THE SECONDARY EMISSION FROM A NICKEL SURFACE DUE TO POSITIVE ION BOMBARDMENT.

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The research treated in this thesis was suggested by and performed under the direction of Dr. R. A. Millikan. The object of the research was, as is suggested by the title, the investigation of the secondary emission from metallic surfaces when bombarded by positive ions. In the apparatus used the metallic surface was nickel and the positive ions were those of the lighter alkali metals.

This problem has been attacked previously by several workers, Fuchtbauer[1], Cheney[2], Baerwald[3], Hahn[4], Badereau[5], and many others. Before discussing the present attack upon the problem it will be well to consider the qualities desired in the apparatus to be used for the experiment at hand. There are five heads under which these qualities come, viz.:

1. Source of Positive Ions. A source of positive ions is desired that will emit a uniform stream of positive ions, with a low dispersion of velocities about the mean. The source of ions should emit at a low temperature, it should not be capable of contaminating the surfaces in the apparatus, it should be capable of working in a high vacuum, and it should not emit quantities of unionized molecules or atoms.

2. Symmetry of Electric Fields. It is highly desirable that the electric fields used have the highest possible degree of symmetry.

3. Shielding. The collecting cylinders should be thoroughly shielded from the primary ray stream, and also from secondary emission from surfaces other than those of the target.

4. Cleaning of the Target Surface. The target surface should be so placed that it can be cleaned before each run, so as to be sure that there are no coats of oxides, or adsorbed gases.

5. High Vacua. The apparatus must be such that a high vacuum can be maintained at all times during the experiment. The high vacuum is desired not only because a mean free path of considerable length is required, but also in order that the target surface, from which the secondary emission is to be obtained, shall not be contaminated due to the presence of gases during the observations (see paragraph 4. seq.).

The methods used to satisfy these several conditions will now be discussed.

1. Source of Positive Ions. The first method tried was that of Baerwald. He got his positive by means of the evaporation of gaseous molecules from a hot metallic surface. This method will give positive ions, but the current obtained has a logarithmic decrement, and a short time after the first heating of the filament the current will be infinitesimal (Richardson[6]). If this method were to have been used, it would have been necessary to break the vacuum in the apparatus every two or three hours, so that the emitter could be given a new supply of the necessary gas. Also, the primary ion current obtained was so small that it would have been necessary to use the most sensitive electrical measuring apparatus. The second method tried for obtaining positive ions was that of Langmuir and Kingdon. In this method metallic caesium was introduced into the apparatus. The metallic caesium has a vapor pressure at ordinary temperatures of an appreciable amount. Langmuir and Kingdon[7] have shown that if an atom of caesium strikes against a metallic surface whose work function is higher than the ionizing potential of the caesium atom, then the metallic surface will take an electron away from the caesium atom and leave it in an ionized state, adsorbed to the metallic surface. Now if the temperature of the surface is raised a sufficient amount of the ionized caesium atom will be evaporated. In this manner

a supply of positive ions can be obtained. The maximum value of this current will occur when every caesium atom striking the surface will be evaporated as an ion. The difficulty with this method is that all of the metallic surface in the tube becomes covered with a mono-molecular layer of caesium atoms so that any part of the tube that gets to a temperature of 400° C or more will radiate electrons. As the energy required to keep the emitter at the temperature necessary for the evaporation of the ions (approximately 1000° C) will keep parts of the tube well above the temperature mentioned above, there will be an indeterminate amount of stray electron emission in the apparatus.

The third method tried, in the search for a satisfactory source of positive ions was the production of hydrogen ions by the diffusion of the gas through hot palladium. On trying this it was found that the number of ions produced was negligible in comparison with the amount of neutral gas emitted. It was quite impossible to keep a satisfactory vacuum in the tube. An endeavor to increase the number of ions by bombarding the palladium tube met with a very small success, and the attempt was abandoned.

The fourth and final method was then tried. In this arrangement aluminum phosphate was coated upon an emitter of sheet molybdenum and the whole heated to about 1000° C by passing an electric current through it. The $AlPO_4$ then emitted positive ions in large quantities, proportional to the amount of the salt present. By this method a positive ion current as large as 100 microamperes can be easily produced. According to Dempster[8] the positive ions are sodium and potassium, and consequently must be due to impurities in the $AlPO_4$. The action of the aluminum phosphate in emitting positive ions of sodium at the low temperature indicated is quite anomalous. The writer thinks that the most probable explanation is that the $AlPO_4$ acts as an electrolyte (perhaps in the solid state) and the sodium ions are brought to the outer surface of the emitter by the electric field applied in the apparatus. This mechanism seems the more probable because the positive ion current

increases rapidly with the field stength. These emitters, when coated with a layer of phosphate perhaps .3 mm thick, will give at least forty hours of service before the coating wears out. During the time the emitter is active the positive current is extremely steady and at the end of the period there is an extremely rapid decrease in the emission. When the coating is new the emitter will not emit any electrons at its ordinary temperature. As the emitter grows older and older, the salt drops off the molybdenum and finally when the emitter is worn out it becomes quite a good electron emitter. The life of the emitter depends upon the temperature at which the emitter is run, if the temperature of the emitter is raised slightly the emission increases, and as there is only a given amount of the alkaline impurity present, the emitter coating wears out with great rapidity. The $AlPO_4$ is used in place of other salts because it is infusible at any ordinary temperature, and because it sticks to the metallic heating surface. This method has been entirely satisfactory except that the exact nature of the positive ion is not known. The proposed method of finding the exact nature of the positive ions will be discussed later.

