I. THE EFFECT OF THE APPLICATION OF HIGH VOLTAGES ON THE THERMIONIC EMISSION FROM A THORIATED TUNGSTEN FILAMENT.

II. SOME EXPERIMENTS ON THE ELECTRONIC BOMBARDMENT OF LEAD AND LITHIUM.

Thesis presented for Degree of M.Sc.

by

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The Effect of the Application of High Voltages on the Thermionic Emission from a Thoriated Tungsten Filament.

In 1913, Langmuir,[1] while endeavouring to determine the electron emission from tungsten, in a high vacuum, observed that certain filaments behaved in an erratic manner. This erratic behaviour was ultimately attributed to traces of thoria which were present. Subsequently, Langmuir discovered that by suitable treatment a tungsten filament containing 1 - 2% of thoria could be made to give at temperatures below about 2200° K an electron emission many times greater than that of a pure tungsten filament at corresponding temperatures. When giving large emissions at comparatively low temperatures the filament is said to be activated. To bring the filament into this state, Langmuir found it necessary to heat for a short time at a very high temperature in the neighbourhood of 2600°- 2800° K and then to heat for a longer period at a temperature of about 2000°- 2100° K, called the activating temperature when the emission rose rapidly from that corresponding to a pure tungsten surface to a steady value which was many times greater. He found it convenient to measure the activity of the surface of the filament by measuring

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(1) Langmuir Phys. Rev. 22. 1923. 360

the emission at some low temperature in the neighbourhood of 1400°- 1500° K, called the test temperature, where the rate of activation was very slow.

According to Langmuir, after such treatment the filament remained indefinitely in an active state, provided the temperature was kept below 2200° K and a high vacuum maintained. He found that the presence of oxidising gases destroyed the activity, but that they could be eliminated by introducing easily oxidisable substances, such as alkali vapours; while in the presence of inert gases, the application of large voltages was found to cause deactivation which he attributed to positive ion bombardment of the filament.(2)

To account for the increased emission from an activated filament Langmuir has suggested that the surface of such a filament is partially or completely covered by a layer of thorium, which is reduced from thoria during the heating to a high temperature, and which subsequently diffuses to the surface when the filament is heated at an activating temperature. He bases this suggestion on the experimental result that the work necessary to remove an electron from a thorium surface is less than that required to remove one from a tungsten surface, indicating that a larger emission would be expected from a thorium surface than from a tungsten surface at the same temperature. (3)

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(2) Kingdon & Langmuir Phys. Rev. 20. 1922. 108.
(3) Langmuir Phys. Zeit. Jahrg. 15. 525. 1914.
K. K. Smith Phil. Mag. XXIX 811. 1915.
Langmuir Trans. Amer. Electrochem. Soc. 354. 1916.

According to Langmuir's theory the rate of diffusion depends both on the temperature and on the concentration gradient of the thorium within the filament, the latter factor being determined by the previous history of the filament. As a result of the observed behaviour of thoriated tungsten filaments Langmuir regards the thorium as forming an adsorbed layer on the surface of the tungsten and from a study of the properties of adsorbed films has concluded that they are, in general, of atomic thickness only. From experimental observations he has supposed the activated thoriated tungsten filament to be in a state of equilibrium at any temperature where a steady emission is measured, and this state of equilibrium exists when the number of thorium atoms arriving at the surface of the filament from the interior in unit time, is exactly equal to the number of thorium atoms leaving it by evaporation in the same time. Langmuir distinguishes two types of evaporation, firstly ordinary temperature evaporation and secondly, what he calls induced evaporation. He suggests that the latter kind of evaporation would be caused if the thorium atoms on the surface were more loosely held by underlying thorium atoms than by underlying tungsten atoms, and so if a thorium atom diffusing from the interior arrived just under a surface thorium atom the latter would leave the surface. Langmuir supposes that this actually occurs

and that diffusion of the thorium to the surface of the filament occurs at all temperatures, but at different rates at different temperatures.

He attributed the decrease of emission caused by heating the activated filament to temperatures above about 2200° K (deactivating temperatures) to a decrease in the total amount of thorium on the surface due to the rate of evaporation of thorium from the surface being faster than the rate of diffusion to the surface. He found that the filament could subsequently be made to give its high emission by heating at an activating temperature (about 2000° K) but that the rate of activation was slightly slower than the initial rate, indicating a decrease in concentration gradient, and this rate fell off in successive activations. The original rate could again be realised by flashing momentarily at a very high temperature (about 2900° K) before running at an activating temperature. In support of this theory Langmuir has produced experimental evidence which seems to justify his conclusions.

On account of the property of thoriated tungsten filaments of giving when activated, large electron emissions at comparatively low heating currents, they have been used, among other purposes, as filaments in thermionic valves. It was found, however, during investigations made

at the G. E. C. Laboratory[4], that although special precautions had been taken to remove impurities from the electrodes of such valves and to maintain as high a vacuum as possible, introducing into the valves substances to remove oxidising gases, deactivation of the filament could be caused by applying large voltages to the anode while the temperature of the filament was far below that necessary to cause any decrease of emission due to temperature alone.

The following investigation was carried out with a view to ascertaining the means by which the high emission from thoriated tungsten filaments is adversely affected by the application in vacuo of large voltages when the temperature of the filament is too low to cause deactivation.

Since six thermionic valves containing thoriated tungsten filaments were at hand and since this device satisfied the conditions which were required for the investigation, the latter was carried out with the aid of these valves. They were all of the same type and the filament was surrounded by an open spiral grid and a cylindrical anode. The diameter of the anode was about 1 cm. and its length about 2 cms, while the diameter of the grid was about 3 mms. During manufacture they had been specially treated with a view to removing gaseous impurities from the electrodes

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(4) Thompson and Bartlett. Journ. Inst. Elec. Eng. 62. 1924. 689.

as completely as possible, and, were evacuated to a high degree. They were also provided with substances to remove oxidising gases.

Preliminary experiments were carried out to see if the properties described by Langmuir could be observed. The valves supplied all gave when first tested, at low temperatures, emissions greater than those corresponding to a pure tungsten surface at the same temperatures, but subsequent experience showed that the emission at these temperatures could be improved. Using a valve as a two electrode arrangement (arrangement A p.28) with grid and anode joined, a few trial experiments showed that the filaments did behave in the manner described by Langmuir. Below filament heating currents of ·50 amps, the rate of activation was very slow. Heating currents ranging from ·50 to ·65 amps, caused activation, the rate of activation increasing with increasing temperature, while heating currents of ·70 amps and above caused deactivation and the rate became greater with increasing temperature.

·50 amps was selected as a convenient test heating current since activation was slow at this temperature. Further observations showed that the emission current at ·50 amps heating current was nearly saturated for 40 volts applied to the grid and anode and was about 6 milliamps (Graph I).

These conditions were therefore adopted as standard test conditions throughout the investigation, namely ·50 amps filament heating current and 40 volts applied between anode and filament, the anode and grid being joined.

The other valves showed similar behaviour. When first tested under standard conditions the maximum emission currents obtained by heating at ·60 amps. ranged from 2 to 13 milliamps. Subsequently it was found that by suitable treatment all the filaments could be made to give a maximum emission of 13 milliamps under standard conditions after heating at ·60 amps.

After the filaments had been subjected to several alternate activations and deactivations the rate of increase of emission when the filament was heated at the activating temperature became slower and the temperature at which it was possible to cause deactivation became lower. On the basis of Langmuir's theory this is probably due to different states of the filament, a lower concentration gradient, for example, causing a slower rate of activation Not only did the rate of activation become slower in successive runs but eventually the final emission reached was much less. Accordingly trials were made of the temperature required to obtain reduction of thoria, with a view to increasing the concentration gradient. The filament under investigation

was heated for short times at temperatures ranging from ·80 to ·85 amps h.c. and slight improvement was observed. At a later period in the history of the same valves, however, no improvement in rate of activation could be detected after heating for short times at heating currents in the neighbourhood of ·80 - ·85 amps. The filament was therefore flashed at increasingly higher temperatures and run at an activating temperature. After flashine at 1 amp. h.c. a great improvement in rate of activation was observed on heating at ·60 amps while after flashing at 1·3 amps h.c a saturated emission was obtained in half an hour, when the filament was subsequently heated at ·60 amps.

