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Gamma-Rays from Light Elements Due to Proton Bombardment

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With the high potential electrostatic generator recently constructed at this laboratory a survey of a number of elements has been made in a search for gamma-ray emission due to proton bombardment. Six of the elements examined gave gamma-rays of sufficient intensity to permit accurate measurement. These six elements are Li, Be, B, F, Na and Al. Their gamma-ray yield was studied as a function of proton energy, or generator voltage, starting with the minimum potential at which the yield was easily measurable. In the region of fairly low potentials the six elements studied gave evidence for resonance excitation of gamma-radiation and at high potentials each of the six elements gave a gamma-ray intensity which increased nearly exponentially with voltage up to the maximum voltage obtainable from the generator (approximately 2 Mev). The measurements were made using a Lauritsen electroscope, and therefore they give the intensity in arbitrary units which depend on the hardness of the radiation and the geometry of the apparatus. The other elements which were bombarded are C, O, Si, K, Ca, Ni, Cu, Zn, Mo, Pt, Pb. Gamma-radiation from these elements was weak and the observed intensities may have been largely due to contaminants.

INTRODUCTION

THE excitation of gamma-rays from the light elements due to high energy proton bombardment has been studied at several different laboratories. Lauritsen[1–3] and his colleagues observed gamma-radiation from Li, Be, B, and F using protons with energies up to about 0.9 Mev. Using 1.2 Mev protons McMillan[4] observed gamma-radiation from Li, Be, and F and obtained indications of a weak intensity from B. The work of Hafstad, Heydenburg and Tuve[5] on the voltage excitation curves of gamma-radiation from Li and F showed the advantages of a monochromatic beam of protons and accurately measured generator voltages. Their curves of gamma-ray intensity from thick targets of Li and F as a function of proton energy show a number of well-defined steps and demonstrate the importance of nuclear energy levels in the interaction of protons with nuclei.

Lithium was shown by these observers to give a sharp resonance emission of gamma-radiation for incident protons with an energy of 440 kv. This resonance is convenient for voltage calibration and upon completion of the electrostatic generator recently developed at this laboratory this lithium gamma-ray resonance was used to determine its voltage scale. Further work on proton bombardment of lithium using voltages up to a maximum of nearly 2 Mev gave results of such interest that it seemed advisable to extend the investigation to other elements. A fairly extended survey was therefore made which

[1] Crane, Delsasso, Fowler, Lauritsen, Phys. Rev. **46**, 531 (1934).
[2] Crane, Delsasso, Fowler, Lauritsen, Phys. Rev. **47**, 782 (1935).
[3] Crane, Delsasso, Fowler, Lauritsen, Phys. Rev. **48**, 102 (1935).
[4] Edwin McMillan, Phys. Rev. **46**, 868 (1934).
[5] Hafstad, Heydenburg and Tuve, Phys. Rev. **50**, 504 (1936).

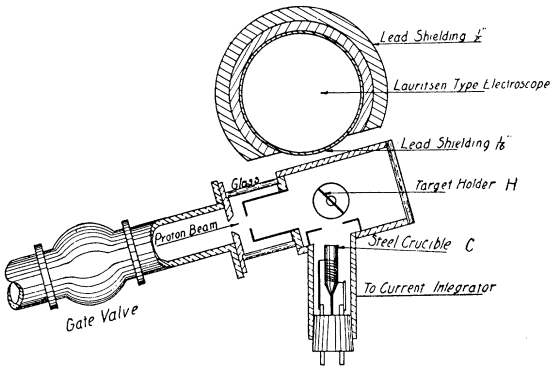

FIG. 1. Target chamber and electroscope.

included most of the light elements up to aluminum and also several heavy elements.