2. Symmetry of Electric Fields. To obtain symmetry of the electric fields the apparatus was constructed of a number of concentric cylinders, the fields were applied between these cylinders and the central positive ion emitter. Thus all of the electric fields in the apparatus have axial symmetry, a very desirable characteristic.

3. Shielding. As will be seen by the figure the collecting cylinders C' and C'' are thoroughly shielded by their position back of the cylinders A and B. The cylinder B is used in order to protect the cylinders C from any emission from the edges of the slot in cylinder A. The cylinder B also protects the collectors from any emission which may arise from the necessarily high temperature at which A runs, due to the radiation of heat from the emitter. The ends of the tube A are closed, because it was found that at low accelerating voltages stray emission could find its way through the then open ends of A and arrive at the collecting cylinders or

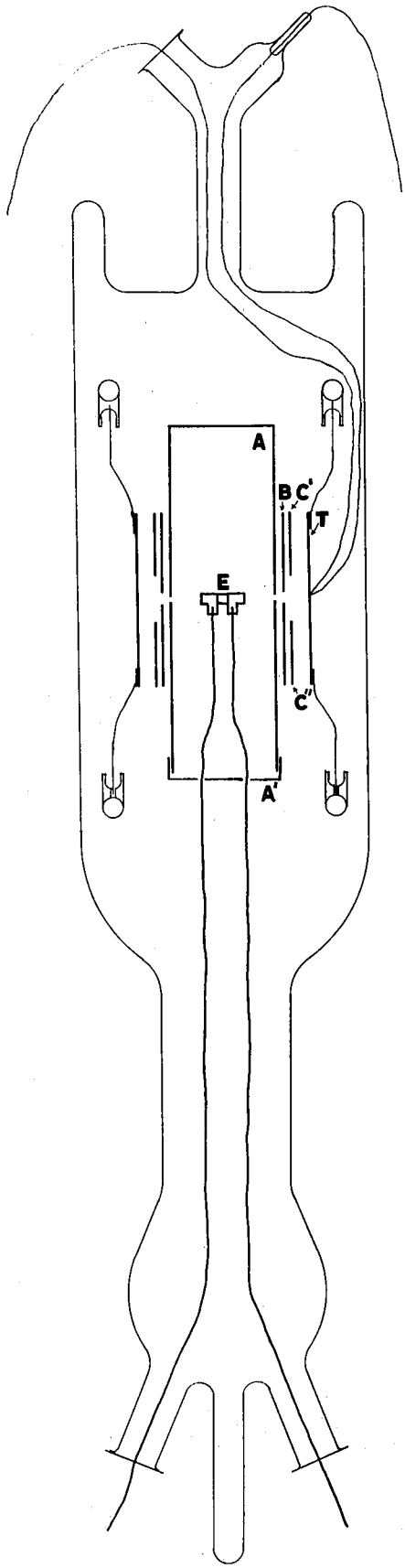

Fig. 1

their supports.

4. The Cleaning of the Target. The cleaning of the target was first attempted by heating by bombardment, but after many failures of the filament and general lack of results by this method it was decided to build an induction furnace. The induction furnace has been extremely satisfactory and has given a minimum of trouble. It will be described in detail later.

5. High Vacuum. The apparatus used was constructed of Pyrex glass, and all of the metal parts were of nickel, tungsten, or copper. The apparatus thus could be subjected to high temperatures and thoroughly out-gassed. The emitter used in the final work did not in any way impair the high vacuum as it emitted no gas after the first hour of operation. The metal parts were so arranged that they could be glowed out at 1000° C by means of an induction furnace. As there were no mercury seals or stopcocks between the liquid air trap and the apparatus, there was nothing to gas in the apparatus itself. Due to these facts and also to the continuous operation of the mercury pumps for months at a time the vacuum was of the highest order. In this way the probability of a primary or secondary particle suffering a collision with a gas molecule is extremely remote. The vacuum was at all times sufficient to give a mean free path of at least fifty meters.

Description of the Apparatus and Experimental Technique.

The first apparatus used in this research has been sufficiently described in the author's master thesis. The present apparatus consists, as has already been mentioned, of a number of concentric nickel cylinders. These nickel cylinders are supported by means of a glass cage which is in turn supported from a glass cup. The cylinders have three wires attached to them at each end and these wires are led into small glass cups which are part of the glass ring which forms the ends of the

glass cage. The wires at the top ring are sealed into the glass cups, but at the bottom they slide, thus allowing for the differential expansion of the cage and the metal cylinders. The three cylinders A, B, and T are supported in this manner, but the cylinders C' and C", not connected to each other, are supported only at their upper and lower ends respectively; the three wires supporting C" being sealed into the lower glass ring. The thermocouple shown in the figure attached to T is made of tungsten and constantin. The two wires are brought out as shown, without being cut at any place; the tungsten wire is sealed directly through the glass while the constantin wire is silver soldered through a copper disk seal. In this manner the cold junction of the thermocouple is completely separated from the apparatus and can consequently be kept at a uniform temperature.