Having ascertained that so long as the temperature of the filament was not increased above a value corresponding to about ·70 amps h.c. the emission from the filament did not alter with time, the effect of applying large voltages to the valve was studied, the heating current being maintained at a value never greater than ·60 amps.

Using the valve first as a two electrode arrangement with grid and anode joined (see Arrangement A. p.29.) and the filament activated, a voltage of 300 was applied at a filament temperature corresponding to ·50 amps h.c. and the subsequent variation of emission with time was observed. The emission rose immediately to a value higher than that

which had been measured under standard conditions and then fell at first rapidly, but later more slowly until a limiting value was reached. (Graph II). The high initial emission may be attributed to the fact that the flattish part of a current-volts curve indicating saturation is not quite flat but slopes up (see Graph I). On returning to test conditions, the emission was much smaller than the original emission, but on heating the filament at ·60 amps h.c. the emission measured at intervals under standard conditions gradually rose again to its former value.

The effects of applying different voltages for varying lengths of time were then observed. Several time-emission current curves were plotted for various voltages on the grid and anode at ·50 amps h.c. a temperature at which activation is almost negligible and at ·60 amps. h.c. which corresponds to an activating temperature. These curves are shown on graphs II and III. After each deactivation the rate of activation became very gradually slower, during successive runs.

A consideration of the current-time curves taken at the same temperature for different voltages shows that they are all of the same form. The emission at first decreases, but ultimately attains a steady value, and the rate of decrease is faster for higher voltages. At ·50 amps h.c. (non-activating) all the curves appear to reach practically

the same limiting value, while at ·60 amps. h.c. (activating) a lower limiting value is reached the higher the voltage. It may be noted in passing that some of the curves exhibit at first, a portion which is less steep than the succeeding portion of the curve.

A comparison of the results obtained for the same voltage at different temperatures was then made by plotting appropriate curves from graphs II and III or IV and V. These show that at the higher temperature the rate of deactivation is faster, but the limiting value reached is higher than at the lower temperature. For higher voltages, however, the difference between the two limiting values at different temperatures is much less.

So far the effects of applying a large voltage at any given temperature have been investigated by observing the emission at that temperature and voltage at intervals during the application of the voltage. In order to examine more closely the condition of the filament after the application of the same voltage at different heating currents, observations were taken at the standard heating current (·50 amps) at intervals during the application of 300 volts to an activated filament at ·60 amps. h.c. and similar observations at intervals during the application of the voltage at ·50 amps. h.c. The observations are plotted on graph VI,

and the curves show that the filament is brought into a greater state of deactivation by applying 300 volts to the filament at ·56 amps h.c. than by applying 300 volts at ·50 amps. h.c.

It may be concluded, therefore, that the decrease in emission observed is a function both of voltage and temperature.

An inspection of the curves on graphs II and VI for deactivation by 300 volts at ·50 amps. h.c. reveals on II decrease of current from 18 to 4 milliamps occurring in 30 minutes, while on VI decrease from 11 to 6·5 milliamps occurring in 90 minutes. This difference is not due to any difference of electric fields or temperature. The only difference ~~in~~ the two cases is that different valves were employed, and in the former case, the initial emission was about 13.M.A. under standard conditions, while with a much slower rate of activation the initial emission in the latter case was about 8.M.A. For both, however, the emission was approximately the maximum obtainable by running at an activating temperature. According to Langmuir's theory, a much lower value of maximum emission is obtained when the concentration gradient of the reduced thorium is very low. It is probable, therefore that the difference observed between the curves in question is due to a difference of distribution

of thorium within the filament. In several instances the genuineness of the differences in the curves obtained under different conditions was tested by returning to the first set of conditions, after having taken a series of observations under the second. The curve of the results then obtained was practically coincident with the original curve under similar conditions, thus showing that any difference produced in the distribution of thorium within the filament was negligible, at any rate during a series of consecutive runs.

Further experiments were made using the valve as a two electrode arrangement but with grid and filament joined (Arrangement B. p.29.). On application of 300 volts to the anode practically no deactivation was observed either at ·50 amps h.c. or at ·60 amps h.c. For comparison, series of observations with arrangement B and 300 volts applied to the anode were alternated with series with 300 volts applied as in arrangement A. The observations are plotted on graphs VII and VIII which show curves at heating currents of ·50 amps and ·56 amps respectively. These curves show that no deactivation occurs when 300 volts is applied in arrangement B at either temperature, but that deactivation does occur for the same voltage and temperature when the field is applied as in arrangement A.

The carbon tube was surrounded by liquid air and the anode bombarded for several days at a low heating current with electrons of 1200 volts energy, a large resistance (·2 megohms) being included in the circuit to avoid the risk of arcing. As before the tube was placed between the poles of an electromagnet. A faint flickering glow was visible in the tube under these conditions. At intervals the heating current was increased when the glow brightened but eventually disappeared except for a bright blue spot where the electrons hit the anode. On increasing the heating current further this spot became red hot and then white hot and increased in size. Finally a bright pinkish column of glow extended between the filament and anode and red flickerings occurred in the space above the filament.

Examination of the glow through the spectroscope showed a many lined spectrum which was identified as the secondary hydrogen spectrum. H α β and γ were very bright and there was also a bright line due to lithium and occasionally the sodium line could be seen.

After lowering the heating current to its original value the glow was very much brighter than before and did not diminish with time although the carbon tube was kept surrounded by liquid air. Experiments are therefore in progress to try and remove the hydrogen.

No positive results have yet been obtained in this investigation.

II. Some Experiments on the Electronic Bombardment of Lead and Lithium.

Early in 1924, Nagaoka, Sugiura and Mishima[1] published an account of observations they had made of certain spectral lines of mercury, which led them to believe that in the mercury nucleus, one proton was slightly detached from the rest of the nucleus and that the remainder of the nucleus resembled very closely the gold nucleus. Nagaoka thought that if this were really so it might be possible to remove the loosely bound proton from the mercury nucleus, thus effecting a transmutation of mercury into gold. Later[2] he attempted to do this by applying an intense electric field to mercury atoms and he concluded from his experiments that he had been successful, because he was able to detect small amounts of gold at the end of his experiments which had not been detected when similar tests were applied at the beginning of the experiments.

Meanwhile Miethe and Stammreich[3] working independently of Nagaoka detected small quantities of gold in mercury vapour lamps which had been in use for some time, and thought that this might have been derived from mercury.

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(1) Nature vol. 113 p. 459. 1924.
(2) Nature.vol. 13 p. 682 1925. vol 14. p. 85. 1926.
Nature. vol 116 p. 95 1925.
J. Physiques et Ra. vol 6 p. 209 1925.
(3) Miethe Naturw vol 12 p. 357 1924. Nature vol 114 p 197 1924.
Stammreich Naturw vol 12 p 744 1924. Miethe & Stammreich
Naturw vol 113 p 635 1925. Z. Techn. Phys. vol 6. p. 7 1925.
Z. anorg Ch. vol 150 p 350 1926

About the same time Soddy[4] suggested that transmutation might be effected not by striking a proton out of the gold nucleus but by adding an electron to it. He suggested that if high speed electrons were made to collide with mercury atoms, some of them would be directed towards the nucleus and they might possess sufficient energy to penetrate the extra nuclear levels of electrons. If so they would be likely to reach the positively charged nucleus and be captured by it. The change is the inverse of a β - ray change and so the gain of an electron by the mercury nucleus would lower the atomic number by unity, and an isotope of gold isobaric with one of the isotopes of mercury would be produced.

Towards the end of 1924 Smits[5] devised experiments and endeavoured to transmute lead into thallium. In essentials his experiments resembled those of Miethe and Stammreich and he concluded that he was successful in obtaining mercury and thallium since observation of the spectrum of lead vapour showed that as his experiment proceeded lines corresponding to mercury and thallium became visible.