APPARATUS AND PROCEDURE

High energy protons for these experiments were obtained by means of a belt type electrostatic generator operating in a steel tank under high air pressure.[6] Fig. 1 shows the construction of the target chamber which was used for all of the work on gamma-ray emission. Several different targets were mounted on holder H which consisted of a sheet of nickel or molybdenum. By means of a threaded rod and a ground brass plug the holder could be moved so that the proton beam impinged on any desired target. Gamma-ray intensity was measured with a quartz fiber electroscope of the Lauritsen type placed as shown in Fig. 1 and provided with lead shielding to cut down the background intensity caused by x-rays from the generator. To obtain gamma-ray intensity from a weak emitter at any particular generator voltage, a run of several minutes was often necessary to give sufficient deflection of the electroscope fiber. As the proton current was often unsteady, the total charge incident on the target during a run was measured by means of a current integrator as shown in Fig. 2.

This circuit makes use of a thyratron tube to periodically discharge a condenser which is being charged by the proton current. As protons enter the Faraday cage in which the target is mounted, the potential of the condenser which furnishes the plate current for the thyratron rises until it reaches the critical discharge voltage associated with the bias used on the grid. The thyratron then discharges the condenser, and immediately becomes nonconducting so that the condenser may recharge. The number of discharges is recorded by a separate thyratron circuit the same in principle as the one described by Dunning.[7]

To prevent the plate condenser from completely discharging and thus causing the potential of the Faraday cage to fluctuate greatly, the condenser C_7 is put in series with a smaller condenser C_4, so that the discharge from C_7 goes into C_4. In this way the voltage of C_7 may drop only 7 volts while the voltage of C_4 changes by 35 volts and extinguishes the discharge. R_8 discharges C_4 before the thyratron becomes conducting again. This seven-volt fluctuation of the Faraday cage has not been sufficient to cause interference in an amplifier brought up near the target for alpha-particle counting.

The positive impulse for setting off the recording thyratron circuit is obtained from the rise in voltage across R_5 accompanying the discharge through the tube. Other resistances and condensers shown in the circuit are used for different sensitivities. Radio gang selector switches, S_1, S_2, S_3, and S_4, change the sensitivity by means of one control; but when the highest sensitivity is used, position 1, the grid bias must be changed

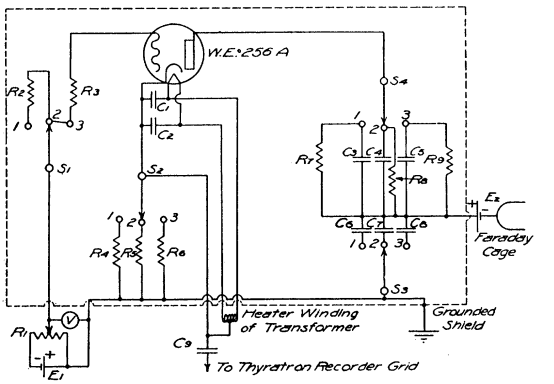

FIG. 2. Circuit of thyratron current integrator.

R_1—25,000 ohm potentiometer, w.w.
R_2, R_3—30 megohm grid leaks
R_4—7000 ohm w.w.
R_5—700 ohm w.w.
R_6—70 ohm w.w.
R_7—50 megohm grid leak
R_8—5 megohm grid leak
R_9—500,000 ohm grid leak
E_1—3 volts
C_1, C_2—0.004 mf condensers
C_3—0.0002 mf mica
C_4—0.002 mf mica
C_5—0.02 mf mica
C_6—0.001 mf } well-insulated
C_7—0.01 mf } condensers
C_8—0.1 mf }
C_9—0.002 mf
E_2—45 volts

[6] The construction and operation of the generator are described in Phys. Rev. **51**, 75 (1937).

[7] John R. Dunning, Rev. Sci. Inst. **5**, 387 (1934).

from its normal −2.0 volts to −2.5 volts. Switch points with the same numbers are used simultaneously.

A shield, represented by the dotted line, keeps the time constant of the grid circuit fixed and provides a chamber which can be dried to prevent surface leakage of charge. However, in spite of the elimination of surface leakage, the two high sensitivity calibration curves of counts per minute as a function of current to the Faraday cage show an apparent leakage. The error is negligible for the low sensitivity, 1 to 10 microamperes; but in the cases of 0.1 to 1.0 and 0.02 to 0.1 microampere sensitivities a small number of counts per minute must be added to the observed number. The Western Electric type 256 A thyratron was selected because tube leakage, which seems to be the cause of the error, is known to be very small.