The cylinder A is a nickel cylinder made by rolling up thin sheet nickel into a tube and riveting it. It is ten centimeters long and three centimeters in diameter and has a slot of one and a half millimeter wide cut around its center, except in three places where legs are left to hold the two parts of the tube together. The upper end of the tube is permanently closed by means of a nickel cap welded to the tube. The lower end of the tube is closed by means of a sliding nickel cap A' which is supported from the emitter leads. Tube A is five centimeters longer than any of the other tubes so that it will have an unobstructed radiating area and thus operate at a lower temperature than would be otherwise possible.

Cylinder B is five centimeters long and three and a half centimeters in diameter, it has a slot around its center to register with the one in A but its slot is three millimeters wide. The two collecting cylinders C' and C" are each four centimeters in diameter and two and a half centimeters long. These cylinders are mounted coaxial with A and B and symmetrically about the plane of the slots in A and B in such a manner that the distance between them is thirteen millimeters. The fourth cylinder T is the target and is five centimeters in diameter and five centimeters. long. This cylinder is pierced with an observation hole approximately eight millimeters in diameter, for observing the emitter. The target has attached to it in the

plane of the slots the thermocouple aforementioned.

The emitter E consists of a strip of molybdenum five mils thick, two and a half mm wide and twelve mm long. This strip is bent into the form of an open circular band. The current is run in to the strip at its ends and heats it. The current necessary to obtain a temperature of 1000° C is about forty-one amperes and the voltage drop across the ends of the leads is less than one volt. The emitter strip is covered with the $AlPO_4$ as was discussed in the paragraph on positive ion sources. The emitter is supported only by means of the emitter leads, which in turn are supported by the copper seals shown on the drawing. This construction was adopted in order to make the emitter easily removable, and has justified itself many times over. By cutting the tube shown in the figure, the emitter, together with its leads and seals can be removed complete, without disturbing the grid system which is supported from the other end of the tube. By this construction the emitter has been withdrawn and replaced with a delay, including the time necessary to make a new emitter, of only four hours. The cap A' which is necessary to close the cylinder A is supported from the emitter leads by means of an insulated clamp. The leads run through the cap in glass bushing, so that there is no electrical contact between the cap and the leads. The cap A' has its own lead which is taken out at the bottom of the tube through a tungsten-pyrex seal. This lead was brought out so as to be sure that A and A' were in electrical contact. This rather elaborate method of supporting A is necessary in order that the emitter can be easily extracted.

The leads from the various cylinders are led out through tungsten-pyrex seals in the cup at the top of the tube. All of the leads from the cylinders are in the cup and the entire assembly of the cylinders is supported from the cup so that by breaking the large annular seal at the top of the tube the cylindrical grid system together with its leads may be withdrawn intact. This arrangement

enormously simplifies the construction of the apparatus, as all of the cylinders can be assembled, and the electrical circuits completed, before the apparatus is sealed into its bulb.

The evacuating system used consisted of a Hyvac pump, a two-stage glass mercury diffusion pump, a liquid air mercury trap, a McLeod gauge, and the apparatus itself. The glass ware was all of pyrex glass and the pumps were all electrically operated. The evacuating system was capable of producing a pressure of less than 10^{-6} mm of mercury.

The induction furnace used to bake out the target consisted of a ten kilowatt ten thousand volt transformer, a glass plate condenser of 0.2 microfarads capacity, and a simple horn gap made of heavy wall copper tubing. The inductor coil was made of a few turns of three sixteenths inch copper tubing wound on the outside of the glass tube inclosing the apparatus. The electrical arrangements were very simple, the transformer charged the condenser through choke coils till the voltage of the condenser was high enough to break down the spark gap, then the condenser discharged through the inductor which was in series with the spark gap. The discharge of the condenser was of course of an oscillating nature and the currents through the inductor were extremely large. The induction furnace would heat the target to 1000° C in a very minutes with expenditure of about four kw. The efficiency of the furnace was about thirty per cent, as shown from computations from the emissivity of the nickel surface. The only trouble experienced with the furnace was from the overheating of the condenser, as the plates would puncture if the furnace was used for more than ten minutes. If the condenser had been inclosed in oil the induction furnace could have run indefinitely.

Electrical Circuits.

The electrical connections used in all of the observations are shown in the wiring diagram (Fig. 2). Here the tube is represented diagramatically. The thermojunction is not shown in the diagram. The heating current for the emitter was provided by three six volt Electra storage batteries of one hundred and fifty ampere hour capacity. These three batteries were used connected in parallel, so that the filament battery had a capacity of four hundred and fifty ampere hours at six volts. This battery was connected through a controlling resistance and an ammeter to the emitter. The emitter current used was usually forty-one and a half amperes. The emission was very sensitive to small irregularities in the heating current, consequently the battery had to be of large size in order that the current should not change. The battery was of such a capacity that the current seldom dropped off more than three tenths of an ampere during two hours. The source of potential marked GEN in the diagram was obtained from the building direct current supply. By working at night it was possible to raise the voltage of the generator to four hundred volts. The accelerating potential for the positive ions is applied between the emitter and the cylinder B, the ions being first accelerated to A and then falling through another field to B. The purpose of this subdivision of the accelerating potential was to provide a retarding potential for any electrons liberated from the edges of the slot in A. The slot in B being twice as wide as that in A, there is a very small chance of the positive ion stream hitting the edges of it. The voltage V_2 was held usually at about seventy-five percent of V_1, it was found by experiment that the ratio of the two potentials was not important as long as it was not near either to one or to zero. The target T was connected back to B through the galvanometer G_1. G_1 was a Leeds and Northrup galvanometer, type R, its sensitivity was of the order eighteen hundred megohms. Its range was controlled by a Leeds

