Radioactivity of Neodymium and Samarium

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Descriptions of methods for determining (*a*) absolute activity of an element, (*b*) the penetrating power of its radiation, (*c*) the deflectability in a magnetic field and (*d*) the sign of the charge of the radiation are given. The methods involve the use of Geiger-Müller counters with screen walls. Neodymium emits beta-rays of maximum $H\rho$ value 355 and 2.4 mm of air penetrating power. The half-life is slightly less than 1.5×10^{12} years. The range of the samarium alpha-particle has been found to be 1.23 ± 0.05 cm in air at N.T.P. and its half-life 6.3×10^{11} years. Gadolinium, praseodymium, tin, iodine and beryllium have been found inactive. They cannot have half-lives less than 10^{14} years if they emit particles of ranges greater than 1 cm. The limit for Be is 10^{15} years.

THE problem of the detection of radioactivity among the lighter elements has become of greater importance during the last few years because of our increasing knowledge of nuclear properties. In this investigation several elements have been examined and the activities of two of them substantiated. As Hevesy, Pahl and Hosemann[1] have stated, the general average stability of the elements does not rule out the existence of relatively active scarce isotopes. The methods of detection are so sensitive that the presence of so small an amount of radium as 10^{-12} parts per unit weight of sample is easily established. However, it is not necessarily true that such activities be attributed to less abundant isotopes. Activities of the abundant isotopes sufficiently low to prevent detection by this procedure can be established by counter methods in general and in particular by the one used in this research.

Description of Apparatus

Detection of feeble activity obviously cannot be better accomplished than by the use of an instrument capable of counting single particles emitted from a sample of large area. As the closest approach to this at present available, Geiger-Müller counters modified by the use of wide-mesh screen walls to allow free entrance of particles of the lowest possible energies have been used throughout this work.

Since there are so many ionizing particles being produced under normal conditions by cosmic rays and small amounts of strongly active substances, the presence of any radiation of the kind sought can obviously be established only by a differential method which will allow an accurate correction to be made for this background radiation.

The apparatus used for the general detection and measurement of activities is shown in Fig. 1a.

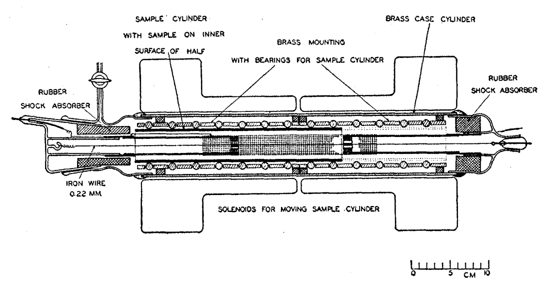

FIG. 1a. Apparatus used for general detection and measurement of activities.

A screen wall counter is placed in a chamber in such a way that a sample deposited on the inner surface of half a steel cylinder concentric with the counter can be brought directly over the counter and removed by moving the sample cylinder through half its length. The steel cylinder was mounted in a sleeve of brass bearings and the whole chamber placed inside of two solenoids capable of giving fields sufficiently intense to move the cylinder. The solenoids were used alternately to move the cylinder from the exposed to the unexposed position at intervals of five minutes. The timing mechanism consisted of a half-minute telechron clock from which a twenty-notch ratchet wheel device operated to close the

[1] G. v. Hevesy, M. Pahl and R. Hosemann, Zeits. f. Physik **83**, 43 (1933).

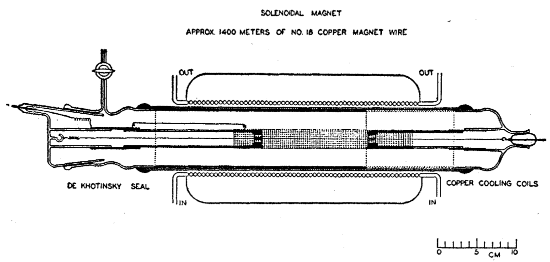

FIG. 1b. Apparatus used for determination of magnetic deflectabilities.

magnet circuits. The whole counter was placed in an iron shield as shown in Fig. 2. The shield affords a protection of at least a foot of iron in every direction and reduces the background count from approximately 25 per half-minute when unshielded to about 15.