At the beginning of 1925 Stumpf[6] suggested that the particle of atomic weight 197, which Aston had attributed

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(4) Nature vol. 114 p 244. 1924.
(5) Naturw vol. 13 p 699. 1925. Nature vol 117. p 13. 1926.
(6) Nature vol. 115 p 172. 1925.

to mercury was really due to gold. Later, Aston[7] concluded that a mercury isotope of atomic weight 197 does not exist so that gold and mercury are not known to possess any isobares. Since this is so a transmutation of mercury into gold in which an electron is added to the nucleus would produce an isotope of mercury which does not already exist. Theoretically this is not impossible but it is likely that transmutation would occur more easily by the addition of an electron if the product were known to be stable under ordinary conditions. From a consideration of the Periodic Table it appears that at present, apart from mercury and gold, lead and thallium are the only two consecutive elements which have been thought to possess isobares, although their existence has not been established. The following investigation was therefore undertaken in order to attempt a transmutation of lead into thallium.

Since this investigation was begun, however, many negative results of attempted transmutations of mercury into gold have been reported,[8] but no more positive results. It has also been shown that gold in the amounts detected by

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(7) Nature vol. 116. Aug 8. 1925.
(8) Sheldon & Estey. 'Sci. Amer'. p. 296. Nov 1925 & p 389 Dec 1925 Nature vol 116 p 792 1925 Tiede. Schleede & Goldschmidt. Naturw vol 13. p 745. 1925. Piutti & Boggio. Lera. 'Rendic. Accad. Sci. Fis. Mat.' Naples. Sept - Dec. 1925; Nature vol.117. p.604. 1926. Haber. Jaenicke & Matthias. 'Naturw'. vol 14. p.405. 1926; 'Z. anorg. Ch.' vol.153. p.153. 1926. Milan Garrett. 'Proc. Roy. Soc.' vol 112(A) - p.391. 1926.

Miethe and Nagaoka, and attributed by them to transmutation from mercury might well have been present in the apparatus before the experiment.

The method of experiment adopted in the present investigation was the bombardment of lead by electrons from a filament. It was proposed to detect any products by heating the apparatus and subsequently passing a discharge through the vapour and examining the spectrum by means of a spectroscope.

A preliminary experiment was made to obtain and examine the thallium spectrum. A piece of pure thallium was introduced into an ordinary glass tube containing two aluminium electrodes (fig. I) and sealed on to a pump.

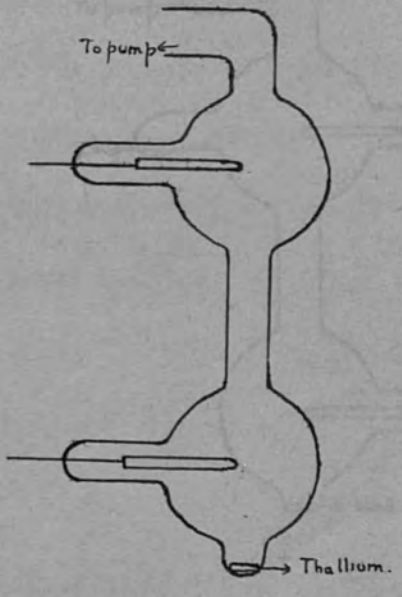

Fig. I.

The tube was exhausted and surrounded by a cylinder of asbestos with holes cut for the leads to the electrodes and a slit, in order to view the spectrum. The tube was heated gently by a small flame placed underneath and a discharge passed from an

induction coil. A greenish glow filled the tube and examination with the spectroscope showed very faint lines and bands, with a very bright green line in the position corresponding to the thallium green line.

A similar experiment was attempted with lead to see whether a lead spectrum could be obtained. Both lead and thallium melt at about 300° C but thallium is supposed to have an appreciable vapour pressure at about 200° C, while lead does not exert an appreciable vapour pressure till about 800° C. Accordingly two pieces of lead were enclosed in a pyrex glass tube into which aluminium electrodes were sealed (fig. II).

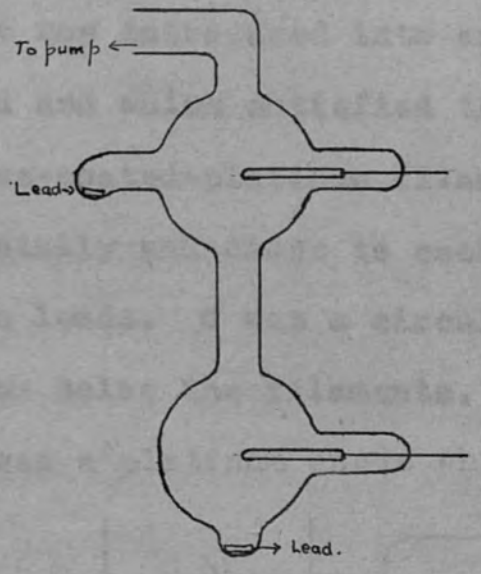

Fig. II

(The lead was prepared from recrystallised lead nitrate by heating to convert to litharge and reducing the latter in a stream of electrolytic hydrogen) The tube was sealed on to the pump by a sealing wax seal and pumped down till the pressure was a few hundred thousandths of a mm. Both pieces of lead

were heated with small flames, but no discharge could be made to pass when the electrodes were connected to an induction coil. On raising the temperature of the lead to cause it to vapourise the glass melted and was drawn in. It was concluded from these experiments that if any thallium were formed it would be possible to detect it spectroscopically at temperatures far below those at which any lead could be detected.

An experiment was then carried out to see whether enough vapour could be produced from thallium by electron bombardment to be detected spectroscopically. A piece of thallium was introduced into an apparatus (fig. III) which was at hand and which satisfied the necessary requirements. F.F. were two lime-coated-platinum filaments about 1 x 5 mms supported horizontally and close to each other in the same plane by platinum leads. G was a circular platinum gauze grid distant about 1 cm below the filaments, on which the thallium was placed. A, was a platinum anode which was not used.

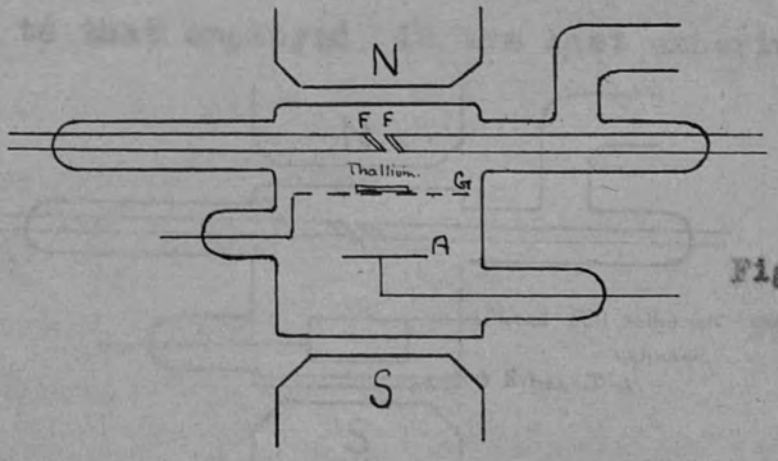

Fig. III.

The tube was sealed on to the pump and pumped down. After testing for leaks the filaments were glowed for some time to get rid of any occluded gas. One of them was then heated and an accelerating field of 100 volts applied between the grid and the negative end of the filament. A faint bluish glow appeared in the tube which increased in brightness as the filament temperature was raised. The tube was placed between the poles of an electromagnet N.S. so that the stream of electrons from the filament should be concentrated on to the thallium. The glow was therefore concentrated in a column between the grid and filament and on examination by means of a spectroscope showed the bright green thallium line and fainter lines and bands which were identified as the hydrogen lines and Angström carbon oxide bands. When the bombardment was stopped it was found that the thallium had melted and alloyed with the platinum grid which had been destroyed.