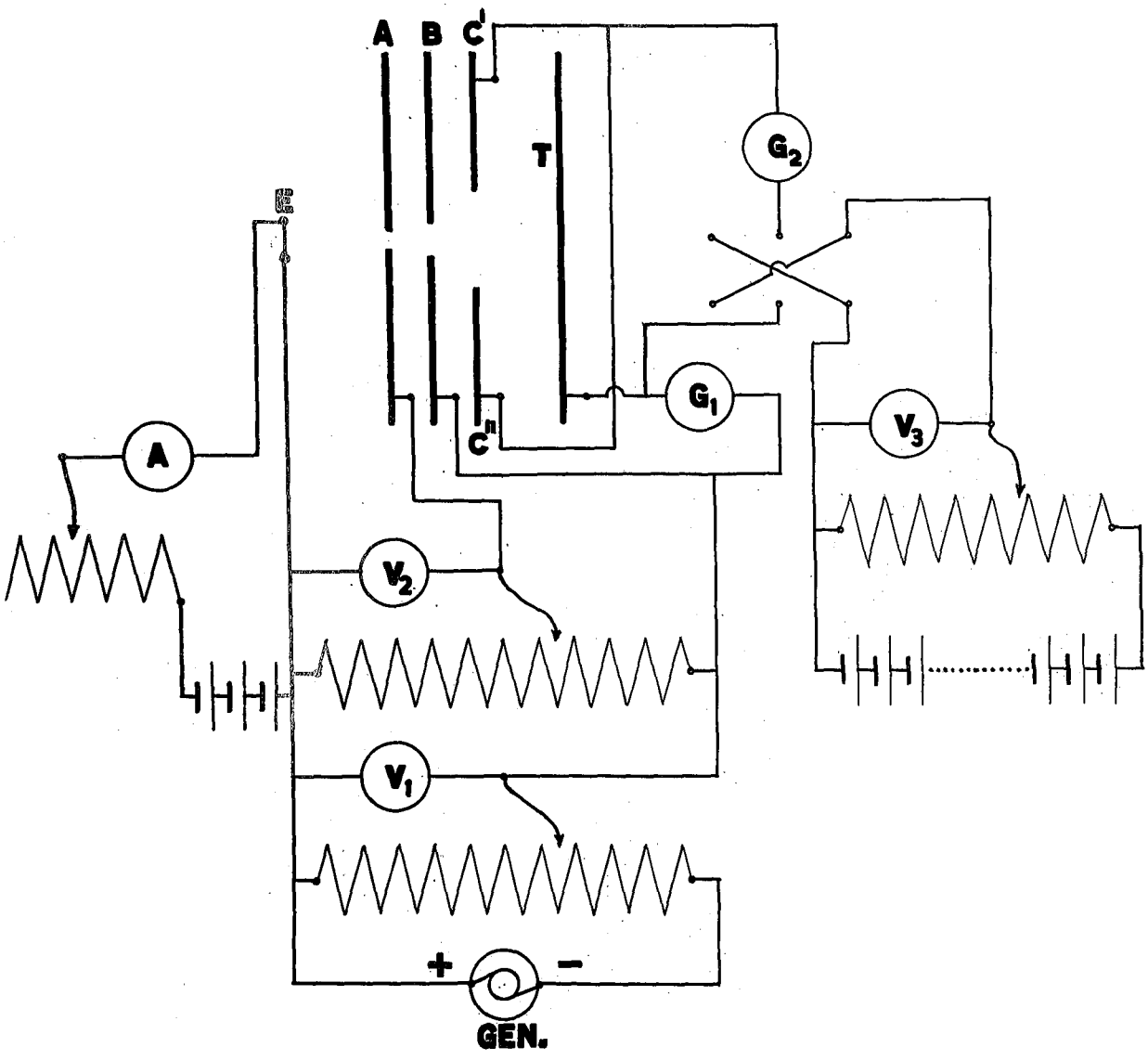

Fig. 2

and Northrup Ayrton Shunt. This galvanometer reads all of the current that arrives at the target and also, as we shall see, the current that arrives on the collectors C, from the emitter. The collectors C' and C" are connected together outside of the tube and then through the galvanometer G_2, through the reversing switch and potential V_3 to the target. Thus the galvanometer G_2 will read all of the current incident on the collectors, while the galvanometer G_1 reads the current arriving from the emitter on the target and collectors. As the collectors are so well shielded that no current can arrive on them directly from the emitter, the current read by the galvanometer G_1 must be only the secondary current from the target or a primary radiation reflected from the target. Thus it follows that the ratio of the readings of galvanometers will be the ratio of the secondary or reflected current to the primary current. The potential V_3 consists of a number of Burgess B batteries and a small storage battery of thirty-five cells. The storage battery was connected across a potentiometer to give small variations in potential while the B batteries were cut in one at a time to give the coarse steps. The voltmeter had a total resistance of one megohm so that it would not run down the B batteries. This high resistance instrument was obtained by using a Rawson Multimeter of ten thousand ohms resistance on the one volt scale, in series with 990.000 ohms, to make it a hundred volt instrument. The reversing switch was used to change the relative polarity of the target and collectors. The galvanometer G_2 was a Leeds and Northrup type HS instrument of 15.600 megohm sensitivity; it was always used with a Leeds and Northrup Ayrton Shunt. The ratio of the sensitivities of the two galvanometers was 8.7.

