of U^{232} .

B. Preparation of Th^{228} Sources

The ideal source for range and range straggling measurements of heavy recoil atoms from alpha-decay would be one that was so thin that there would be no degradation in the energy of the recoil escaping from the surface of the source. It is possible to produce extremely thin sources by collecting recoils from alpha-decay that have been stopped in air at atmospheric pressure upon negatively charged collectors under a potential of a few hundred volts per cm. The mean-free-path for the recoil atoms at atmospheric pressure is so short that the atoms do not accelerate to high enough velocities to penetrate the surface of the collector plate. These atoms in turn decay by alpha-emission, producing the recoil atoms whose ranges are to be studied. With other known methods for source preparation there is a high probability of producing films over the surface of the source material.

Uranium-232 (74y, α) 32 was selected because its daughter, Th^{228} (1.91 y, α) 33 possessed a half life long enough that it would provide a source that would not decay appreciably during the duration of the experiment. Ra^{224} (3.64 d, α) 34 possesses a convenient half life for range experiments.

Uranium-232 was electroplated on the inner surface of a 125-ml platinum dish. The 6 M HCl solution was neutralized with NH_4OH until the solution was just acid to methyl red.²⁶ The liquid volume was

controlled so that an area of about 25 cm^2 of the platinum dish was in contact with the plating solution. Thus it was hoped the U^{232} thickness would be about $1 \mu\text{g}/\text{cm}^2$, and the maximum number of Th^{228} recoils would escape into the air. The electrodeposition was made by plating at 10 v with a current of 4 amp for 15 min. It was found that $\sim 30 \mu\text{g}$ of U^{232} had been plated on to the platinum dish. This was determined by pulse-height analysis of a few aliquots of the remaining solution.

Platinum foil was cut into strips $3/8$ -in. wide and 3 in. long to be used for the Th^{228} collectors. A strip was placed into a slit in a cork and supported in a buret clamp. The clamp was attached to a ring stand on the base of which the platinum dish rested. The lower end of the platinum strip was bent into a right angle, and the strip was lowered to a point which was approximately 3 cm from all parts of the platinum dish. Five 300-v batteries were connected in series with a 1 megohm resistor between each pair of batteries and leads were attached to the ring stand and the platinum strip, making the platinum strip the negative electrode. The entire unit was housed under a bell jar in a glove box as protection against airborne impurities settling on the platinum strip and platinum dish. The two sources used in the range experiments were each made by collecting Th^{228} recoils for about 3 months. The resulting source strengths were $\sim 10^6$ disint./min. in Th^{228} .

C. Experimental Apparatus

A vacuum system, shown in Figs. 1 and 2, was constructed to be used in the range measurements on Ra^{224} recoil atoms. A mechanical pump was connected to the system in series with an oil-diffusion pump. During evacuation, the pressure of the system was read from two meters controlled by thermocouple gauges. The oil-diffusion pump was protected by a unit which would shut off the pump if the pressure of the system rose above 100 microns during evacuation or if the flow of cooling water to the pump stopped. Two glass traps inserted into liquid nitrogen were placed in the system between the pumps and the bell jar in order that any volatile matter would be trapped out of a gas before it reached the bell jar.

MU-18168

Fig. 1. Schematic diagram of the vacuum system used in the measurement of ranges of Ra^{224} recoil atoms: (a) bell jar, (b) diffusion pump, (c) mechanical pump, (d) glass traps in liquid nitrogen, (e) thermocouple gauges, (f) differential oil manometer, (g) mercury manometer.

ZN-2277

Fig. 2. Vacuum system used in the measurement of ranges of Ra^{224} recoil atoms: (a) oil manometer, (b) power supply, (c) diffusion-pump control, (d) thermocouple gauge for forevacuum pressure.

This was a precaution taken to prevent a film of any foreign matter from covering the source. Cylinders of gases were connected to the system by means of tygon tubing attached to the regulator valves. The system was so constructed that all gases entering the bell jar had to pass through a liquid-nitrogen trap. The pressures of the stopping gases were measured with the aid of a differential oil manometer that had been calibrated against a mercury manometer to which it was connected.

A unit, which was housed inside the bell jar, was constructed from glass (Fig. 3) to support the source and aluminum parallel plates. Glass was used for this unit instead of plastic because of the tendency of plastic to adsorb gases on its surface and outgas. Aluminum strips, 2.4 cm by 12.0 cm with lines marked 2 mm apart along the lengths of their surfaces, were used as recoil collectors. The marked segments were numbered from the edge nearest the source. A copper wire, which served as the contact for the negative plate was brought into the bell jar through a rubber stopper cemented into the base plate.