The chamber was generally filled with oxygen, air, hydrogen, argon or helium at 2 or 3 cm of Hg pressure. The voltage necessary for counting ranged between 600 and 1500 under these conditions. The sample cylinder and chamber wall were usually kept at a potential about 45 volts positive with respect to the counter to stabilize the counts and render the effect of leakage currents less important.

By placing the sample cylinder at a sufficiently high negative bias with respect to the counter wall it is possible to increase the effective diameter of the counter to the size of the sample cylinder and effectively place the sample inside the counter.

The amplifying circuit is represented diagrammatically in Fig. 3. Essentially, the count phenomenon in the counter produces a fall in potential of the wire which lasts a time determined by the value of the shunt resistance to the ground represented in the figure as a three-element tube with filament and grid connected together to the counter wire and the plate to the ground. It seems essential that the value of this resistance be adjusted to certain values between 10^8 and 10^{10} ohms, somewhat dependent upon the counter being used. This has been mentioned in an earlier publication[2] in which the use of solution resistance units was advocated. The tube type of leak is more satisfactory because of its ready adjustability. The current heating the filament is varied by a sensitive rheostat. A milliammeter in the filament circuit insures reproducibility. Kovarik[3] first pointed out this general type of circuit for point counters. The tube resistance device is described by Rutherford, Chadwick and Ellis.[4]

The output in the plate circuit of the screen-grid tube has been used alternatively to operate a telegraph relay, a two-tube thyratron relay or

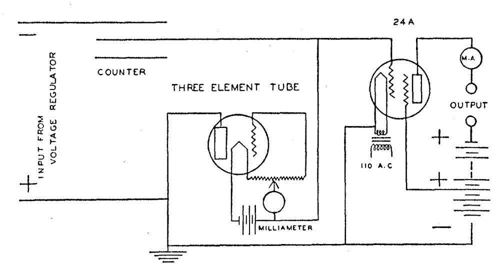

FIG. 3. Recording circuit.

coupled with a two-tube resistance coupled amplifier which gave output impulses of 10–20 m.a. to actuate the telephone relay. The latter was the most satisfactory.

An ordinary magnetic counter operated from the telephone relay to record the counts. A magnetically controlled automatic camera built to use standard 35 mm motion picture film was focussed to take pictures of the dial readings at intervals of 30 seconds. A telechron clock controlled the camera, as well as the sample-moving mechanism described above.

The whole assembly for activity determination functioned automatically, taking readings

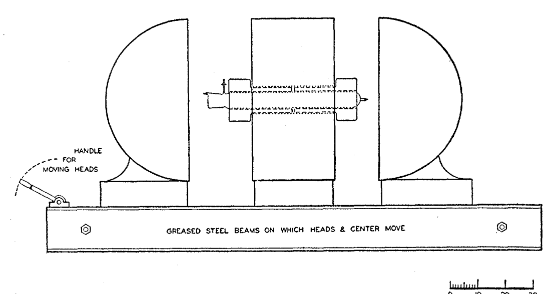

FIG. 2. Complete counter and shield assembly.

[2] Libby, Phys. Rev. **42**, 440 (1932).
[3] Kovarik, Phys. Rev. **13**, 272 (1919).
[4] Rutherford, Chadwick and Ellis, *Radiations from Radioactive Substances*, pp. 52–53, The Macmillan Co., 1930.

every half-minute with sample being moved either over or away from counter every five minutes for as long an interval as was necessary to establish definitely the value of the activity. An illustrative calculation of the time necessary for an effect of given size is given later.

Fig. 1b shows the apparatus used for the determination of magnetic deflectabilities. The chamber has a heavily wound water-cooled solenoid around the central portion where the counter is. The sample cylinder is enlarged to the size of the chamber wall. The magnet produces fields up to 900 gauss over the counter region. Since the distance from the sample to the counter is 1.7 cm, the radius of the circle of motion of the particle just missing the counter under the most advantageous conditions of energy and direction of emission must be 0.85 cm and the $H\rho$ value of this limiting particle is 0.85 of the field in gauss for this condition. Consequently the determination of the limiting $H\rho$ values for a radiation consists of plotting the curve of activity against field strength and noting the lowest field just capable of annihilating the effect. This type of curve is illustrated by Fig. 7 for Nd_2O_3 beta-radiation. The magnet is cooled by a layer of copper tubing wound around the chamber in bifilar manner so as to keep the chamber temperature uniform. Movement of the sample cylinder is accomplished by tipping the chamber and the use of the shield is foregone. Needless to say, this particular apparatus will serve for relatively low energy radiations only but can be modified by use of a larger chamber for higher energy particles. D. Bocciarelli and G. Occhialini[5] first suggested and used this geometrical arrangement in their work on potassium. The cooling coil next to the chamber is necessary to prevent a gradual rise in the counting voltage due to heating of the chamber.