The thermocouple (not shown in the diagram, but it was attached to the target) was read by means of a Rawson Multimeter used as a Millivoltmeter.

Methods of Procedure.

As the object of this particular piece of work was to determine as much as possible about the secondary radiation from a metallic surface, the first and most obvious thing to do was to run a saturation curve of the secondary and reflected radiation from the metallic surface. This saturation curve will enable the velocity distribution of the various radiations to be easily determined. These saturation curves were all taken at constant accelerating potentials, by varying the collecting potential between the target and the collectors. The run was started by setting the primary accelerating potential at a given value, then the collecting potential on the collector was made negative and equal in magnitude to the accelerating potential. The collecting potential was then decreased slowly to zero and then increased in the opposite direction (positive) until it was twice the value of the primary voltage. Thus at the beginning of the run all of the reflected positive ions were collected while all of the secondary electrons were kept from arriving at the collector. Then as the potential of the collector was decreased some of the reflected positive ions with high velocity stopped arriving at the collector, and as the voltage decreased more and more of these high speed positives failed to arrive at the collector, while simultaneously some of the secondary electrons that had high speed commenced arriving at the collectors. As the voltage goes through zero the electrons are beginning to be collected and the reflected positive ions are repelled. It was found that there were high speed reflected positive ions with very high energy of nearly equal to that of the primary beam. When the voltage reaches that of the primary ions the curve reaches a saturation value. The curve always starts with the current in such a direction as to indicate that the ions are positive; as the collecting voltage approaches zero the current becomes smaller, and in the vicinity of zero volts the curve takes a sharp break upwards, showing that there is a cluster of positive ions with velocity grouped around zero volts.

These curves prove that the secondary electron current does reach a saturation value and thus justifies the taking of a curve showing the variation of the secondary electron emission with the primary ion energy, provided that it is certain that the curve is taken when the current is saturated. Our preceding curves all showed that the secondary current is saturated when the collecting potential for electrons is equal to or greater than the primary accelerating potential. Curves were consequently taken in this manner with the collecting potential twenty-five percent greater than the primary potential. The curve obtained is shown in Fig. 6. The reason that the saturation values could not be taken from the saturation curves directly was that the history of the surface had changed between the different runs. The surface was glowed out between observations, and the different curves were taken over a considerable time, so that the condition of the surface could be quite different during the different runs. For another thing, according to Dempster (loc. cit.) the kind of positive ions given off by the $AlPO_4$ might vary during the life of the emitter so that two saturation curves did not necessarily represent the same conditions. For these sufficient reasons it was thought better to make a separate run of the saturated values. Attempts have been made before to get a curve which shows the relation between the primary energy and the quantity of secondary emission, but in all previous works no attempt has been made to determine the shape of the velocity distribution, consequently the results obtained might not be for the saturation value.

Discussion of Curves.

The saturation curves shown in Figs. 3, 4, and 5 are all plotted with the ordinates representing the ratio of the secondary to the primary current expressed as a percent. The abscissas represent the collecting potential, and the numbers at the end of the curves show the value of the primary ion energy for that