The 1-5/16-in.-long recoil collimator was constructed from 3/8-in.-diam. copper tubing with a copper flange attached. The source was located between an aluminum disc and the flange of the collimator. The flange and aluminum disc were screwed tightly together. The aluminum disc was screwed to a copper plate, and the collimator was centered between the parallel plates through a hole in the glass support. It was found that when a polystyrene collimator was used, the Ra^{224} recoils collected only at the edge of the collector strip nearest the source.

D. Experimental Procedure for Range Measurements

As a precaution, the Th^{228} source was removed from the collimator and flamed before an experiment in order to remove any matter which may have settled on it. The source appeared bright and clean to the naked eye after flaming. A collector strip was positioned on the lower parallel plate, the bell jar was seated over the base plate, and the system was evacuated to ~100 microns with the mechanical pump. The valve to the diffusion pump was then opened, and the system was evacuated to $\sim 10^{-3}$ mm

ZN-2278

Fig. 3. Unit used in Ra^{224} range measurements: (a) glass support for source and parallel plates, (b) aluminum collector strip in position for collection on the lower aluminum parallel plate.

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Hg. The bell jar was flushed twice with the stopping gas; each time the system was evacuated with the diffusion pump. After flushing was completed the pumps were valved off from the system. The gas pressure was adjusted, with the aid of the differential oil manometer, to such a value that the mean linear range of the recoil atoms in the gas would be ~10 cm, assuming that the Lindhard-Scharff range-energy relation in Eq. (2), was a good approximation. This placed the peak Ra^{224} activity near the center of the collector strip. The bell jar was isolated from the rest of the system during the collection period. The lower parallel plate was maintained at a potential of -200 v relative to the upper plate during collection. Because of the low source-activity level and the small solid angle of collimation, it was necessary to collect Ra^{224} recoils for periods of 12 to 16 hr. At the end of each experiment the gas pressure in the bell jar was remeasured with the oil manometer to determine whether any pressure change had occurred during the recoil collection period.

E. Counting Procedure

The aluminum collector strip was cut into segments along the lines scratched and numbered on its surface. The amount of Ra^{224} deposited on each segment was then measured by alpha pulse-height analysis (Fig. 4).

F. Results

The counting results from each collector segment were treated as follows. The Ra^{224} activity of a segment, derived from pulse analysis, was divided by the length in millimeters of that segment. This placed the Ra^{224} activity collected on the aluminum strip on a relative basis. From the calculated density of the stopping gas in a given experiment, the linear ranges were converted to units of $\mu\text{g}/\text{cm}^2$ of stopping gas.

Differential range curves were obtained when the counts per minute (c/m) of Ra^{224} per mm were plotted on a relative scale versus the

MU-18169

Fig. 4. Alpha pulse-height analysis of a segment of the Ra²²⁴ collector strip with D₂ as the stopping gas.

cumulative stopping medium thickness in $\mu\text{g}/\text{cm}^2$. Figures 5, 6, and 7 show the range curves for Ra^{224} recoils in H_2 , D_2 , and He , respectively.

The experimental range results for Ra^{224} are summarized in Table I. The mean range, R_o , and the straggling parameter, ρ , are listed, together with the theoretical values for these quantities calculated from Eqs. (2) and (4). Possible sources of error in the straggling parameter, ρ , and their magnitudes are treated in the discussion of range results for 96.8-keV Ra^{224} recoil atoms.

Table I

Experimental and theoretical values for 96.8-keV Ra^{224} recoil atoms				
Gas	Experimental		Theoretical	
	R_o ($\mu\text{g}/\text{cm}^2$)	ρ	R_o ($\mu\text{g}/\text{cm}^2$)	ρ
H_2	3.9 ± 0.1	0.136	3.02	0.0542
D_2	6.6 ± 0.1	0.145	6.09	0.0765
He	6.6 ± 0.2	0.162	6.24	0.107
N_2	7.0 ± 0.3	0.238	6.87	0.192
Ne	7.3 ± 0.4	0.264	7.14	0.224
A	9.4 ± 0.6	0.315	8.98	0.292

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Fig. 5. Range curve for 96.8-keV Ra^{224} recoil atoms in H_2 .

MU-18166

Fig. 6. Range curve for 96.8-keV Ra^{224} recoil atoms in D_2 .

MU-18170

Fig. 7. Range curve for 96.8-keV Ra^{224} recoil atoms in He.

III. Th^{226} RECOIL ATOMS PRODUCED BY He^4 IONS ON Ra^{226}

A. Preparation of Ra^{226} Targets

Radium targets were prepared by vaporizing anhydrous RaCl_2 in vacuo on to 0.001-in.-thick aluminum foil which was attached to the inner top surface of the bell jar. Isotopically pure Ra^{226} was used. The procedure used was to transfer aliquots of the radium chloride solution on to a tantalum filament. With the bell jar removed, the water was evaporated at atmospheric pressure from the electrically heated filament until there was no longer any sign of water vapor. As a precaution against vaporizing any impurities from the filament on to the aluminum foil, the tantalum filament was flashed twice at atmospheric pressure by rapidly increasing and decreasing the electric current passing through it. The bell jar with the aluminum foil attached to the top was then seated over the filament and evacuated. When the bell jar was evacuated to $\sim 10^{-3}$ mm Hg, the filament was flashed several times to vaporize the RaCl_2 .