In the seven months of operation the calibration has shifted toward slightly less sensitivity and less tube leakage. Consequently if absolute data are wanted the calibration should be checked periodically.*

Results

Lithium

For the preparation of lithium targets metallic lithium in crucible C of Fig. 1 was evaporated onto target holder H, which was perpendicular to the stream of lithium vapor during evaporation and was then turned into position for proton bombardment. Work with a small electrostatic generator at this laboratory had shown that gamma-ray emission from lithium has a sharp resonance for protons at about 440 kv which agrees with the results of Hafstad, Heydenburg and Tuve. This 440 kv resonance was carefully studied with the large generator using lithium films of several different thicknesses and the resonance position was used to determine the sensitivity of the generating voltmeter with which the large generator is equipped. Details of the calibration experiments are given in the paper which describes the large generator.

After the low voltage calibration work several runs were taken on lithium targets over the entire voltage range of the generator. Curve A of Fig. 3 shows the yield from a target having an absorption thickness of about 30 kv for 440 kv protons. The resonance peak at 440 kv is much broader from this film than from several of the thinner films used for voltage calibration. An accurate determination of the width of the 440 kv level is being made with the small generator and will be given in a later publication. Above the 440 kv resonance, curve A drops to a level which is only slightly above the x-ray background (background intensity is not subtracted) and at 900 kv shows another comparatively gradual rise with a broad maximum at about 1000 kv. Above 1200 kv the yield curve has a smooth rise which is approximately exponential up to the maximum generator potential. For curve B the lithium film used for curve A was moved about 5/16 inch so that the proton beam hit at a different place. Results of this run, which was taken to check the resonance peak at 1000 kv, show good agreement with curve A.

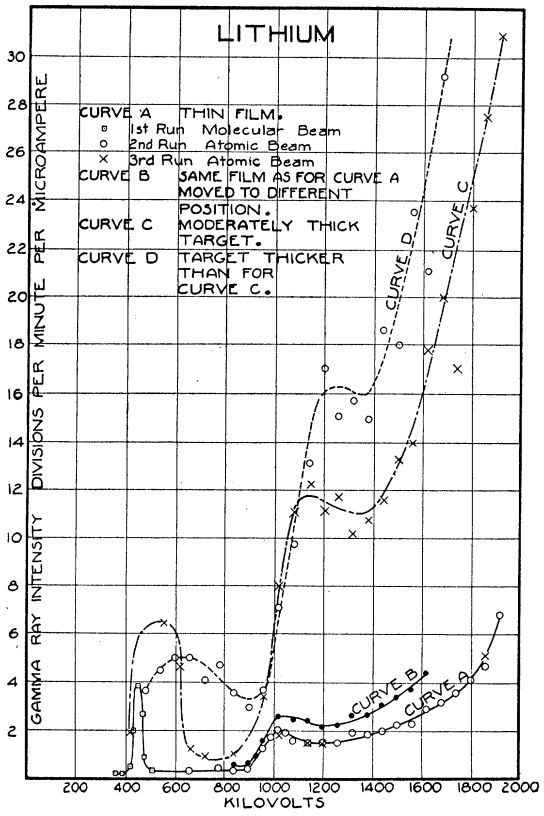

FIG. 3.

* For precision work it is necessary to keep the thyratron heater current constant. A 5 percent drop in the heater current decreases the count 3 percent.