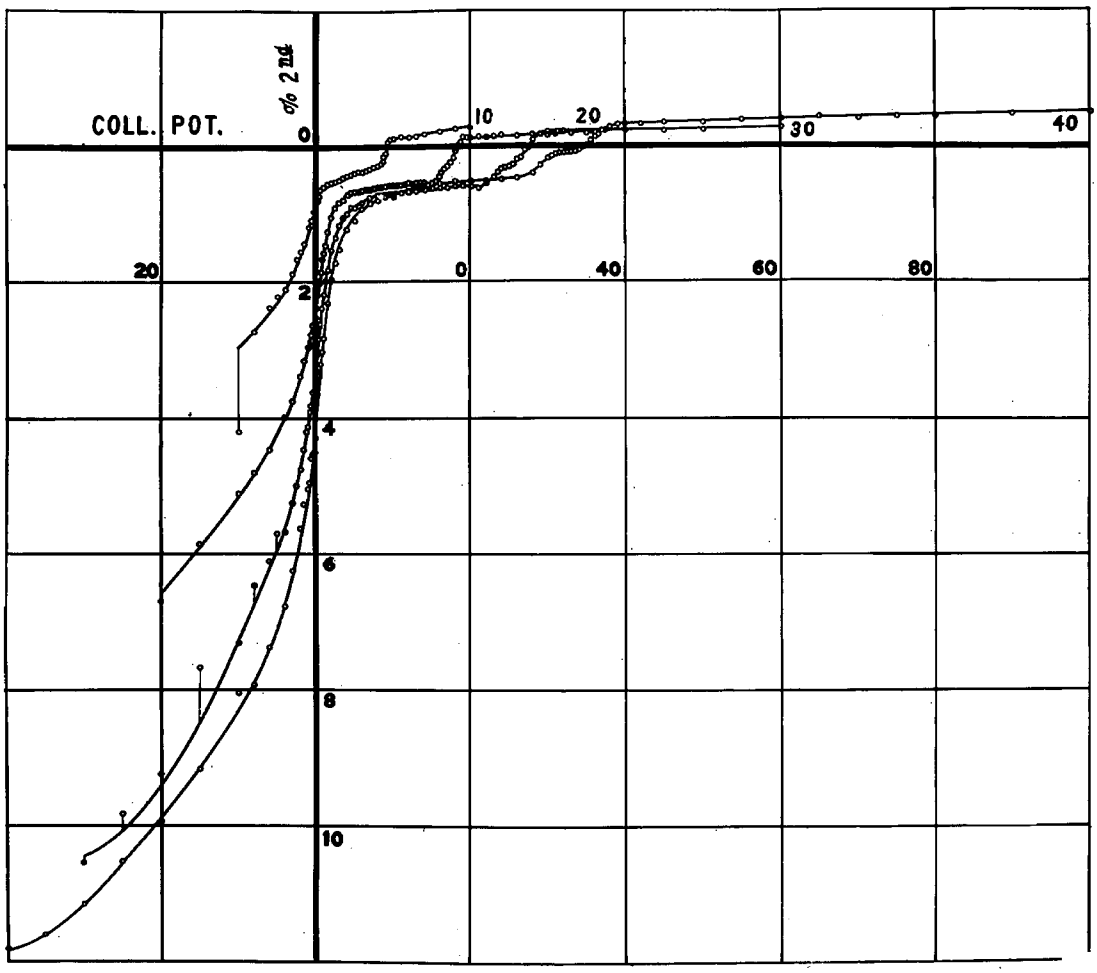

Fig. 3

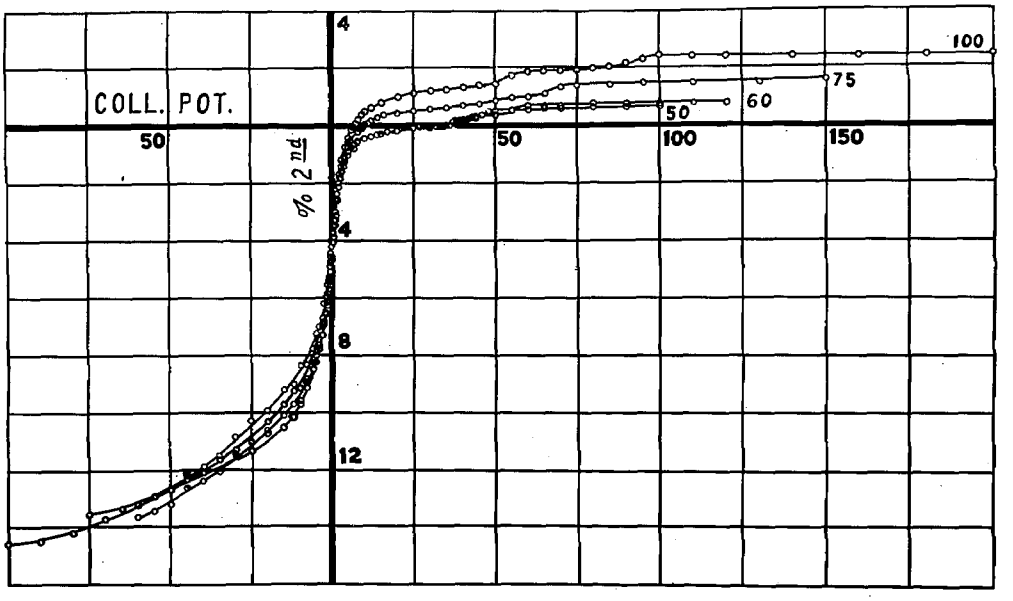

Fig. 4

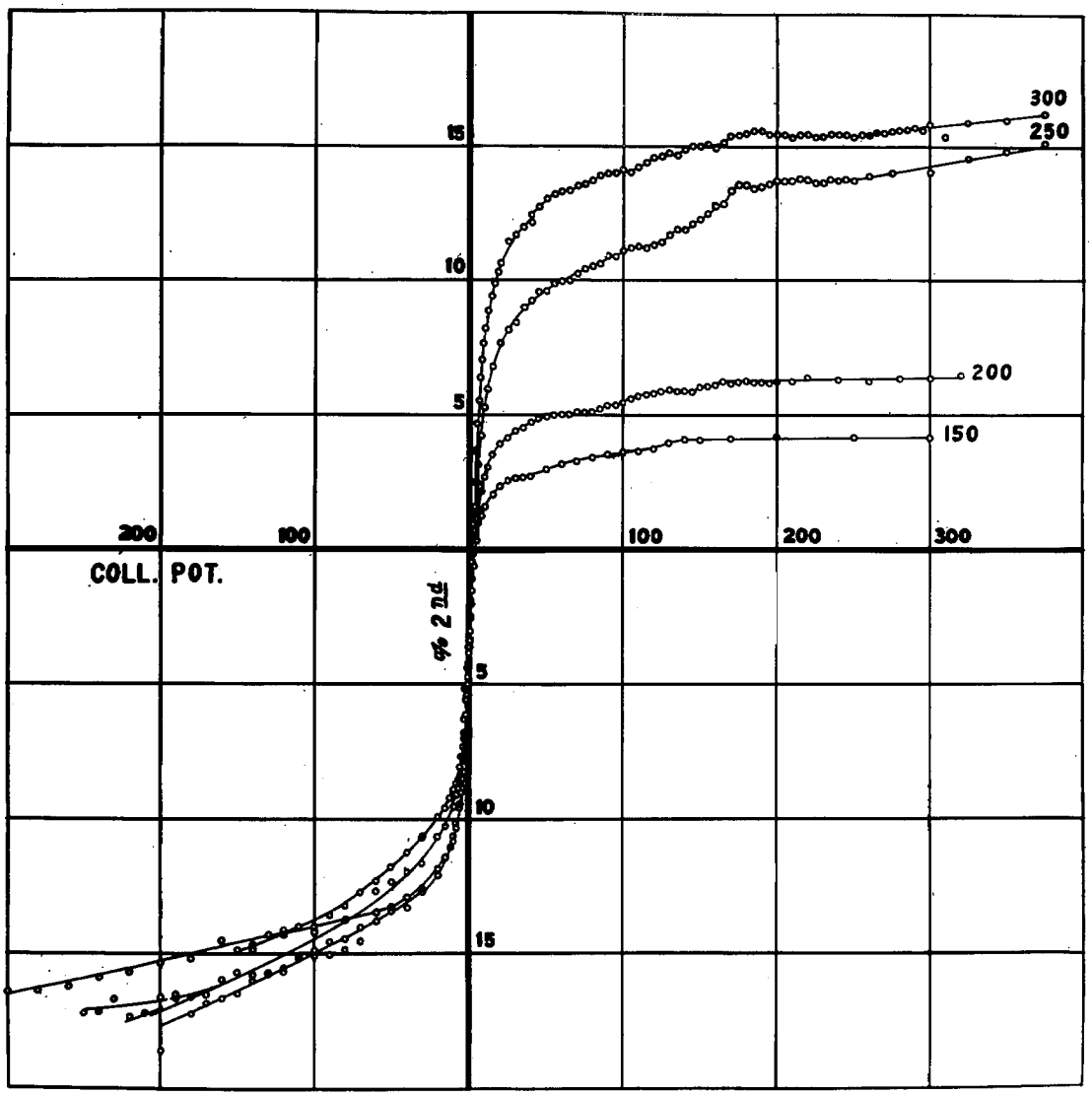

Fig. 5

particular curve. In the plots the ordinates above the axis represent secondary electron emission, that below reflected or secondary, positive ions; the abscissas to the right represent positive collecting potentials, those to the left negative. It will be noticed that the points to the left of the y-axis are rather scattered and do not lie on a smooth curve as do the points to the right of the y-axis. This condition is due to the pulling of the ions out of the primary beam by the potential of the collector. It will be noticed that as the negative collecting potential becomes nearer to that of the primary beam the points scatter more and more. It will also be noticed that curves to the right of the axis are practically identical for all of the different voltages being simply traced further and further to the left as the voltage of the primary ions increases. There would be no point in increasing the negative potential on the collector to more than the accelerating potential as then the collector would simply gather in the ions from the primary beam and the results would be of no value. It will be noticed that all of the curves have strong points of inflection in the vicinity of zero volts. This large change in the ordinates must mean that there are large quanitites of reflected positive ions with energies in the neighborhood of zero volts, as the change is far greater than the total electron saturation current could possibly cause. After this the curves flatten out, and pass through one or more new breaks, and finally reach their saturation value when the collecting potential equals the primary ion voltage. These curves all reach a satisfactory saturation value when the collecting potential has the correct value. The fact that in the curves for