The amount of Ra^{226} deposited per unit area was determined by cutting measured areas from the radium-coated aluminum foil and counting these samples in a low-solid-angle alpha-counter. The Ra^{226} thickness was calculated from the solid angle of the counter and the specific activity of Ra^{226} . Targets were made from those samples with 1 to 2 $\mu\text{g}/\text{cm}^2$ Ra^{226} on them.

B. Experimental Apparatus

All the Th^{226} recoil-atom range measurements were made using a recoil target assembly²³ which was adapted for these specific measurements. The catcher-foil holder pictured by Vandebosch²³ was removed from the target chamber. A parallel plate insert, shown in Fig. 8, was constructed with two brass plates cemented with Epon into lucite blocks. A 3/8-in.-inner-diam. aluminum ring was cemented into one lucite block to serve as a collimator for the Th^{226} recoil atoms. A lucite disc with a brass rod cemented through the center was cemented into the hole in the base of the target chamber to serve as the contact for the negative

ZN-2279

Fig. 8. Recoil target assembly adapted for range measurements of Th^{226} recoil atoms: (a) recoil target assembly, (b) parallel-plate insert, (c) aluminum collector strip, (d) target-chamber cover.

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electrode. Copper tubing, through which the stopping gas was introduced, was attached to the brass pipe protruding from the target-chamber cover. A copper tubing coil inserted into a Dewar flask of liquid nitrogen between the block and a mechanical pump served as a trap for any Rn^{222} which emanated from the Ra^{226} target during bombardment. Gas pressures were read from a centimeter scale attached to a U-tube mercury manometer which was connected to the copper tubing. This gas control system and the insert for the recoil block were designed and constructed by B. G. Harvey and J. R. Morton.

C. Experimental Procedure for Range Measurements.

1. Handling of Ra^{226} Targets

It was necessary to invoke precautionary measures in handling the Ra^{226} targets in order to avoid possible contamination of personnel, the recoil target assembly, and the cyclotron bombardment room. At least one-half hour before each series of experiments, the Ra^{226} targets to be used were baked under a heat lamp to remove the emanation daughters. Spare targets were placed in a lead cave at the cyclotron.

2. Cyclotron Bombardment Procedures

All bombardments of Ra^{226} with helium ions were made with the Crocker Laboratory 60-in. cyclotron. Aluminum foil was used to degrade the helium-ion beam to the desired bombarding energy. Calculations of the helium-ion energies were made from the range-energy curves of Aron, Hoffman, and Williams.³⁵ Collimation of the beam upon the target was achieved by means of a graphite collimator with a 1/4-in. circular hole. The nonradium side of the target foil, which faced the incoming beam, was cooled by helium gas which circulated in the space between the target and aluminum degrading foils. Maximum beam currents of 0.50 microampere (μa) were used in order to avoid possible vaporization of Ra^{226} from the target foil. A helium-ion beam energy of 41.6 Mev, which is near the peak of the $\text{Ra}^{226}(\alpha, 4n)\text{Th}^{226}$ reaction, was chosen after reference to the excitation function for that reaction measured by Vandenbosch.²³

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An aluminum collector strip, 2.4 cm by 8.4 cm, marked in a manner similar to that described previously was used. After the recoil atom collector strip was placed in position on the lower parallel plate and the target chamber cover tightened down, the target chamber was evacuated by means of a mechanical pump. The chamber was flushed twice with the stopping gas before adjusting the gas pressure to the desired value. The gas pressure was adjusted so that the mean linear range of the recoil atoms in the stopping gas would be about 5.5 cm. The electrical leads from the dc power supply were attached to the block in such a manner that the parallel plate on which the collector strip rested would be negative with respect to the upper parallel plate and the chamber walls. A collecting potential of about 600 v was maintained between the parallel plates during the bombardment.

The duration of helium-ion bombardments was usually between 15 and 30 min. which was usually sufficient to produce adequate quantities of Th^{226} . Another advantage of these short runs was that the probability of pressure changes in the stopping gas was considerably lowered. The mercury levels in the manometer were closely watched for any change in gas pressure during bombardment.

At the end of bombardment the gas was pumped slowly out of the target chamber in order that any Rn^{222} that escaped from the target into the chamber would be trapped in the coil of copper tubing inserted in the liquid-nitrogen Dewar. The chamber was then let up to atmospheric pressure and the chamber cover removed. The collector strip was quickly slipped from its resting place into a container and rushed to the counting room.