The instrument used to determine the sign of the charge of a magnetically deflectable particle consists of the deflection instrument itself modified by the insertion of sloping vanes between sample and counter, as shown in Fig. 4. Four sets of readings are taken, two of them with sample over the counter and the field direction reversed automatically at regular intervals by an adaptation of the ratchet wheel mechanism mentioned

[5] D. Bocciarelli, Nature **128**, 375 (1931).

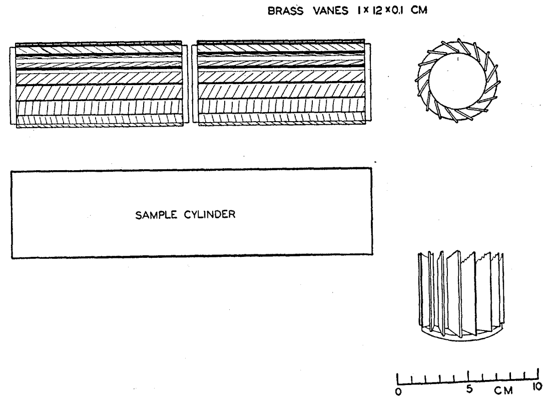

FIG. 4. Vanes for insertion in deflection apparatus to allow charge determination.

above; and the other two consisting of the readings with sample removed. The instrument was calibrated both by calculation of field directions and the effect with potassium radiation. The particles bending up from the sample when starting originally parallel to the vanes will obviously reach the counter more frequently than those curving oppositely.

The ranges of radiations in various gases are measured in the apparatus shown in Fig. 5. Two counters of approximately the same construction are placed in a large chamber with the sample deposited on the chamber wall over one of them. Two curves of count rate against pressure of gas are then run. The range of the radiation is determined from the lowest pressure at which the two curves start paralleling each other. The difference between the two count rates can be traced to the point at which it assumed a constant value. Curves of this kind are shown in Fig. 8 for

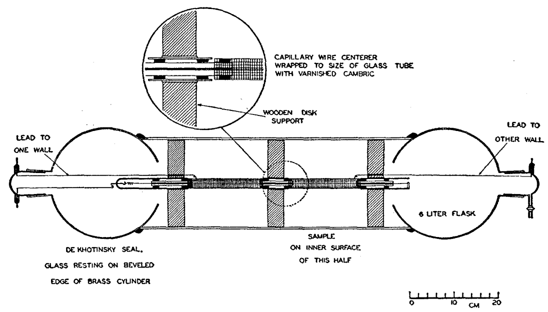

FIG. 5. Apparatus for determining ranges of α-particles in various gases.

samarium alpha-radiation. For the instrument shown the sample counter distance is 8.7 cm so a particle of 1 cm range should cease to be effective at a pressure of 8.7 cm of air, i.e., 76/87.

Characteristics of Counters

The general properties of screenwall counters are these: (1) In general, the properties of the counters agree with those of the solid wall type studied and reported upon so extensively by many workers.[6] (2) Each counter when in proper counting condition has a voltage range of from 10 to 100 volts within which the applied voltage can be varied without changing the count rate. However, the size of the "kicks" varies with the voltage in this interval and the amplification must be sufficient to prevent variation of kick size causing failure of mechanical counting. (3) Counters have been made with copper, aluminum and steel walls combined with wires of tungsten and of the above materials in the various possible ways. In none of these cases has it proved essential that the materials be cleaned or treated in any way other than to remove moisture. (4) The voltage-pressure curves are very dependent on the character of the gas. The noble gases apparently give flat portions at low pressures in the curves of voltage against pressure. (5) Very pure helium is not a good gas to use. It is difficult to get the voltage adjusted correctly, and the permissible voltage range appears to be smaller than for the less pure gas. (6) Counter heads should have high resistances to prevent spurious counts due to leakage currents from wall to wire. They should center the wire accurately to insure the maximum size of the counting voltage range. (7) The counters are often photoelectrically sensitive, especially if aluminum is present. Copper and iron both show the effect under the proper conditions.