low voltages (Fig. 3) the curves do not cross the axis till they are practically at the saturation voltage means that there are large numbers of positive ions reflected with high velocities. There seems to be one group of these reflected positive ions whose energy is about 0.9 that of the incident beam. Other groups of ions are reflected with different energies. This phenomenon serves to mask the

emission of the secondary electrons so that it is only in the curves where the e energy of the incident beam is high that the curve becomes negative when the collecting potential is low. This tendency for the secondary current to remain positive even when the collector has a considerable positive potential is quite remarkable, as it was not expected that the reflected and secondary positive ions would be such a large fraction of the primary ion current.

After it was discovered that the curves all reached a saturation value when the collecting potential was higher than the primary ion potential, a curve was taken by varying the accelerating potential and maintaining the collecting potential always twenty-five percent greater. This curve (Fig. 6) shows the variation of the total secondary current with the positive ion voltage. It will be noticed that the secondary electron current for values below fifty volts is practically negligible. The secondary current increases rapidly with the primary voltage reaching a value of twenty-two percent at three hundred and eighty volts. This curve seems to have a very rapid rate of increase so that by extrapolating along the curve it appears that the secondary current would be equal to the primary current at about nine hundred volts. Badereau's points seem to lie fairly near to this curve, though this must be accidental as he was not working on the saturated part of the curve and there are only four of them in the voltage range covered.

Project for Future Work.