D. Counting Procedure

When the aluminum collector strip reached the counting room after Th^{226} collection, it was quickly cut into segments along the marked lines and gross alpha-counted in a 52%-geometry argon-flow proportional counter. Corrections were made for the decay of the 30.9-min³⁶ Th^{226} between the time of beam shutdown and counting, and for the decay of the sample while counting.

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E. Results

The gross alpha-activity was normalized in a manner similar to that described in the section on the results of the Ra²²⁴ recoil-atom range measurements.

The differential range curves for 725-kev Th²²⁶ recoil atoms in D₂, He, and N₂ are shown in Figs. 9, 10, and 11, respectively. Table II lists the experimental and theoretical values for the mean range, R_o , and the straggling parameter, ρ .

Table II

Experimental and theoretical values for 725-kev Th ²²⁶ recoil atoms				
Gas	Experimental		Theoretical	
	R_o ($\mu\text{g}/\text{cm}^2$)	ρ	R_o ($\mu\text{g}/\text{cm}^2$)	ρ
D ₂	32 ± 1.0	0.219 ± 0.026	45.0	0.076
He	30 ± 2.2	0.255 ± 0.095	45.8	0.119
N ₂	38 ± 3.7	0.277 ± 0.027	50.0	0.191
A	40 ± 2.3	0.289 ± 0.051	64.4	0.292

MU-18163

Fig. 9. Range curve for 725-keV Th^{226} recoil atoms in D_2 .

MU-18164

Fig. 10. Range curve for 725-keV Th^{226} recoil atoms in He.

Fig. 11. Range curve for 725-keV Th^{226} recoil atoms in N_2 .

IV. DISCUSSION AND CONCLUSIONS OF RANGE RESULTS

A. 96.8-kev Ra²²⁴ Recoil Atoms

It is necessary to consider the magnitude of contributions to the range straggling of effects other than collisions of the recoil atoms with the stopping medium.

The fact that the source nuclide, Th²²⁸, is not monoenergetic is of some significance. The most prominent alpha-particle energies are 5.421 Mev (72%) and 5.338 Mev (28%).^{37,38} The energy difference is only 83 kev; hence, two peaks will not be distinguishable in the differential range curves. Since only about 30% of the recoil atoms are generated by the lower-energy alpha particles, the contribution to the range straggling from this source is less than 0.005 in ρ .

It is difficult to determine the magnitude of the contribution of source thickness to the range straggling with the type of sources that were used in this work. One can compare the results obtained with two different sources to determine if there is a significant difference. Collections of Ra²²⁴ recoils were made in H₂ by the use of two different sources that were prepared and treated in the same manner. The range and range-straggling results of these experiments agreed within 2%.

Thorium-228 was collected only on platinum. No other material was used to collect the Th²²⁸ recoils. Since Th²²⁸ recoils, collected as they were at atmospheric pressure, do not penetrate the metal surface, I believe that the surface effects on recoiling atoms are negligible. The magnitude of chemical bonds is in the region of several ev. Metallic bonds are of the same order of magnitude. A comparison of this energy with the recoil energy of Ra²²⁴ shows that this is insignificant. The diffusion of one heavy metal into another is an extremely slow process even at highly elevated temperatures. The effect of diffusion of the Th²²⁸ into the platinum at room temperature during the period of time that the range experiments were conducted is considered to be negligible.

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The back-scattering of the heavy Ra^{224} recoils in platinum is not considered to be a significant surface effect on the recoils.

It is not possible to solve the problem of electric deflection of the recoil atoms rigorously because the charge of the atom and its velocity are not constant over its entire path. Consider a recoil atom of charge e and mass m moving in a uniform electric field of strength X . The atom will experience a force in the direction of X equal in magnitude $F_z = Xe$. Acceleration of such a charge is $\frac{F_z}{m} = \frac{Xe}{m} = a$. Suppose the atom passes for a distance d between the plates where X exists. It is accelerated for a time, $t = \frac{d}{v}$, perpendicular to its initial direction. Then we have

$$z = \frac{a}{2} t^2 \quad (5)$$

and

$$z = \frac{Xe}{2m} \frac{d^2}{v^2} \quad (6)$$

Field strengths of 50 to 125 v/cm were used in recoil collection. Recoils have initial velocities of $\sim 10^7$ cm/sec. We see that the displacement in the direction of the field becomes serious only near the end of the recoil atom's range, but, since the recoil has little range left to travel where the deflection becomes prominent, the effect can be considered small. Radium-224 recoils were collected in H_2 at 50 v/cm and 100 v/cm to determine whether there was a noticeable effect on range straggling produced by the field strength. No change was noted.