Photoelectric Effect and Sensitivity of the Counters

Necessarily, the question of the sensitivity of the counters to particles of the lowest energy must be considered because of its bearing on the significance of negative results obtained from certain elements. Certainly one cannot say with assurance that failure to observe radiations of the more common ionizing powers indicates inactivity of any sort, especially in view of the low energy radiations from samarium and neodymium discussed below.

Fortunately, the photoelectric effect serves to establish the requisite sensitivity. Experimentally, the effect is most obvious with counters having aluminum screen walls. In fact, as many as 100 photoelectrons per minute have been counted from a wall area of 2 or 3 cm^2 with a 150-watt tungsten lamp at 5 or 6 feet. The chamber walls of 2 mm Pyrex glass as well as the bulb glass intervened. The phenomenon must be associated with the screen wall rather than the wire of the counter, for changing from aluminum to copper screen of the same mesh and wire size changes none of the counter characteristics except the response to light. Copper and iron emit photoelectrons in appreciable number when illuminated by direct sunlight. Most screen aluminum shows the effect but certain samples were found which did not until they had been cleaned with acid.

Since the Pyrex glass used is known to pass no light below 3000A and the photoelectric thresholds for iron and copper are given values in the I. C. T. between 2665–3050A for mechanically cleaned surfaces, it becomes clear that the instrument must be capable of counting electrons of very low energy. As a further, more quantitative test, the photoelectric threshold of the aluminum screen was determined by means of a monochromator with a tungsten lamp source. The procedure was to build a counter about the length of the monochromator slit, shield it from all light except that coming from the slit and observe the setting of the monochromator when the count rate became equal to the zero rate. A value of 5000A was found. The highest value given in the I. C. T. is 4770 and the average of all values given is 3900.

At first thought one might conclude that these results are of little significance as to the ability of the counter to detect low energy electrons because the voltage applied to the counter constitutes an accelerating potential of about (but not exceeding) 2000 volts per cm. This objection is answered rather conclusively, however, by the

[6] F. Burger-Scheiflin, Ann. d. Physik (5) **12**, 283 (1932); W. Schulze, Zeits. f. Physik **78**, 92 (1932); C. Bosch, Ann. d. Physik **19**, 65 (1934).

work of Lawrence and Linford[7] on the effect of high electrostatic fields on the photoelectric threshold of alkali metal surfaces. They checked the Schottky formula for the effect rather well and showed the effect for these surfaces to be thoroughly predictable. As an example, they found a field of 69,000 volts per cm to shift the limit for a potassium film deposited on tungsten from 5600 to 5900A. Assuming that the aluminum surface is not greatly different from potassium as far as this effect is concerned and with the formula they verified, it appears that a field of the order of that used in the counters should shift the threshold by about 30A providing the true threshold in the absence of a field is 4800A. These figures indicate that electrons are counted almost down to the threshold where they emerge from the metal with practically zero energy. At least they are counted when they have energies of 1 volt or so, since 0.05 volt corresponds to a shift of about 100A in the region of 4800A and these considerations have shown that the threshold as determined by the counter method is most probably within 100A of the true value. Consequently, the instrument must be capable of detecting any beta-ray that might be emitted from a radioactive body.

Furthermore, since alpha-rays necessarily have higher ionizing powers than betas, the above conclusion applies to them also. Finally, since gammas are recorded through their photoelectric secondaries they, too, must be included and the general conclusion that the method is capable of detecting any radiations of these types which can be emitted by a radioactive body is established.

Counting Efficiency

Having established the ability of the counter to respond to all possible radiations to some extent it is only necessary to demonstrate the high percentage of the eligible particles actually counted to establish the method as one of precision.