There are a great many other things to be done in the future on this problem. The field of work must be extended to other target surfaces, to other kinds of positive ions, and also to higher voltages. For the different kinds of positive ions, hydrogen together with the series of alkali metals would be most staisfactory. Hydrogen, ionized, would give as a bombarding particle a proton, which is the

smallest known object. The series of the alkali metals would give as ions a seires with complete K, L, M, N, and O shells. This will be very desirable, as then the second ionizing potential will be very high, and consequently the probability that an ion will be doubly ionized is very remote. In order to determine the kind of ions that are given off by the emitter it will be necessary to use a mass spectrograph. The work at higher voltages can be done with the present apparatus but a source of potential of about two thousand volts will be necessary.

The writer wishes to express his deep indebtedness to Dr. Millikan for his help during the progress of this research, Also he wishes to thank Mr. G. A. Alles amd Mr. A. L. Raymond for their help in preparing the aluminum phosphate used, and he desires to express his deepest appreciation to Mr. W. Clancy and Mr. J. Pearson for their help in the design and construction of the apparatus used.

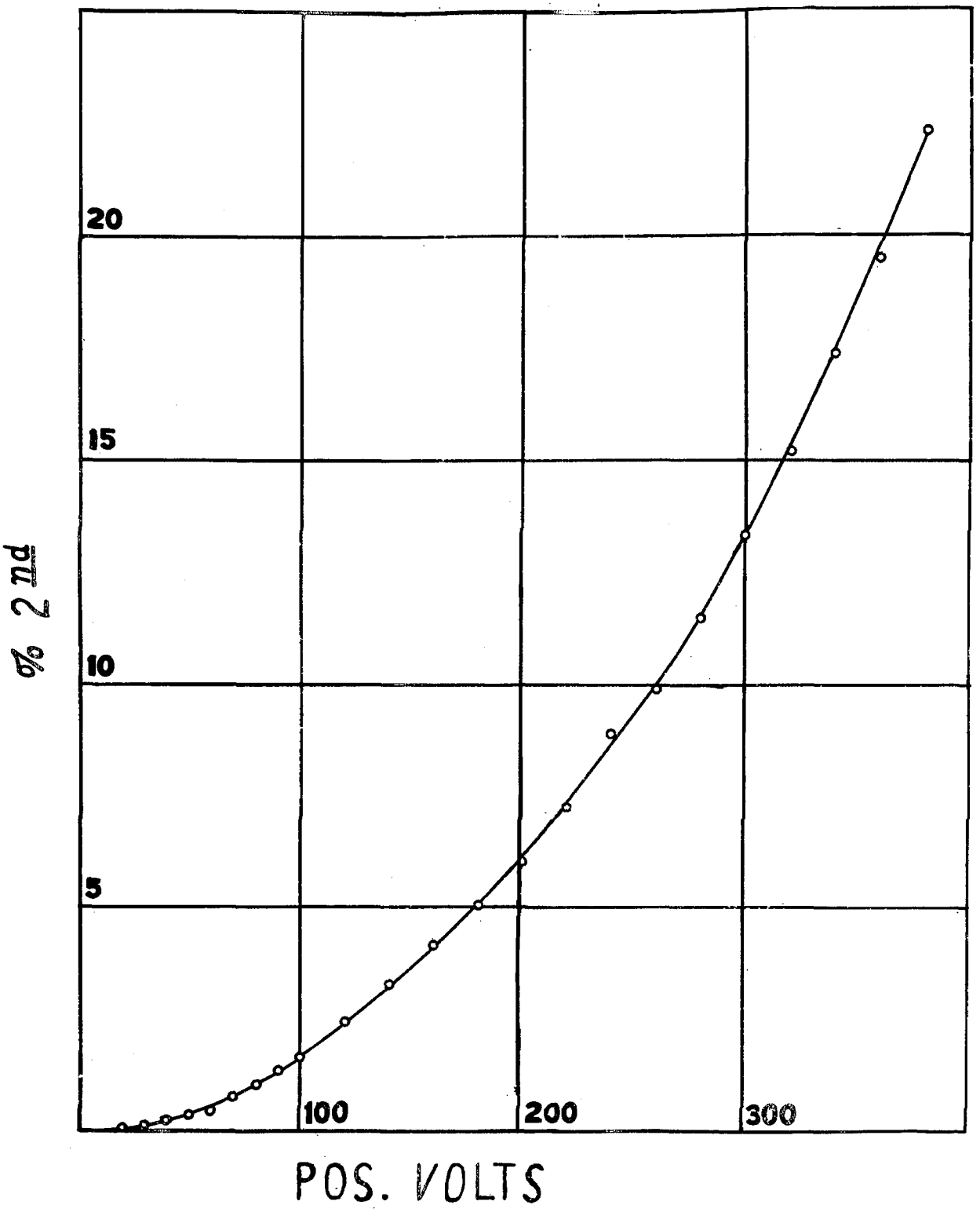

Fig. 6

Bibliography

1) Fuchtbauer a. Phys. Zeits. 7, 153 and 748, 1906.
b. Ann. d. Phys. 23, 308, 1907.

2) Cheney Phys. Rev. 10, 335, 1917.

3) Baerwald a. Ann. d. Phys. 41, 643, 1913.
b. " " " 65, 167, 1921.
c. " " " 60, 1, 1919.

4) Hahn Zeits. f. Phys. 14, 355, 1923.

5) Badereau Phys. Zeits. 25, 137, 1924.

6) Richardson Emission of Electricity from Hot Bodies, ed. 2, p. 196.

7) Langmuir and Kingdon Phys. Rev. 11, 320, 1922.