The drift velocity, $\vec{w}$, of an ion in a uniform electric field is represented by equation,³⁹

$$\vec{w} = \frac{De\vec{E}}{ekT}, \quad (7)$$

where D is the diffusion coefficient, e , the ionic charge, $\vec{E}$, the electric field strength, ϵ , the average agitation energy, k , the Boltzman constant, and T , the absolute temperature. The lateral drift of an ion in the uniform electric field may then be calculated with the aid of the relation

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$$l(t) = (2 D t)^{1/2}, \quad (8)$$

where the length, l , represents the root-mean-square distance of spread for ions at the time t . If an ion travels a distance, d , in the direction of the field, it will take a time of $t = d/\vec{w}$. Upon substitution of $d/\vec{w}$ and the value of $\vec{w}$ from Eq. (7) into Eq. (8), one sees that D cancels, giving

$$l = \sqrt{\frac{2d\epsilon kT}{Ee}}. \quad (9)$$

For Ra²²⁴ collection, we have $\vec{E} = 100$ v/cm, and $d = 2$ cm. This value of ϵ is close to 1 for positive ions,³⁹ and $\frac{kT}{e}$ is 2.5×10^{-2} v. With the aid of Eq. (9), we find the r.m.s. distance of spread to be 3.2×10^{-2} cm. This produces a negligible error when compared to other errors in the experiment.

Rutherford,⁴⁰ Franck,⁴¹ and Briggs^{42,43} agree that recoil atoms have the same values for drift velocities as the positive ions of the stopping gas. For H₂, the mobility of the positive ion is 5 cm²/v-sec at atmospheric pressure.⁴⁴ With a field of 100 v/cm at a pressure of 3.7 mm Hg, we have $\vec{w} = 1.03 \times 10^5$ cm/sec and $D = 24.6$ cm²/sec. Then, $l = 3.1 \times 10^{-2}$ cm is in agreement with the value of l obtained above. For the case of stopping the more energetic Th²²⁶, the lateral drift will be even less, since the gas pressures used were much higher, and it is well known that D is proportional to $1/p$.

An angle of 8.2 deg is subtended by the collimator for Ra²²⁴ recoils. The range straggling produced by collimation is less than 0.01 in ρ . It was thought that an accumulation of high charge on the glass unit supporting the source and parallel plates may have contributed to the range straggling observed. The glass unit and the inside of the bell jar were coated with Aquadag⁴⁵ to make them conducting. A Ra²²⁴ collection was done in H₂, and the range straggling result agreed with the straggling observed in H₂ without the Aquadag coating on the glass.

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It was not possible to vary the stopping gas pressure radically and still keep Ra^{224} activity on the collector strip, but it was observed that the range straggling was unchanged by an approximately 40% change in gas pressure.

If one keeps in mind that the Lindhard-Scharff equation was derived from first principles and uses only fundamental constants in its final form, the agreement between experimental and theoretical ranges can be considered as good. A comparison of the experimental and theoretical values of the mean range, R_0 , (Table I) indicates that in the energy region of ~100-kev the experimentally measured ranges are greater than theoretically predicted by the Lindhard-Scharff equation. Baulch and Duncan¹³ obtained values of $5 \mu\text{g}/\text{cm}^2$ and $10.5 \mu\text{g}/\text{cm}^2$ for the range of $\text{ThC}(\text{Tl}^{208})$ in H_2 and A, respectively. These are larger than the theoretical ranges, also.

The range straggling measured for stopping in H_2 is nearly three times as great as the theoretical value. No explanation can be found for this at present. The stragglings observed for stopping in all the gases employed in this work are greater than the theoretical values calculated.

B. 725-kev Th^{226} Recoil Atoms

The Th^{226} recoils were produced at the fringe of the magnetic field of the cyclotron. As with the deflection of the recoil atoms in the electric field, we find that we cannot solve the problem for magnetic deflection of the recoils exactly because of the constant variation of the radius of curvature of the recoil atom with its velocity. The radius of curvature may be represented by

$$R = \sqrt{\frac{2mE}{H^2 e^2}} \quad (10)$$

If we consider all quantities on the right side except the recoil energy, E , to be constant, we have

$$R = K(E)^{1/2}, \quad (11)$$

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where $K = \sqrt{\frac{2m}{H^2 e^2}}$. We see, then, that the recoil atom will undergo the greatest deflection when it possesses the greatest energy. In the fringe of the magnetic field where the recoils were generated and collected, the effect of straggling produced by magnetic deflection of the recoil atoms is not important.