An investigation was made of the effect of bombarding lead with electrons using a somewhat similar apparatus to that employed in the last experiment (fig IV)

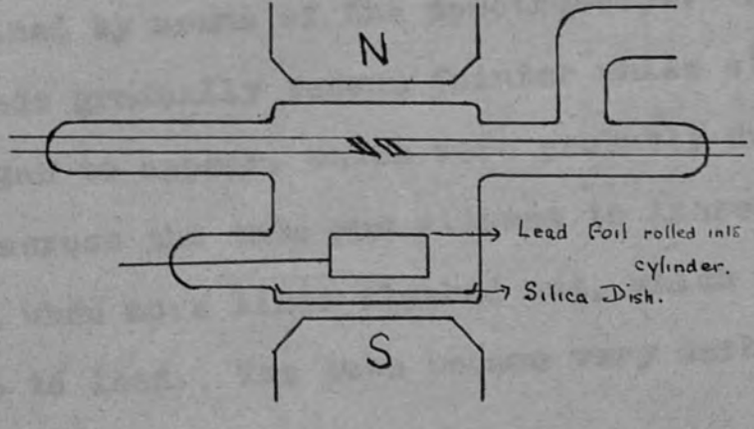

Fig. IV

Two lime coated filaments similar to those used in apparatus fig. III, were sealed into the tube to act as a source of electrons. The anode was made of pure lead foil rolled into a cylinder and supported by a platinum wire. A silica dish covered the bottom of the apparatus so that if the lead melted in the course of the experiment the risk of its cracking the glass tube would be avoided.

After pumping down the tube and glowing the filaments one of the latter was heated and 100 volts applied between filament and anode. As before the apparatus was placed between the poles of an electromagnet in order to concentrate the electron stream. A faint bluish glow appeared in the tube which became brighter when the filament temperature and accelerating voltage were increased. Examination of the spectrum of the glow showed that it was due to hydrogen and carbon oxide.

An attempt was made to get rid of these gases by bombarding the lead gently and pumping. This was continued for several days. At intervals the glow was made brighter by increasing the filament temperature and accelerating field, and examined by means of the spectroscope. The Angström carbon oxide bands gradually became fainter while other lines and bands began to appear, which were probably due to lead. The current across the tube was allowed to increase till an arc occurred when more lines flashed out, which were probably also due to lead. The tube became very dark presumably owing

to deposition of lead on the walls, so that the glow could no longer be seen. Heating the tube in a bunsen flame partly removed the deposit and the glow was observed to exhibit only the Angström bands and hydrogen lines, which had become very bright again. The lead was accordingly bombarded gently and pumped as before for several days, but before any diminution in the brightness of the carbon oxide bands was again observed both filaments burned out.

It was clear from this experiment that chemically pure lead contains occluded gas, chiefly carbon dioxide and hydrogen. This was released from the lead when the latter was bombarded and consequently the energy of the electrons was ultimately expended in ionising the gas so that by the time they reached the lead they would have only a few volts energy.

An attempt was made, therefore, to remove the occluded gas from lead before subjecting it to bombardment. A few small pieces of lead were introduced into a glass tube which was sealed on to a pump. The lead was heated gently till molten and the tube pumped down till no pressure was registered on the McLeod Gauge the lead being allowed to cool and remelt several times. After some time the whole tube blackened, and when it was cut down and the lead removed, the latter was covered with a black fluffy powder, which however was easily rubbed off leaving a bright surface. The metallic globule was cut into several pieces and melted in vacuo into a glass tube

drawn out and sealed off at one end so as to form a thin cylinder about 2 mms in diameter by 2 or 3 cms long. After cooling, the lead was removed and used as the anode in an apparatus exactly similar to that shown in fig. IV.

On bombarding with electrons of 100 volts energy, a faint glow appeared at first, but vanished after bombarding for some time and pumping. On increasing the accelerating field to 440 volts, however, a bluish glow appeared after a short time and spectroscopic examination showed the presence of hydrogen and oxides of carbon as well as other lines and bands. While endeavouring to examine the arc spectrum the apparatus cracked. No sign of the thallium line was detected.

It is probable that during the foregoing experiments no electrons struck the lead with more than a few volts energy for any considerable time. Before any adequate test of the possibility of transmutation could be made along these lines precautions would have to be taken to ensure that the energy of the electrons was not expended in ionising occluded gas in the lead or lead vapour. The difficulty of realising this experimentally has not yet been overcome.

The element lead was chosen for the foregoing investigation because there was a possibility that it was isobaric with thallium, and that the latter might be produced

more easily if isobars of lead and thallium existed. Such isobars have not been definitely established. An attempt was therefore made to effect transmutation of lithium into helium, elements for which isobars are not known to exist naturally, but whose properties lend themselves more readily to the conditions of the experiment.

Lithium itself cannot be used because of its low melting point and reactive character. Lithium oxide has been selected as a compound which does not vapourize till an exceedingly high temperature, although the presence of another element is a disadvantage since it lessens the number of possible hits on lithium atoms.

The great advantage of the use of lithium is that the product helium is gaseous at all temperatures which are readily attainable, so that if a carbon tube surrounded by liquid air is attached to the experimental tube everything except helium and some hydrogen will be absorbed. If any helium is produced, therefore, it should readily be detected.

Another experimental advantage in the use of lithium is that with the electric fields available an electron can be given enough energy to strip the lithium nucleus of electrons and this will possibly increase the chance of its reaching the nucleus.

Lithium oxide was selected as being the most suitable compound of lithium to use, firstly because it is

unaffected by heat till 1000° C. when it sublimes and secondly because the proportion of lithium to oxygen is as great as possible namely 2 : 1. For preliminary experiments it was prepared by dissolving metallic lithium in water, evaporating the product to dryness and igniting on a small platinum dish. The latter covered by a thin layer of a substance which contained a large proportion of lithium was used as the anode in the apparatus shown diagrammatically in fig. V which resembles the other tubes which have been used in this investigation.

The carbon tube was heated and the apparatus pumped out till the pressure registered by the McCleod gauge was a few hundred thousandths of a mm. Both filaments were

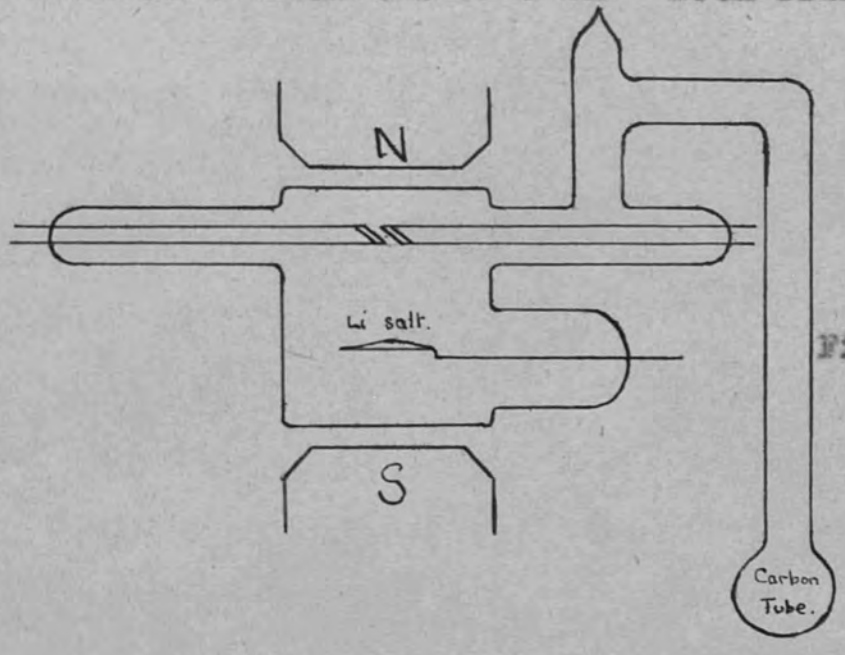

Fig. V.

glowed for some time to remove occluded gas and the tube was then sealed off, in order that any helium which might subsequently be produced would be contained in as small a volume as possible and therefore be more readily detected with the spectroscope.