For example, the counter in the absolute activity apparatus shown in Fig. 1a, having a copper screen wall presenting 71 percent open space can be calculated by consideration of the solid angle subtended at each part of the sample by the counter and adding over the sample surface, to offer free passage to 14 percent of the particles from the sample. Since potassium is known[8] to emit 27,500 particles per half-minute per mole, the number passing through the counter must be 3850 per half-minute per mole of sample. The calibration experiments for the activity apparatus of Fig. 1a showed 3740 ± 200 counts per half-minute per mole of KCl in sample. The check seems quite satisfactory.

Statistical Distribution of Counts and Calculation of Error Limits

The question of the statistical distribution in time of the background radiation is of importance to this research only in that it indicates how the limits of error of the average results should be fixed. Bateman[9] has shown that the particles emitted from a radioactive body should be distributed in time according to a law requiring that the standard deviation from the average rate observed should be just the square root of that rate. This simple result might be expected to hold for the zero count in this work. Many data have been obtained in the course of this work which substantiate this. The details of this aspect of the work are to be submitted for publication separately. The probable errors given below have been calculated on the assumption that the average rate is sufficiently high to warrant the use of the error law relation that the probable error is 0.67 times the standard deviation.

The validity of the law allows its use to calculate the length of the runs necessary to establish definitely an effect of given size. With the activity instrument in the shield the rate without sample, zero rate, usually is about 15 per half-minute. By using this value, the length of the shortest satisfactory run to establish an effect of 1.0 count per half-minute with probable error of $\frac{1}{3}$ the effect can be calculated from the relation

$$0.6745 \times 2^{\frac{1}{2}}(15/(n/2))^{\frac{1}{2}} = 0.3$$

to have the value 300, n being the total time in half-minutes and $n/2$ the number of intervals with and without sample exposed.

[7] Lawrence and Linford, Phys. Rev. **36**, 482 (1930).

[8] W. Mühlhoff, Ann. d. Physik **399**, 205 (1931).

[9] H. Bateman, Phil. Mag. **20**, 704 (1910).

Methods of Proving Specificity of Activities Observed

Probably the most difficult phase of the problem of establishing the activity of a weakly active element is the proof that the activity observed is not due to minute amounts of known strongly active substances. In general, impurities of this sort present in such small amounts as 10^{-12} grams per gram vitiate any effects that could be expected. Three general methods of attack are used. The first consists of chemical purification of the substance investigated, observing any change in the activity resulting; the second depends upon observation of marked differences between the properties of the radiation and that of any previously known type; and the third is the checking of the growth of the activity after purification to a constant final residual activity against the growth curves for known radioactive substances.

Removal of such small amounts of impurities is most easily accomplished by the general method of co-precipitation. A considerable amount of a homologue of the impurity being removed (for example, Ba^{++} for Ra^{++}) is placed in solution with the substance being purified and a reagent added to precipitate the homologue of the impurity without taking out the substance purified. The co-precipitation methods, in general, prove adequate if the operation is repeated once or twice. The removal seems to be unexpectedly thorough, possibly because of a general covering up of the radioactive precipitate before it has opportunity to come to equilibrium with the solution and redissolve. All of the known radioactive substances which have sufficiently long lives to contaminate seriously a sample are homologues of sodium, barium, lanthanum, zirconium, tellurium, lead or the noble gases.

Obviously, it will prove difficult, in general, to purify any one element from all of the above. For example, the rare earths cannot easily be freed of traces of actinium and mesothorium. Consequently, there usually remains some doubt as to the source of any residual activity after all of the possible purifications have been carried out. This can sometimes be removed by allowing the sample to stand and by determining its activity at intervals as well as noting any changes in its characteristics such as penetrating power, deflectability, etc. Since the most probable impurities are those which have no homologues precipitated chemically, the growth curves and general changes in penetrating power, etc., to be expected from them are known and generally make the data obtained highly pertinent. Of course, it is possible that the character of the radiation after purification may obviate the growth curve step.

Results

Beta-radioactivity of neodymium[9a]

Samples of neodymium salts prepared by the late Professor James of New Hampshire and by Professor B. S. Hopkins of Illinois, as well as others of unknown history, were used. The first measurements on all of them showed a definite activity which seemed to have about the same value for all. However, thorough purification reduced the activity to a final residual value which was the same for all of the samples. All samples were changed to Nd_2O_3.