Calculations by Harvey⁴⁶ show that the large range straggling observed for the Th^{226} recoils can be accounted for, in part, by the emission of the four neutrons from the excited compound-nucleus. Neutron emission from a compound-nucleus is expected to be isotropic²² and will broaden the differential range curve but will not change R_0 . If we assume isotropic distribution of the neutrons, the momentum imparted to the nucleus by the four neutrons can be represented by

$$P_{4n} = (\sum_1^4 p_n^2)^{1/2}, \quad (12)$$

where p_n is the average neutron momentum, randomly oriented. The probability that it has an angle θ to the beam is $W_\theta = \sin \theta$. If it does have an angle θ , the resultant momentum is

$$P = (p_\alpha^2 + p_{4n}^2 + 2p_\alpha p_{4n} \cos \theta)^{1/2} \quad (13)$$

at an angle

$$\omega = \arctan \frac{p_{4n} \sin \theta}{p_\alpha + p_{4n} \cos \theta}, \quad (14)$$

where p_α is the momentum imparted to the nucleus by the He ion. The projection P_b of P on the beam axis is

$$P_b = P \cos \omega. \quad (15)$$

The kinetic energy E_b associated with the momentum P_b is

$$E_b = \frac{P_b^2}{2m}, \quad (16)$$

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where m = the mass of the recoil atom. Therefore, to obtain the range (or energy) distribution, one plots E_b versus $\sin \theta$.

The Q for the $\text{Ra}^{226}(\alpha, 4n)\text{Th}^{226}$ reaction is 30 Mev. If we assume that at an incident He ion energy of 41.6 Mev, there is residual excitation in the nucleus of 5 Mev, then, there is 6.6 Mev of kinetic energy available for 4 neutrons. Making the assumption that the kinetic energy is distributed evenly among the 4 neutrons, each neutron has 1.65 Mev of kinetic energy. We then choose values of $\sin \theta$ between 0 and 1 and calculate values of E_b . The plot of $\sin \theta$ versus E_b yielded a distribution curve with a full width at half-maximum of 69%. This is a reasonable order-of-magnitude agreement with the observed range straggling for Th^{226} . More accurate values would necessitate the programming of Monte Carlo calculations on an electronic computer.

Values of the mean range, R_0 , measured experimentally for 725-kev Th^{226} in different stopping gases are less than the theoretical values calculated. Experimental ranges do agree with the theoretical values to ~25%. These results, together with those measured for Ra^{224} , seem to indicate that the range-energy relation for stopping by nuclear collisions is not linear as is predicted theoretically by Bohr.

The range stragglings measured for both Ra^{224} and Th^{226} are greater than the theoretical stragglings. In the case of Th^{226} the additional contribution to the range straggling by neutron emission from the recoil atom is present. The range distributions about R_0 appear to be Gaussian.

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V. HEAVY RECOIL ATOMS PRODUCED BY H^2 IONS ON Bi^{209} A. Preparation of Bi^{209} Targets

Reagent-grade bismuth metal⁴⁷ was vaporized in vacuo from an electrically heated tantalum filament on to a 0.001-in thick aluminum foil. The foil was attached to the inside top of a bell jar so that its distance from the filament to all parts of the foil was nearly constant. Foils of different bismuth thickness were produced by varying the length of time the filament was heated.

The bismuth thickness was determined analytically. Accurately measured areas were cut from the bismuth-coated aluminum foil. The bismuth was dissolved by placing a few drops of concentrated HNO_3 on the foil sample. Upon dissolution of the bismuth, the solution was carefully transferred into a 5-ml volumetric flask. The transfer pipette was washed several times with distilled water. Each time the wash solution was added to the flask. After this, 2.0 ml of 10% thiourea solution was added to the volumetric flask to produce the yellow bismuth-thiourea complex.⁴⁸ The solution was then diluted to volume. A Beckman model B spectrophotometer set at 440 m μ (minimum transmission for bismuth-thiourea complex) was used to measure the transmission of the solution. The concentration of bismuth was read from a curve of percent transmission as a function of bismuth concentration. From the concentration of the solution and the known area of the sample foil, the thickness of bismuth in micrograms per square centimeter was calculated. The analysis showed that for each foil the bismuth thickness was very uniform over the entire surface. Aluminum foils, with bismuth thicknesses varying from 0.6 to 42.5 $\mu g/cm^2$, were prepared for target material to study the effect of target thickness on the efficiency with which recoil atoms escape from the target.

B. Experimental Procedure

The deuteron beam of the Crocker Laboratory 60-in. cyclotron was used for all the Bi^{209} bombardments. The bismuth target was inserted

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into the recoil target assembly with the bare side of the aluminum facing the incoming beam. Helium gas circulating in the space between the target and degrader foils served to cool both target and degrader foils. The incident deuteron beam was collimated by a graphite disc with a 1/4-in.-diam hole. The target chamber was evacuated to a pressure of ~50 microns during the bombardment. The recoil atoms escaping from the target were stopped in a 0.00025-in.-thick aluminum catcher foil. The catcher foil was 4.5 cm square. It was supported in the catcher-foil holder (shown by Vandebosch²³) at a distance of 0.7 cm from the target. Maximum beam currents of 0.50 μ a were used to avoid vaporization of the bismuth target material. The deuteron beam passed through the catcher foil and stopped in an insulated Faraday cup at the back end of the target chamber. An electrometer measured the beam current at the Faraday cup.