The effect of applying a much larger voltage in the same two arrangements was investigated at ·56 amps.h.c. In these circumstances deactivation was found to occur in both cases. With arrangement A very rapid deactivation occurred and a limiting value was reached in five minutes, but for arrangement B deactivation was much slower and the limiting value was much higher and was not attained till about an hour after the application of the voltage. Time current curves plotted from observations taken in the two cases are shown on graph IX.

In arrangement B with 600 volts applied at ·56 amps h.c. the anode became red hot after a few seconds and cooled slowly as the emission decreased. Whilst the anode was getting hot the emission decreased more slowly than it did later, giving a discontinuity in the time emission current curve (graph IX). No heating was visible when the filament was deactivated using arrangement A with the same voltage and heating current. (This point is referred to again later see p. 15.)

Another point of difference between the deactivation curves is that the initial emission for arrangement A is much larger than that obtained with arrangement B for the same voltage and temperature as may be seen from graphs VII VIII and IX, although the initial emission under

standard conditions had been the same. This may be ascribed to the fact that saturation is not reached in arrangement B till a much higher voltage has been applied than is necessary to produce saturation in arrangement A (graph X).

To ascertain whether this difference of emission was a determining factor in the difference in deactivation observed for the two arrangements, the heating current was decreased so that the initial emission in arrangement A was nearly equal to that in arrangement B, and observations of time and emission were plotted (graph XI). The resulting curves show that even when the initial emissions are not very different there is a great difference in the rate of deactivation. With arrangement A the rate of deactivation at a particular voltage and heating current is much faster than the rate when the same voltage is applied in arrangement B at the same heating current.

Further evidence that initial emission is not the main deciding factor in deactivation was obtained from the following experiments. From a series of characteristic curves (anode - current - grid - volts curves) plotted for different values of the anode voltage it was found that the amplification factor of the valve is 10, that is, if on the straight part of the characteristic for a particular anode voltage, an emission corresponding to a particular grid

voltage is selected, the same emission is given when the anode voltage is increased by 10 and the grid voltage decreased by 1. The emission along the straight part of the characteristic may therefore be written as some function of $V_A + k V_G$ where V_A and V_G represent anode and grid voltages and k is the amplification factor, in this case equal to 10. A value of V_A was selected so that very slight deactivation occurred when V_G was zero, and observations of the current were taken at intervals. After reactivation V_A was increased by 100 and V_G decreased by 10 when another series of observations was taken and so on. The observations are plotted on graphs XII and XIII, XII giving curves of series of observations taken at a heating current of ·50 amps and XIII curves taken at ·55 amps h.c. If deactivation were a function mainly of emission, the time current curves should be coincident at each heating current. Graphs XII and XIII both show, however, that the time current curves are not coincident. In both series deactivation increases for increasing anode voltage and the effects are more marked at the higher heating current. These observations show that deactivation does not depend only on the value of the initial emission.

The fact that the anode became red hot when a voltage of 600 was applied in arrangement B (see p.13), together with the fact that less deactivation occurred with

arrangement B than with arrangement A for the same voltage and heating current, suggests that deactivation cannot be caused by the action on the filament of atoms of any gaseous impurity released from the anode by bombardment. Such a possibility is unlikely in view of the special treatment undergone by the valves during manufacture in order to eliminate such impurities, but these observations provide confirmatory evidence that deactivation is probably not due to this cause, since if it were, more deactivation would be expected in arrangement B than in arrangement A.

Apart from the effect of a high temperature another possibility which may cause decrease in emission is according to Kingdon and Langmuir(5) the bombardment of the filament by positive ions. Horton and Davies(6) have detected a positive ion current from a platinum surface when this is bombarded by relatively slow moving electrons at pressures which cannot be detected on the McCleod gauge and have shown that it is probably due to positive ions of the material of which the electrode is composed. It is possible, therefore, that in the present case sufficient positive ions are produced from the electrodes by electron bombardment to be responsible for the decrease in emission observed. Experiments were

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(5) Kingdon and Langmuir Phys. Rev. 20. 1922. 108.

(6) Horton and Davies Proc. Roy. Soc. 1920. (A). 97. 23.

therefore carried out to see whether a positive ion current could be detected under any circumstances.

A negative voltage of 10 was applied to the anode of a valve whose filament was activated and the grid voltage increased from 0 in small stages. (arrangement C. p.29.). The negative current collected by the grid was measured on a milliammeter and a sensitive galvanometer was included in the anode circuit. The direction of deflexion which corresponded to a negative current was ascertained by shunting the galvanometer and allowing electrons from the filament to get over to the anode. As the grid voltage was increased (as in arrangement C) a slight deflexion was observed on the galvanometer indicating a positive current, which increased with increasing grid voltage. When the latter had reached about 80 volts, however, the direction of deflexion of the galvanometer changed showing that electrons were reaching the anode. The anode voltage was accordingly lowered to - 30, for which value no change in the direction of deflexion of the galvanometer was observed for any value of the grid voltage applied. The latter was increased up to 100 volts, when decrease of the negative current, indicating deactivation of the filament set in. The positive current was inappreciable till the grid voltage had a value of about 30 after which it increased rapidly with the grid voltage (graph XIV), the emission current being that obtained at a heating current of ·56 amps. The magnitude of the positive ion current detected after the grid voltage had

been increased to about 90 volts was of the same order as those found by Langmuir to be responsible for deactivation in his investigations, namely 10^{-7} amps. This observation, therefore, justifies, the suggestion that deactivation is due to positive ion bombardment of the filament.

With the arrangement which has just been described (C) if positive ions were derived from the electrodes they must have come from the grid since no electron bombardment of the anode occurred. The positive current actually measured on the anode must have been due to positive ions removed from the outer surface of the grid, while those returning to the filament and causing deactivation must have been derived from the inner surface of the grid. If therefore more positive ions could be removed from the inner surface of the grid deactivation would be expected to be greater, and this result would presumably be achieved if more electrons hit the under side of the grid.

Investigations were carried out to see whether deactivation would vary when different fractions of the emission were collected by the grid. Using a valve as in arrangement A, and applying 120 volts to the anode the current collected by the anode and the total emission current were observed at intervals. (graph XV) Similar observations were taken with grid voltage still 120 and anode voltage reduced to 33 (graph XVI). Graph XV shows that when grid and anode

are at the same potential with respect to the filament, about 9/10 of the total current is collected by the anode and 1/10 by the grid. When the anode voltage is reduced to 33 however, about 6/10 of the total current is collected by the anode and 4/10 by the grid (graph XVI). The deactivation produced in the filament, measured by the decrease of the total negative emission is practically identical in the two cases. This suggests that 3/10 of the total current striking the grid was as effective as 9/10 striking the anode, for in the arrangement with the anode negative to the grid, positive ions removed from the anode would be in a reverse field and would not reach the filament while in the case where grid and anode are at the same potential positive ions removed from the anode would be able to reach the filament. It appears therefore that for a given bombardment of the grid more positive ions reach the filament than for the same bombardment of the anode.

To investigate this point further, experiments were made with another valve keeping the grid potential at 120 and giving the anode a voltage of +100 in one case and -30 in the other. The total currents and anode currents were observed in both cases, with the first arrangement a negative current being measured on the anode and with the second a positive current. These observations are plotted

on graphs XVII and XVIII. Since in both cases the anode is negative to the grid no positive ions will reach the filament from the anode in either case, but they are able to reach the filament from the grid. Deactivation would be expected to be favoured by an arrangement of fields which is likely to cause more electrons to hit the inner surface of the grid. This is actually found to be the case (graphs XVII and XVIII). The curves show that much greater deactivation is obtained when all the electrons are collected by the grid and when it is probable that many of them strike the inner surface.

The observations of the amount of deactivation obtained for various arrangements of fields support the idea that deactivation is due to the removal of positive ions from the electrodes.

Returning to graphs XV and XVI and comparing with XVII it appears that the deactivation indicated by XVII is much slighter than in XV and XVI. Such differences in curves taken under similar conditions with different valves have been observed before (see p. 11) and are possibly to be ascribed to a difference in concentration of thorium within the filament.