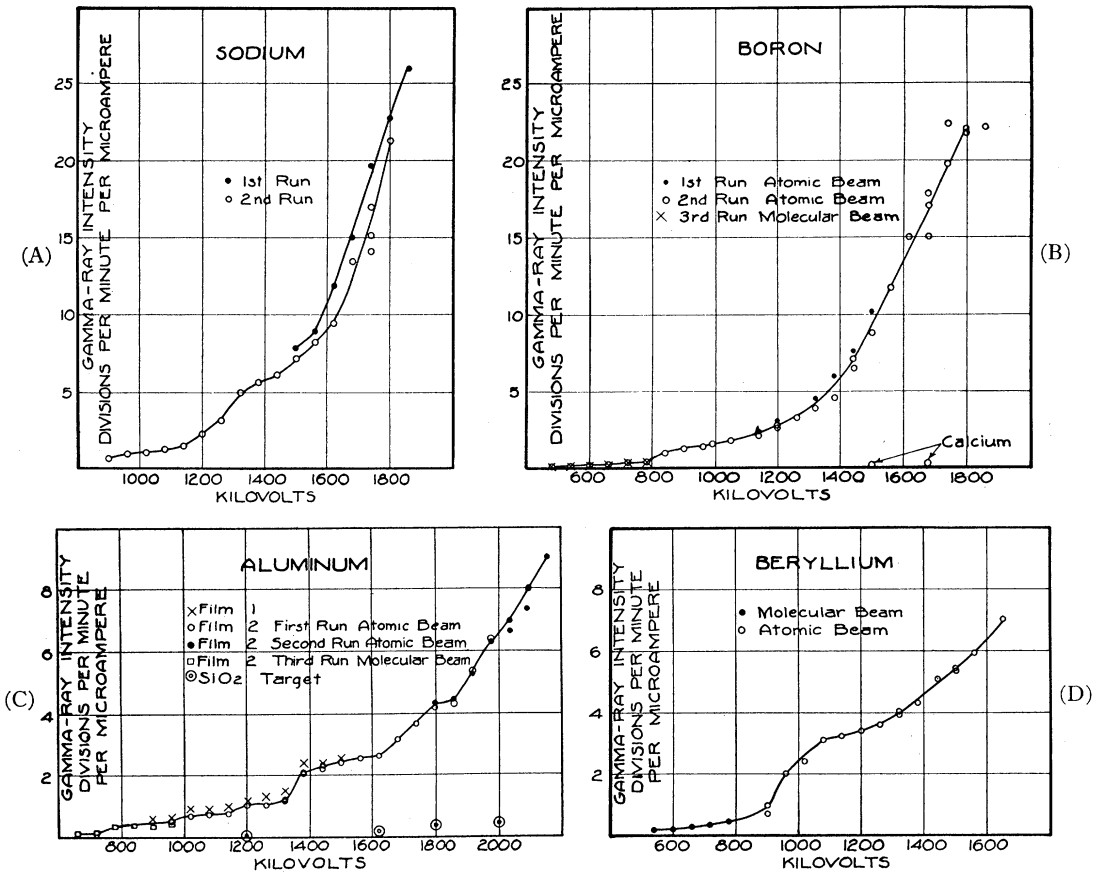

FIG. 4.

Curves *C* and *D* show the results obtained from two comparatively thick lithium films. During these runs the charging belts of the generator were in poor condition and caused considerable voltage fluctuation so that the data are not very reliable, but in both curves the effect of the 1000 kv resonance is quite clearly shown. It was expected that the lithium film used for curve *D* would completely stop the most energetic protons and would thus give a thick target yield curve, but the form of the curve shows that the film was not sufficiently thick.

The lithium gamma-ray resonance at 440 kv is known to be due to the Li^7 isotope and recent work of Rumbaugh and Hafstad,[8] using voltages up to 1100 kv showed that the 1000 kv resonance is also due to Li^7. It therefore seems quite likely that all of the observed gamma-radiation from lithium is from Li^7. Recent work of Delsasso, Fowler, and Lauritsen[9] indicates that the energy spectrum of lithium gamma-radiation from the 440 kv resonance consists of a strong line at 17.5 Mev and a weaker line at about 14.0 Mev. Gaerttner and Crane,[10] however, find indications of a number of low energy components in the gamma-ray spectrum.

The reaction responsible for gamma-ray emission from lithium is still not definitely established. The product is probably stable Be^8 with all available energy emitted as gamma-radiation but there seems to be a possibility that the gamma-radiation carries away most, but not all, of the available energy and that two short range alpha-particles are emitted.[11]

[8] Rumbaugh and Hafstad, Phys. Rev. **50**, 681 (1936).