C. Counting Procedure

After each bombardment, the target was cut out of the target holder, and a 1-in.-diam. sample was cut from the center of the catcher foil with a steel cutter. The circular segment was cut to produce a sample of convenient size for the counting chambers. Gross alpha counting showed that in every case all the alpha activity was on the circular segment of the catcher foil.

Alpha pulse-height analysis was used to identify the $\text{Po}^{208}(2.93 \text{ y}, \alpha)$,⁴⁹ $\text{Po}^{209}(103 \text{ y}, \alpha)$,⁵⁰ and $\text{Po}^{210}(138.4 \text{ d}, \alpha)$ ⁵¹ present in the target and catcher and to resolve their alpha activities. Pulse-height analysis had to be delayed several days in many instances because of the high beta-gamma activity produced in the aluminum foil of the target and catcher. It was necessary to wait until most of the beta-gamma activity had disappeared because poor resolution in the pulse-height analysis was produced by large quantities of beta-gamma activity. Polonium-210 was produced by the (d,n) reaction and by decay of the short-lived isomer of $\text{Bi}^{209}(5.01 \text{ d}, \beta^-)$.⁵² The target and catcher foil were counted on the pulse-height analyzer and proportional counter as soon as possible after bombardment and again after most of the Bi^{210} had decayed.

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D. Treatment of Data

A measure of the fraction of the recoil atoms that escaped from the target for each nuclear reaction was desired in order to compare the results for reactions proceeding by different reaction mechanisms. Percent recoil efficiency is a measure of this fraction. It is defined by the expression

$$\frac{c/m \text{ in catcher}}{c/m \text{ in target} + c/m \text{ in catcher}} \times 100. \quad (17)$$

No decay corrections were applied to the Po^{208} and Po^{209} alpha activities resolved from the counting and pulse-analysis data. The recoil efficiencies of the alpha-emitting recoil atoms from the (d,2n) and (d,3n) reactions were calculated by means of Eq. (17). The solution of growth and decay equations yielded the activities of Bi^{210} and Po^{210} present in the target and catcher at the end of bombardment. These results were then used to calculate the percent recoil efficiency for the (d,n) and (d,p) reaction products.

E. Results

Table III lists the recoil efficiencies calculated for recoil atoms from the various nuclear reactions studied, together with the target thicknesses and incident deuteron energies used. The maximum deuteron energy attainable with the experimental apparatus used was 23.6 Mev. Because of the long half life of Po^{209} , it was possible to determine the recoil efficiencies for the (d,2n) reaction only near the peak of the excitation function. The large limits of error for the Po^{209} recoil efficiencies are the results of poor counting statistics.

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Table III

Variation in percent recoil efficiencies for different reaction products and for various target thicknesses and incident deuteron energies					
Deuteron energy (Mev)	Bi target thickness ($\mu\text{g}/\text{cm}^2$)	Percent recoil efficiency			
		(d,n)Po ²¹⁰	(d,2n)Po ²⁰⁹	(d,3n)Po ²⁰⁸	(d,p)Bi ²¹⁰
15.0	0.6	91.0 $\pm$ 12.8	91.5 $\pm$ 15.4	97.4 $\pm$ 2.3	98.6 $\pm$ 12.6
	3.4	80.2 $\pm$ 9.7		96.6 $\pm$ 2.6	94.5 $\pm$ 5.1
	7.5	83.0 $\pm$ 21.3	92.8 $\pm$ 13.9	95.7 $\pm$ 1.4	89.8 $\pm$ 12.6
	14.0	54.0 $\pm$ 25.6	79.0 $\pm$ 16.9	82.8 $\pm$ 1.8	77.1 $\pm$ 7.9
	23.8	52.5 $\pm$ 15.4	64.3 $\pm$ 14.8	70.2 $\pm$ 1.7	71.0 $\pm$ 6.5
	42.5	10.2 $\pm$ 26.3	55.0 $\pm$ 12.2	55.4 $\pm$ 1.9	45.3 $\pm$ 9.3
17.2	0.6	92.7 $\pm$ 5.4		99.2 $\pm$ 1.8	96.7 $\pm$ 7.1
	3.4			98.4 $\pm$ 2.4	
	7.5	85.8 $\pm$ 5.3		96.0 $\pm$ 2.1	87.8 $\pm$ 4.0
	14.0	76.5 $\pm$ 13.2		93.0 $\pm$ 3.1	88.7 $\pm$ 4.5
	23.8	48.3 $\pm$ 28.0		80.1 $\pm$ 2.1	67.4 $\pm$ 12.5
	42.5	27.3 $\pm$ 9.1		59.1 $\pm$ 2.0	46.2 $\pm$ 5.6
23.6	0.6			98.8 $\pm$ 2.1	
	3.4	83.0 $\pm$ 10.5		98.7 $\pm$ 2.2	96.2 $\pm$ 8.5
	7.5	79.2 $\pm$ 12.3		97.7 $\pm$ 1.3	93.4 $\pm$ 9.2
	14.0	64.6 $\pm$ 6.5		93.0 $\pm$ 1.4	78.9 $\pm$ 7.2
	23.8	51.3 $\pm$ 9.8		86.4 $\pm$ 2.0	63.9 $\pm$ 5.5
	42.5	25.1 $\pm$ 13.4		68.6 $\pm$ 3.1	48.3 $\pm$ 9.3