Since the rare earths cannot be readily purified of traces of actinium, the significance of this final residual activity was not clear. One sample was therefore purified as thoroughly as possible for all other possible impurities, and allowed to stand for seven months. After that time the activity was found to be unchanged within fifty percent. In addition, absorption experiments showed that 5 mm of air completely absorbs the radiation. These tests were made by placing thin screens over the sample in the activity counter, Fig. 1a, and also by varying the gas pressure in the deflection instrument, Fig. 1b, when there was no magnetic field present. For example, a pressure of 3 cm of air in the last case gave an absorbing layer of 0.6 mm. The data from one of these runs are presented in Fig. 6.

Magnetic deflection experiments were then performed. It was found possible to bend the hardest of the radiation into circles of 7.5 mm radius or less by a field of 500 gauss. Fig. 7 shows these data. The abscissae are the $H\rho$ values of particles which would just miss the counter under the most favorable conditions of emission and with the given field. The maximum $H\rho$ value

[9a] A brief report on this work has appeared previously. Libby, Phys. Rev. **45**, 845 (1934).

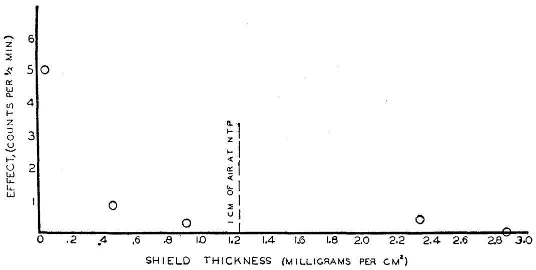

FIG. 6. Absorption curve for Nd beta-radiation.

apparently is close to 355, which agrees well with the observed range according to the data of Schonland[10] on cathode rays. This means a range of 2.4 mm in air at atmospheric pressure and 15°C.

These data seem to make the presence of any of the members of the known radioactive series in significant amounts extremely improbable. The possible presence of a rare earth impurity (such as element 61) which might emit such a radiation was checked by repetition of the deflection experiments with samples of samarium, lanthanum and praseodymium, none of which showed a deflectable radiation of intensity greater than one-third of that of neodymium. The intensity of the alpha-radiation from the samarium was about ten times that from the neodymium samples in the absence of a field.

There are two aspects of the growth curve data. The first is the relatively small change in the total activity and the second is the low penetrating power of the whole of the final radiation. Calculations for actinium itself show that there should have been over a two-fold increase in total activity but, more important, more than two-thirds of the final effect should have been due to alpha-rays, all of greater penetrating power, of course. It should be mentioned also that the whole notion of actinium itself acting as a detectable radioactive substance is improbable because of the known fact that its transition has never been detected except through its products. Calculations were also carried through for RaD, assuming that somehow it might give beta-rays of the right character. After seven months its activity should nearly have tripled in intensity and

[10] Schonland, Proc. Roy. Soc. (London) **A104**, 235 (1923); **A108**, 187 (1925).

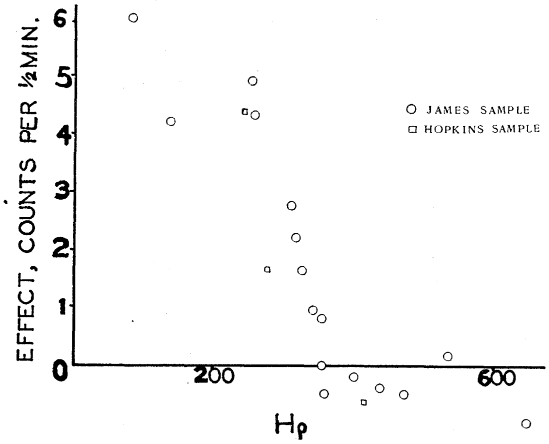

FIG. 7. Effect of magnetic field on Nd beta-radiation.

should have come to consist of 36 percent of beta-rays with $H\rho$ larger than 600 but less than 1200, 39 percent of betas with $H\rho$ larger than 1200, and 25 percent of alpha-radiation.