[9] Delsasso, Fowler, and Lauritsen, Pasadena meeting, Am. Phys. Soc. (1936).

[10] Gaerttner and Crane, Phys. Rev. **51**, 49 (1937).

[11] A thorough discussion of the process of gamma-ray emission from lithium is given by Professor G. Breit in a paper by L. R. Hafstad, N. P. Heydenburg and M. A. Tuve, Phys. Rev. **50**, 504, 510–514 (1936).

Beryllium

The target used for this work was a thick piece of beryllium metal obtained from the Belmont Smelting and Refining Company, who guaranteed the metal to have a purity of 98 percent or better. As the shape of the yield curve (Fig. 4D) is considerably different from the yield curves given by other elements it seems improbable that much of the observed gamma-radiation could have been due to contaminants.

The curve of Fig. 4D indicates that beryllium has a broad resonance level for gamma-ray excitation at approximately 990 kv. In other regions the curve shows a smooth increase of gamma-ray yield with voltage.

Gamma-radiation due to bombardment of beryllium by protons was first observed by Crane, Delsasso, Fowler and Lauritsen[2] and was later observed by Hafstad and Tuve.[12] From cloud chamber measurements of the gamma-ray energy Lauritsen concluded that the gamma-ray spectrum consists of a number of lines of which the hardest component has an energy of about 6 Mev. From these results he concluded that the reaction is probably as given by the equation $_4Be^9+{}_1H^1={}_5B^{10}+\gamma$. To account for low energy components of the gamma-ray spectrum Lauritsen assumed that the radiation may be emitted in a number of steps. Low energy gamma-radiation might, however, be due to an excited product nucleus in the reaction $_4Be^9+{}_1H^1={}_3Li^6+{}_2He^4$ or in the reaction $_4Be^9+{}_1H^1={}_4Be^8+{}_1H^2$.[13]

Boron

A target of metallic boron was prepared in the following way. Granulated boron metal was spread onto a molybdenum sheet to form a layer about 1 mm thick. This was wetted with distilled water and after the water evaporated the granules were found to be firmly enough bound so that the target could be inverted and still remain intact. The boron used in this work was of unknown purity but the observed gamma-ray intensity as shown by Fig. 4B is of such magnitude that only fluorine and possibly lithium would be serious as contaminants. As yield curves from lithium and fluorine are considerably different in shape from the boron yield curve it seems unlikely that either of these materials could have contributed appreciably to the intensity observed from the boron target. The yield curve for boron as shown in Fig. 4B gives a smooth rise of intensity with voltage except for a weak resonance at approximately 820 kv.

Crane, Delsasso, Fowler and Lauritsen[3] have reported the discovery of gamma-radiation from boron due to proton bombardment and from cloud chamber measurements of the gamma-ray energies they concluded that some, and perhaps all, of the radiation is due to the reaction $_5B^{11}+{}_1H^1=C^{12}+\gamma$. Calculations of Breit and Wigner indicate, however, that the process involving emission of an alpha-particle leaving Be^8 in an excited state may also take place with an appreciable probability.

Fluorine

For the investigation of gamma-radiation from fluorine two different targets were used. The first target was a thick layer of CaF_2 powder which adhered satisfactorily to a molybdenum plate after being wetted with distilled water and then permitted to dry. For the preparation of the