VI. DISCUSSION OF RESULTS FOR THE SYSTEM $\text{Bi}^{209} + \text{H}^2$

Donovan²² has measured the angular distributions of heavy recoil atoms produced by deuterons on Bi^{209} . He found that the angular distribution of Po^{208} determined experimentally could be reproduced theoretically by Monte Carlo calculations, on the assumption that the (d,3n) reaction proceeded by a compound-nucleus mechanism with isotropic emission of neutrons. If a reaction proceeds by compound-nucleus formation, the entire momentum of the incident particle is transferred to the struck nucleus. By conservation of momentum, one can calculate the recoil energy of the heavy recoil. Referring to Table III, we see that the fraction of Po^{208} recoil atoms escaping from a target of given thickness does decrease with a decrease in incident deuteron energy as would be expected. The change in recoil efficiency with energy at constant target thickness does not appear to be a rapidly varying function of the deuteron energy over the range of incident energies studied. It is possible to derive mean ranges for recoil atoms stopping in Bi^{209} from the recoil efficiency data provided the target thickness is much greater than the range straggling.²⁶ The mean range, R_0 , for a recoil atom stopping in a heavy material is calculated by multiplying the target thickness by the recoil efficiency. Mean range values for Po^{208} in Bi^{209} and the Po^{208} recoil energies are tabulated in Table IV. These results are for bismuth targets of $42.5 \mu\text{g}/\text{cm}^2$. These mean ranges correspond to what one would expect for complete momentum transfer to the struck nucleus.

Table IV	
Range-energy results for Po^{208} recoil atoms stopping in Bi^{209}	
Recoil energy (kev)	R_0 ($\mu\text{g}/\text{cm}^2$)
140.2	23.5
160.7	25.0
220.4	29.2

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There is little one can conclude about the (d,2n) reaction mechanism, since the recoil efficiencies for Po^{209} were measured at only one energy.

In some instances large errors in the recoil-efficiency results for Po^{210} and Bi^{210} were produced because the pulse-height analysis could not be carried out soon after the end of bombardment. The (d,n) and (d,p) reactions must be proceeding by mechanisms other than compound-nucleus mechanisms, since the recoil efficiencies of Po^{210} and Bi^{210} are less than those for Po^{208} for all deuteron energies and target thicknesses with the exception of Bi^{210} for a $23.8 \mu\text{g}/\text{cm}^2$ target at 15.0 Mev. If the (d,p) and (d,n) products were produced by pure stripping reactions, one would expect nearly the same recoil efficiencies for Po^{210} and Bi^{210} , since the neutron or proton penetrating the nucleus has to go in with little kinetic energy. If the penetrating particle goes in with too high kinetic energy, its binding energy is exceeded, and it will be re-emitted. If enough energy is transferred to the nucleus, other reactions will be favored over (d,p) and (d,n) reactions. In all cases it was found that the recoil efficiencies of Po^{210} are less than those of Bi^{210} . From Donovan's angular-distribution results it can be seen that the (d,n) products are peaked more in the forward direction than are the (d,p) products. This would lead to higher recoil efficiencies for Po^{210} than for Bi^{210} . Since Donovan did not measure the angular distributions for these products above a deuteron energy of 12.5 Mev, no attempt is made to resolve the different types of results.

I have confidence that the recoil-efficiency results for Po^{210} and Bi^{210} are reliable because:

- (a) The results calculated for the (d,n) and (d,p) reactions came from the same counting data used for the (d,3n) product.
- (b) The range results calculated for Po^{208} , when plotted on a range versus energy curve, fell in line with results determined by Alexander and Winsberg⁵³ and by Donovan⁵⁴ for other heavy recoil atoms produced in thick targets.
- (c) It was possible to calculate cross-sections for the (d,n) and (d,p) reactions from the counting data that agreed well with cross-sections for these reactions measured by Kelly and Segre.²⁵

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Since the limits of error in the recoil efficiencies for Po^{210} and Bi^{210} do overlap, it may be that the true recoil efficiencies lie somewhere between the values determined for each of them.

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