It would also be expected from the distribution of current between anode and grid shown by graphs XV and XVI that the anode current shown by graph XVII would be greater than 2/3 of the total. For, the anode voltage in the

observations from which graph XVII is plotted is intermediate in value between the anode voltages in the observations from which graphs XV and XVI were plotted, while other conditions are the same. The fraction of current collected by the anode for the intermediate voltage would therefore be expected to be between 6/10 and 9/10 and be nearer the value 9/10. From the amount of deactivation observed in these three cases it has been concluded that there is a difference in distribution of thorium under the surface of the filament in the two valves. This suggests that differences in the distribution of the negative current may be due to this cause, although no reason suggests itself as to why this should be so.

Further evidence of differences in the distribution of the negative current between grid and anode when the only apparent difference has been a difference in concentration gradient of the thorium was obtained from the following experiment. The valve by the aid of which graph XV was plotted, was connected up and its emission observed under exactly the same external conditions as those previously employed, the only difference being that the maximum emission under standard conditions was 6.m.a. instead of 10.m.a. The second series of observations are plotted on graph XIX and in the latter case the ratio of the current collected by the anode to the current collected by the grid is 4:1 while in the former case (graph XV) the ratio was 9 : 1.

Up to the present, since the idea of deactivation by bombardment of the filament by positive ions was introduced, the experimental arrangements which have been considered have been such that any positive ions from the electrodes causing deactivation must have been derived mainly from the grid. In previous experiments, however, namely those with arrangement B with 600 volts applied (graph IX), the positive ions responsible for the deactivation must have been derived from the anode, for since the grid in this arrangement is at the same potential as the filament very few, if any electrons will be collected by it. Any positive ions removed from the anode will return either to the grid or to the filament, those reaching the latter causing deactivation. If more positive ions were collected on the grid and fewer went to the filament less deactivation would be expected to occur and the following experiment shows this to be the case.

Observations of the negative current were taken at intervals for a valve with 600 volts applied to the anode and -10 to the grid (arrangement D) p. 29. and compared with observations of the negative current with time for 600 volts applied in arrangement B the initial total emission being the same in both cases. These observations are plotted on graph XX and show that less deactivation occurs for conditions where fewer positive ions would be expected to strike the filament.

As a result of the preceding experiment it was thought that it might be possible to prevent all the positive ions from returning to the filament by giving the grid a sufficiently large negative voltage. Earlier experiments with arrangement D with a different object in view (p. graphs XII and XIII cf. with graphs II and III) had also shown that it was possible to reduce deactivation of the filament by applying a negative voltage to the grid. An investigation was therefore made to see whether by a suitable arrangement of fields, positive ions could be collected on the grid, while no deactivation was observed in the filament. Accordingly a valve was connected up as in arrangement D and different negative voltages applied to the grid during each run. The variations of positive and negative currents with increasing anode voltage were observed. In every case deactivation ultimately occurred, but not till a higher anode voltage for a larger negative grid voltage. The relations between voltages and positive and negative currents before deactivation set in are shown on graphs XXI and XXII. The ends of the curves indicate the voltages for which decrease in negative current was observed during the time required to take the observation.

The failure to prevent deactivation of the filament occurring may be due either to the open structure of grid or possibly to the production of positive ions from thorium atoms in the space between the grid and filament.

As in graph XIV the positive current is increasing very rapidly with increasing voltage at the moment when deactivation sets in, and in order to obtain some insight into the behaviour of the positive current under conditions for which deactivation could first be detected, a more careful examination was made of the variation of the positive and negative currents with time for these conditions.

Connecting the valve up as in arrangement D the filament temperature was maintained constant as before, and the grid voltage given various values in successive runs. The anode voltage was increased slowly starting a little below the value for which deactivation was expected to occur, knowledge of which was obtained from the previous experiment. When a decrease was observed in the negative current the anode voltage was maintained constant and the variation of positive and negative currents with time observed. The observations are plotted on XXIII and XXIV. The curves show that the positive current goes on increasing after the negative current has begun to decrease, which suggests that positive ions are being produced by some secondary action. During the time in which the positive current is increasing the negative current is decreasing but at a much slower rate than subsequently occurs. Such flat portions have been observed before in time-emission curves (see graphs II III IV VI VII IX XI XII and XX), and

are in agreement with observations by Kingdon and Langmuir on the deactivation of thoriated tungsten filaments by bombardment of the filament by positive ions derived from certain gases.

Finally, experiments were carried out to investigate whether any deactivation occurred after leaving a valve with a very low voltage applied for a long time. According to Langmuir bombardment of thoriated tungsten filaments by positive ions causes removal of thorium. The equilibrium condition of the surface (p.3) is therefore destroyed and a new equilibrium condition set up where the rate of diffusion of thorium to the surface is equal to the rate of removal by evaporation and bombardment. Although the rate of diffusion alters very slightly with time, yet since thorium is being continually removed from the surface of the filament, a time must come when the concentration gradient of the thorium has diminished. The rate of diffusion will then become less and decrease in the emission will be observed. If deactivation is produced by the removal of thorium from the surface of the filament by positive ion bombardment it is possible that this process occurs at low voltages, but that it cannot be detected ordinarily owing to the immediate replacement of the thorium atoms so removed by others from the interior, If, however, the concentration gradient of the thorium were low, after leaving for some time with a small voltage applied, deactivation would be expected to occur. If then, the voltage was cut off the deactivation would be expected either to cease

or to become much slower. To test this, a valve whose filament was in an active state was connected up with the grid 3 volts positive to the filament and the anode 80 volts positive to the filament. These voltages were selected as being far below those hitherto found necessary to cause deactivation in similar arrangements (cf. arrangement B+graphs VIII and IX). After heating for 21 hours at ·50 amps. h.c. no appreciable decrease in emission was observed. The anode voltage was then increased to 194 and heating continued for a further 93½ hours but still no deactivation occurred. Subsequently, however, a similar experiment was carried out when the concentration gradient of the thorium within the filament was low and deactivation with low voltages could then be detected.

In the experiment just described the filament was in an activated state and giving a maximum emission of 8.m.a. The later experiment was carried out with a valve whose filament was giving under standard conditions a maximum of 4.m.a. It was connected up as in arrangement B and the anode was made 80 volts positive to the filament. On leaving for some time with voltage applied decrease in emission was observed, but in leaving for a further period with voltage cut off, an increase in emission was observed. The voltage was applied and cut off alternately for varying periods and the observations are given in Table I. These numbers show

that there is a definite decrease in emission during the period for which the voltage is applied, and this admits of an explanation on the theory of bombardment of the filament by positive ions.

Table I.

Time volts applied.	Time Volts Cut off.	Emission
		4·05 m.a.
21¼ hours		1·78 m.a.
	10¼ hours.	2·08 m.a.
·2¼ "		2·18 "
	11. "	2·7 "
11¾ "		1·7 "
	12¼ "	1·98 "
47¾ "		1·08 "
	12 "	1·4 "
36 "		1·08 "

The results so far obtained in this investigation may all be explained by the theory of bombardment of the filament by positive ions, which are probably derived mainly from the electrodes. Deactivation appears to be a complicated function of several factors and there is scope for much more investigation in this subject, in connexion with which further experiments are in progress.

Summary.

The experiments described in this paper indicate that the deactivation of thoriated tungsten filaments in vacuo by the application of large voltages, is probably due to positive ion bombardment of the filament.

The main reasons for these conclusions are:-

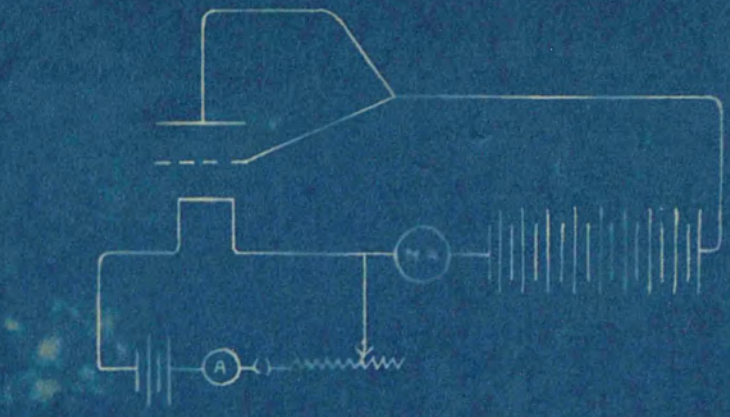
Firstly, that a positive ion current can be detected in the valve of the same order of magnitude as that of the positive ion current which Kingdon and Langmuir observed when deactivation of thoriated tungsten filaments was produced in the presence of gases.