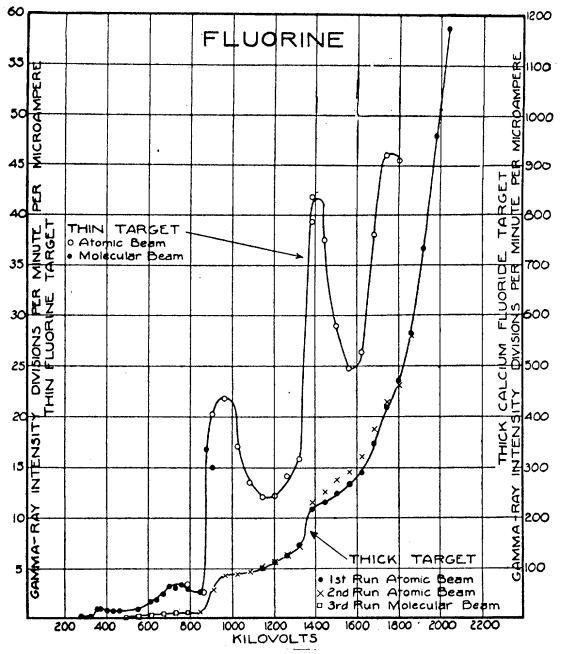

FIG. 5.

[12] Hafstad and Tuve, Phys. Rev. **48**, 306 (1935).

[13] The probability of gamma-ray emission in these reactions is discussed in a paper by Breit and Wigner which will be published soon in the Physical Review.

second target, which was to be thin, an iron sheet was immersed in a weak solution of hydrofluoric acid and the resulting thin layer of combined fluorine was found to be satisfactory as a target.

The gamma-ray yield from fluorine as shown by the curves of Fig. 5 is outstanding because of the number and prominence of resonance levels and because of the great intensity of the yield. Two separate runs on the CaF_2 target show good agreement and peaks in the thin target yield curve correspond closely to steps in the thick target curve. The contribution of Ca to the gamma-ray intensity from CaF_2 is negligible since the yield at 1680 kv from a target of metallic calcium was less than 0.1 percent of the yield from CaF_2 at the same voltage. Because of an oversight iron was not investigated for gamma-ray emission, but the great intensity of the fluorine radiation and the correspondence of the thick and thin target yield curves indicate that the iron background did not contribute appreciably to the thin target yield.

Hafstad, Heydenburg, and Tuve[5] made a careful investigation of gamma-ray emission from a thick CaF_2 target using protons with energies up to about 1050 kv. They observed sharp resonances at 328, 892, and 942 kv and a broad resonance between 600 kv and 700 kv. Because of lack of time the experimental points in the yield curves of Fig. 5 were not taken at closely enough spaced voltages to resolve the 892, 942 kv doublet. A comparison of the curves of Fig. 5 with results obtained at Washington shows, however, that the voltage scales at the two laboratories agree to within a few percent over the voltage range from 328 kv to 900 kv.

In addition to the low voltage resonance peaks which check the results of Hafstad, Heydenburg, and Tuve, the curves of Fig. 5 show a prominent resonance at approximately 1400 kv and give indications of another resonance at 1760 kv.

From their calculations Breit and Wigner find that for fluorine the most probable reaction giving gamma-radiation is $_9F^{19}+{}_1H^1={}_8O^{16}+{}_2He^4$ with formation of O^{16} nuclei in an excited state and subsequent emission of radiation. This conclusion is supported by the work of Crane, Delsasso, Fowler, and Lauritsen who made cloud chamber measurements of the energy of gamma-radiation from fluorine bombarded by 0.9 Mev protons. They concluded that the radiation is probably monochromatic with an energy of 5.4 Mev. If the reaction is assumed to be one of radiative capture with formation of stable Ne^{20} the total energy available would be 13.9 Mev. The absence of radiation harder than 5.4 Mev supports the conclusion of Breit and Wigner that the reaction involving radiative capture is much less probable than the reaction leading to formation of excited $_8O^{16}$ nuclei.

Sodium

A thick sheet of sodium metal obtained from Mallinckrodt gave the yield curve shown in Fig. 4A. Although the purity of the sodium is not known an examination of the shape of the yield curve and the yield intensity shows that the results could hardly be due to contaminants. As a further check on these results gamma-ray intensities have been measured recently from targets of sodium metal, analytical reagent, obtained from Mallinckrodt. The impurities listed in the analysis of this metal could not possibly contribute appreciably to the gamma-ray yield. Although fluorine was not listed in the analysis we were assured by correspondence with the Mallinckrodt Company that the fluorine content is almost certainly less than 0.0015 percent and is therefore negligible.