At this stage of the work the discovery of induced radioactivity with the emission of positrons from short-lived intermediates in transmutation processes was announced. It then seemed essential to determine whether the neodymium case was one of positron emission by definitely establishing the charge of the particles. This was done with the instrument previously described and shown in Figs. 1b and 4. Runs were made during which the sample was left in one position, either over the counter or away from it, readings were taken every half-minute, and the direction of the flow of the current was reversed every five minutes. The whole was done automatically as previously indicated. The best set of results is given in Table I.

TABLE I. *Count per half-minute.*

	Negatives pass	Positives pass	Effect
Sample present	23.37 ± 0.108	23.07 ± 0.107	0.30 ± 0.15
Sample absent	23.38 ± 0.108	23.98 ± 0.110	-0.60 ± 0.15

Each of the four values given is the mean of about 900 readings. The runs with sample present and sample absent were made on different days and the absolute values are therefore not significant. The effect, 0.90 ± 0.22, of the addition of the sample appears conclusive. The effect indicating low energy positives from the wall in the absence of the sample remains unexplained but definite. Further work will be done on this point.

Calculation of apparent half-life of neodymium

In order to calculate the specific activity it is necessary to calculate the thickness of the sample from which no emitted particles will fail to count because of self-absorption in the sample. It is assumed that this layer has a thickness just the range of the particle in the sample. Necessarily this introduces an error tending to make the effective sample assumed somewhat too large and making the rate of decay calculated too small. The data of Schonland[11] give 2.75×10^{-4} grams per cm² as the range of electrons of $H\rho$ value 355. This requires the saturated layer to have 8.18×10^{-7} moles per cm². The air interceding between sample and cylinder was 0.253×10^{-4} grams per cm², so the saturated Nd_2O_3 layer should have 7.43×10^{-7} moles per cm² or 1.435×10^{-4} moles for 193 cm².

Table II shows the results of three runs on the rate of emission of particles from the saturated layer.

TABLE II. *Counts per half-minute from 193 cm².*

46.1×10^{-7} moles/cm²	4.7 ± 0.7
90.8×10^{-7} moles/cm²	6.0 ± 0.9
Not measured, but approximately the same	5.0 ± 0.8
Average (weighted according to length of run)	5.2 ± 0.5

These data show that the effect per mole of sample is $(3.6\pm0.35)\times10^4$ counts per half-minute. A standardization run with KCl gave 3740 ± 220 for the effect from K. By assuming the neodymium betas to be just as efficient in counting as those of K because of the large openings in the screen wall and by using the data of Mühlhoff[12] on the activity of potassium, the decay constant has the value

$$\mathrm{Nd}=\frac{36{,}400\pm3500}{3740\pm220}\times1.5\times10^{-21}=(1.46\pm0.20)\times10^{-20}\mathrm{sec.}^{-1}.$$

The apparent half-life of neodymium therefore cannot exceed 1.46×10^{12} years and is probably close to half that value.

Alpha-radioactivity of samarium

A samarium sample prepared by Professor James was used throughout this work. The activity of the element was easily detected and the extraordinary characteristics of the radiation in being wholly absorbed by a shield of stopping power 1.3 cm of air made it seem specific. Notice of the excellent work of Hevesy and Pahl[13] appeared at this time, clearly establishing the radiation as belonging to samarium itself. The range and decay constant were then measured.

The instrument shown in Fig. 5 was used to measure the range. Oxygen gas was employed at temperature of 25°C. Fig. 8 shows the data for

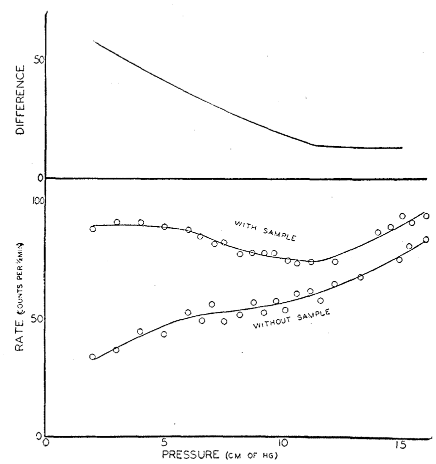

FIG. 8. Effect of pressure on count rates for counters in range finder with Sm sample.