Secondly, the majority of time-emission curves obtained during this investigation show an initial flat portion similar to that obtained by Kingdon and Langmuir in their experiments.

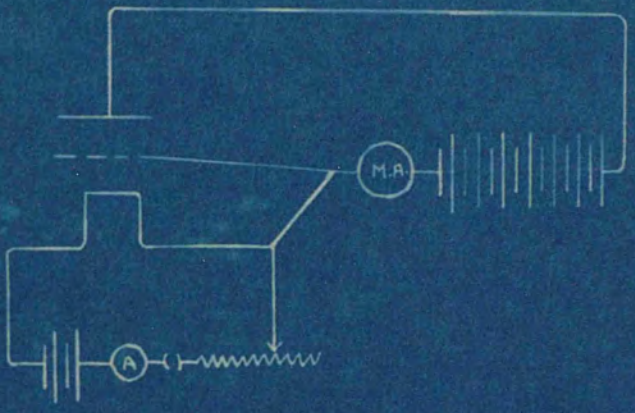
Thirdly, this hypothesis can explain practically all the experimental results so far obtained.

The main sources of the positive ions are probably the grid and anode, but it is possible that some may be derived from other sources, perhaps thorium vapour.

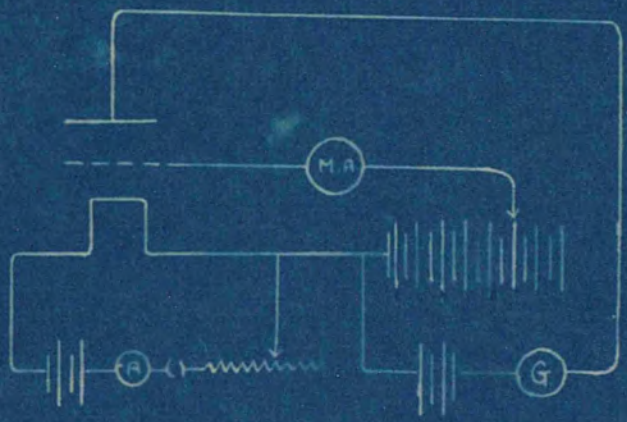
Deactivation by the application of relatively high voltages to thoriated tungsten filaments appears to be a function of the filament temperature, applied voltage, and condition of the surface and interior of the filament.