Gamma-ray yields from sodium were found to be considerably affected by the layer of sodium oxide and sodium hydroxide which formed on the targets when they were being mounted. Two targets of sodium metal analytical reagent were studied, and of these two the first was exposed to air much longer than the second. At 1800 kv the gamma-ray yield from the first was 0.75 of the yield shown in Fig. 4A and from the second the yield was 1.2 of the value given by Fig. 4A. Since oxygen has been found to be inactive these results indicate that the yields of Fig. 4A are not due to contaminants but that the intensities are somewhat lower than would be obtained from pure sodium.

The yield curve of Fig. 4A shows one broad resonance between about 1150 kv and 1320 kv.

Gamma-radiation from sodium is probably due

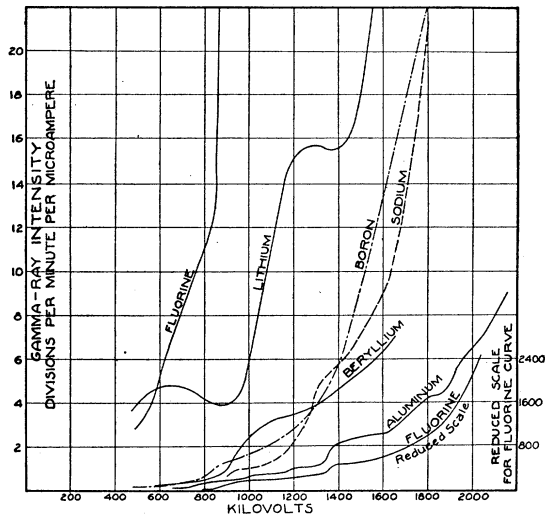

FIG. 6. Comparison of yield curves.

to one of the two following reactions:

$$_{11}Na^{23}+{}_{1}H^{1}={}_{12}Mg^{24}+\gamma, \quad (1)$$

$$_{11}Na^{23}+{}_{1}H^{1}={}_{10}Ne^{*20}+{}_{2}He^{4}, \quad (2)$$
$$_{10}Ne^{*20}={}_{10}Ne^{20}+\gamma,$$

or possibly both reactions may occur.

Using the new mass values of Aston and Pollard and assuming an energy of 1.8 Mev for the incident protons we find 11.5 Mev for the available energy in reaction (1) and 4.4 Mev in reaction (2). In reaction (2) part of the energy will go into kinetic energy of the alpha-particle.

Because of the great difference between the energy changes in these two reactions a determination of gamma-ray hardness should give information regarding the relative probability of the reactions. An attempt has been made to measure the hardness of the radiation by a determination of its absorption coefficients in lead and copper. The values obtained were 0.52 cm^{-1} for lead and 0.35 cm^{-1} for copper.

In the book *Quantum Theory of Radiation* Heitler gives calculated absorption coefficients for hard gamma-radiation, but from his absorption curves our experimental absorption coefficients for lead and copper do not give consistent values for the gamma-ray energy.

Lauritsen[14] has recently shown that the energy of hard gamma-radiation cannot be determined by ordinary measurement of absorption coefficients and it therefore seems that from our data no conclusion may be drawn regarding the hardness of sodium radiation.

[14] Delsasso, Fowler and Lauritsen, Phys. Rev. **51**, 391 (1937).

Aluminum

Alcoa aluminum foil of sufficient thickness to stop the most energetic protons was used to investigate aluminum radiation. Two different foils were tried, giving the results shown in Fig. 4C. As the data on film 1 were taken before adequate shielding was provided for the electroscope, x-ray background from the generator gave a considerable contribution to the observed intensity. Before taking data on film 2 the shielding arrangement as shown in Fig. 1 was adopted and the x-ray background was then considerably less than 0.1 of the aluminum intensity over all regions of the yield curve above 1200 kv.

Taking into account the difference in background intensity for the two aluminum foils studied their yield curves show good agreement and check runs on film 2 also agree well. The yield curve shows a very prominent resonance at approximately 1370 kv. Weak resonances are also indicated at approximately 750 kv, 990 kv, and 1160 kv, but more work must be done using thin aluminum targets before these can be definitely established. At higher voltages the yield curve gives indications of two broad resonances, one at 1620 and the second at 1850 kv.