one of the runs, the lower curve in both representing the activity of the counter without the samarium sample and the upper that with the sample. The significant pressure is, of course, that at which the difference between the two count rates becomes constant, showing that the samarium alphas are no longer counting. The curves apparently give the critical pressure at 11.2 cm of Hg. Another run gave the same value. Since the distance between the counter and the sample was 8.7 ± 0.1 cm, the equivalent distance at 76 cm at 15°C would be

$$8.7\times(11.2/76)\times(288/298)=1.236\ \mathrm{cm}$$

in oxygen which is the range value. The results of Gurney[14] on the relative stopping powers of oxygen and air for alpha-particles show that within one or two percent the average stopping

[11] Schonland, Proc. Roy. Soc. (London) **A104**, 235 (1923); **A108**, 187 (1925).
[12] Mühlhoff, Ann. d. Physik **399**, 205 (1931).
[13] Hevesy and Pahl, Nature, **130**, Dec. 3 (1932).
[14] Gurney, Proc. Roy. Soc. (London) **A107**, 340 (1925).

powers for alphas ranging uniformly from zero to 1.2 cm in range must be the same. Therefore the value for the range in air at N. T. P. found is 1.23 ± 0.05 cm, the error estimate being an upper limit.

The calculation of the range of the 1.23 cm particle in Sm_2O_3 is complicated by the deviations from the Bragg-Kleeman rule for atomic and molecular stopping powers for alpha-particles of short range. The best value is apparently close to 4.9×10^{-4} cm or 1.01×10^{-5} mole per cm^2. This figure is not of much importance to the following calculation of the half-life of samarium, because the samples used were thinner. It can therefore be assumed that the whole of the samples used have been effective. Table III presents the results of three determinations made in the activity counter of Fig. 1a.

TABLE III. *Samarium effect (counts per half-minute).*

Sample thickness	Effect for 193 cm^2	Effect per mole
0.0441×10^{-5} moles/cm^2	16.0 ± 0.9	$80,000\pm 6,000$
0.152	53.5 ± 0.2	$91,800\pm 3,400$
0.0153	5.1 ± 0.8	$89,500\pm13,000$
Weighted average		$88,900\pm 2,900$

$$Sm=1.5\times10^{-21}\times\frac{88,900}{3740}=(3.56\times0.1)\times10^{-20}\text{sec.}^{-1}.$$

The value for the half-life is $(6.3\pm0.5)\times10^{11}$ years. Both the values for the range and activity differ somewhat from those of Hevesy, Pahl and Hosemann.[15] The range value is slightly larger than the 1.13 reported by these authors, in accordance with their belief that their value might be slightly low. The discrepancy between the values for the specific activity is perhaps larger than one might have hoped. It is perhaps pertinent that the samarium samples used in this research were never purified from radioactive impurities as carefully as possible because the radiation was found to have no component capable of penetrating more than 1.5 cm of air. If such a component were present it could not possibly have constituted more than 10 percent of the total radiation. It has been observed several times that thick samples gave concordant results only when the limitation of solid angle for deep particles was taken into account as in the formula of Evans.[15a] Application of this formula to the data of Hevesy, Pahl and Hosemann gives the half-life obtained in the present work.

Other elements

Praseodymium, gadolinium, tin and iodine have all been tested for activity. It is difficult to express the limiting values for the decay constants because they depend on the ranges of the particles which might be emitted. However, it can be said with considerable certainty that none of the above elements can emit particles of range greater than 1 cm of air and at the same time have a half-life less than 10^{14} years.

Beryllium

As previously reported,[16] no activity has been found in BeO samples purified sufficiently well. Measurements were made sometime ago with the activity counter represented in Fig. 1a when the sample cylinder was maintained at a sufficiently negative potential with respect to the counter wall to place the sample effectively inside the counter with the sample cylinder as its wall. These substantiated the limit of possible activity for particle emission mentioned in the above publication. The results can be summarized in the relation

$$t_{\frac{1}{2}}>1.1\times10^{15}R,$$

in which $t_{\frac{1}{2}}$ is the half-life in years and R is the range of any emitted particle in cm of air at N. T. P. This relation is most rigorous for alpha-particles but is not greatly in error for beta-particles.

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[15] Hevesy, Pahl and Hosemann, Zeits. f. Physik **83**, 43 (1933).

[15a] Evans, Phys. Rev. **45**, 29 (1934).

[16] Libby, Phys. Rev. **44**, 512 (1933).