A comparison of the yield curve from aluminum with the fluorine yield curve (see Fig. 6) shows that the two are quite similar in general shape and that each has a prominent resonance at approximately 1.37 Mev. It was therefore suspected that the radiation observed from aluminum might be due to a small amount of fluorine present in the aluminum as an impurity. By correspondence with Dr. F. C. Frary, Aluminum Research Laboratory, Aluminum Company of America, we were assured, however, that the fluorine content of this aluminum is of the order of 0.001 percent or less and that there is no possibility of a 0.25 percent fluorine content which is the amount necessary to account for the observed intensity from aluminum.

Breit and Wigner find that the most probable reaction giving gamma-ray emission from alumi-

num is $_{13}Al^{27}+_1H^1=_{11}Si^{28}+\gamma$ where the total energy available for radiation is 13.6 Mev with an incident proton energy of 1.5 Mev.

Other elements

Table I shows the results of a few measurements of gamma-ray intensity from targets giving low yields. To serve as a comparison the yield values from aluminum are included. SiO_2 gave a very low yield and it was at first believed that the observed intensity was entirely due to x-ray background from the generator. Later work showed, however, that a Pt target gave still lower yields and that some of the observed intensity from SiO_2 was actually due to gamma-radiation from the target. It is quite probable, however, that part or all of the observed intensity was due to contaminants, either on the surface or present in the compound as impurities. Intensities from targets of Mo, Ca, Ni, C, brass, and Pb_2O_3 were all above the Pt intensity and therefore above the x-ray background of the generator but as in the case of SiO_2 the effects may have been due to contaminants.

Radiation from targets of K_2CO_3 and $CaCl_2$ were fairly intense but the yield values given are not very reliable because of a possible surface contamination of sodium. These materials were placed on the target holder together with a sheet of sodium which had absorbed a considerable amount of water vapor. When the target chamber was pumped down sputtering occurred from the sodium and probably caused contamination of the K_2CO_3 and $CaCl_2$ targets.

TABLE I. *Gamma-ray intensity in divisions per minute per microampere.*

Kilovolts	1200	1440	1500	1620	1680	1740	1800	1980
Al	1.0	2.3	2.4	2.7	3.1	3.7	4.3	6.4
SiO_2	0.084			0.19			0.37	.50
Pt					0.052		0.25	
Mo						0.38		
Ca		0.18			0.31			
Ni							0.58	
C							0.39	
Brass							0.70	
Pb_2O_3				0.98				
K_2CO_3							1.5	
$CaCl_2$			1.1					

Discussion of Yield Curves

In order to show the relative intensities of gamma-radiation from the six elements studied and to compare the shapes of their yield curves the thick target yields are all plotted to the same scale in Fig. 6. CaF_2 yield values from Fig. 4 were multiplied by a factor of 2.11 to convert them to yields from pure fluorine. Because of the high intensity of the fluorine radiation very little of the yield curve can be shown when plotted to the same scale as that used for weak emitters, and a curve plotted to a reduced scale is therefore included. The lithium yield curve shown is not entirely satisfactory for comparison since the target used was not sufficiently thick to completely stop incident protons of high energy.

The curves of Fig. 6 show that the relative intensities of gamma-radiation from the elements bears no simple relationship to the atomic number of the elements or to the energies released in the reactions. Fluorine, for example, is of higher atomic number than boron, has less reaction energy, and yet gives an intensity, at 1800 kv, 45.5 times as great as the intensity from boron. To explain the observed intensities of gamma-radiation from the elements studied and to explain the shapes of their yield curves, Breit and Wigner in their paper show that it is necessary to take into consideration the nuclear energy level systems of elements involved in the reactions.

The work reported in this paper was intended to be only a preliminary survey and further studies will be made using thin targets and closely spaced voltages in order to determine the widths and positions of nuclear energy levels more accurately.

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