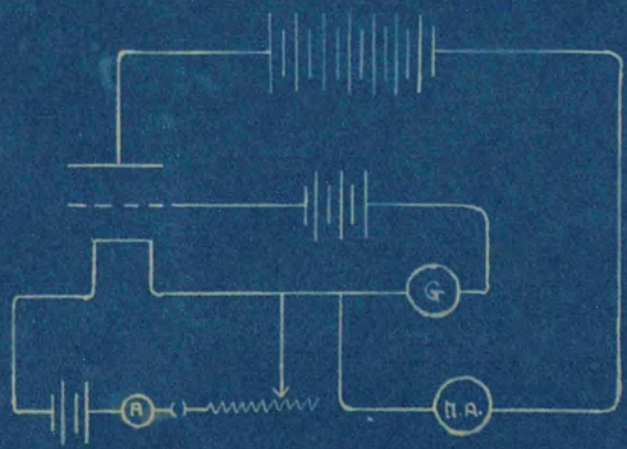
A



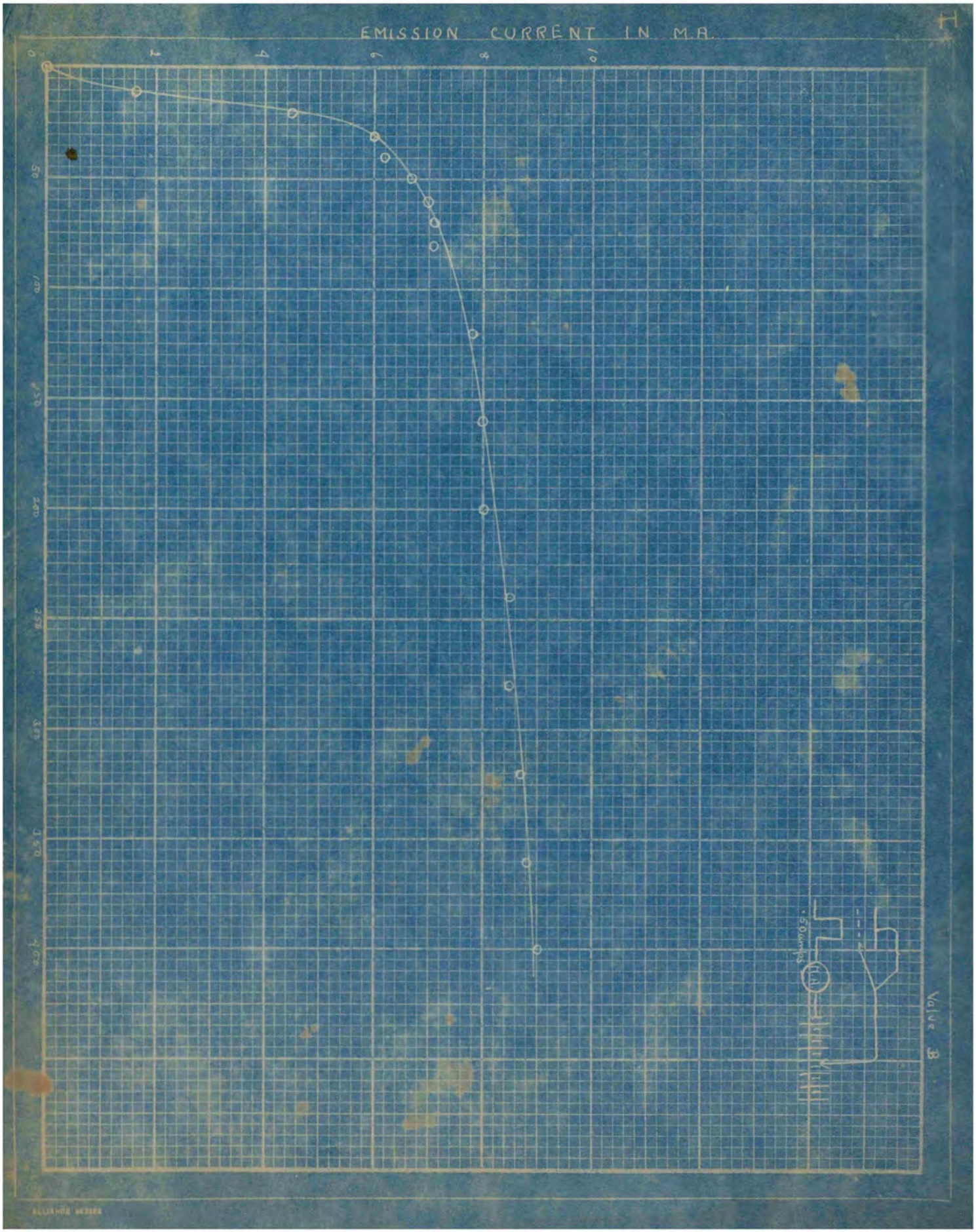
B

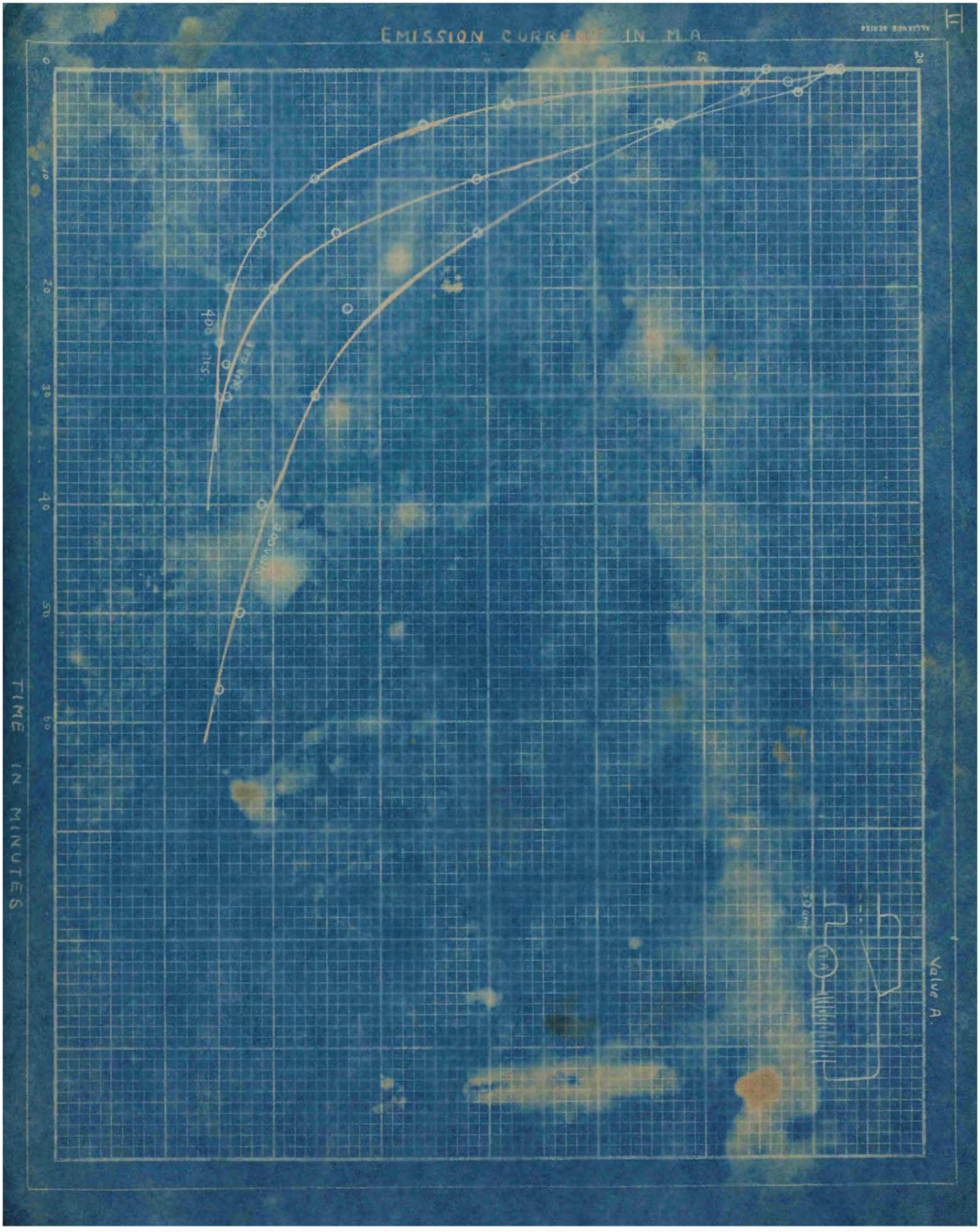


C



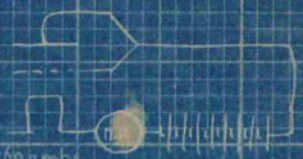
D


EMISSION CURRENT IN M.A.
0
2
4
6
8
10
50
100
150
200
250
300
400
Valve B


EMISSION CURRENT IN M.A.
TIME IN MINUTES
400 Volts
300 Volts
200 Volts
Valve A.

III

Valve A



EMission CURRENT IN MA

8
7
6
5
4
3
2
1
0

100 volts

10
20
30

TIME IN MINUTES.

IV

EMISSION CURRENT IN M.A.

0 5 10 15 20

10 20 30 40 50

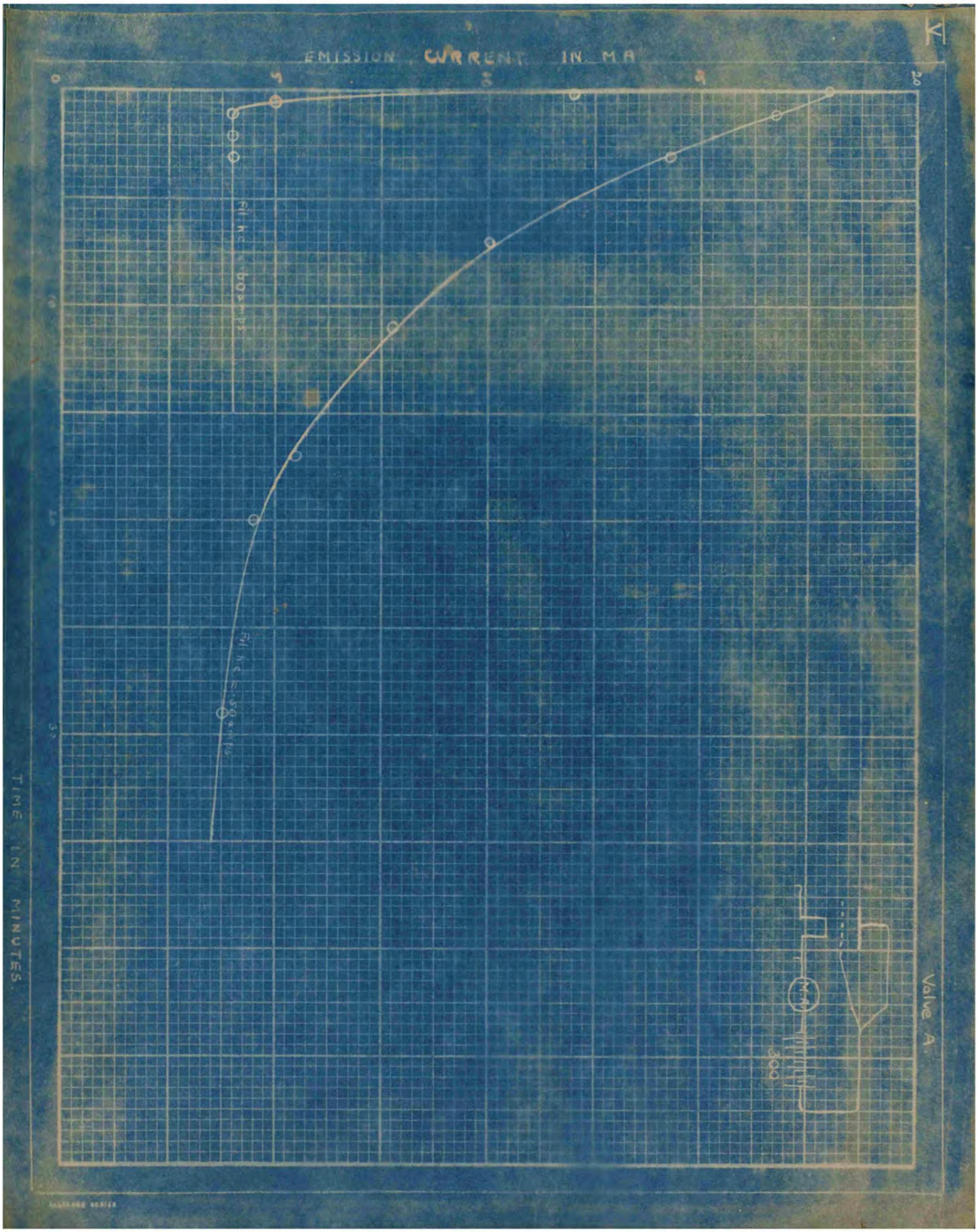
TIME IN MINUTES

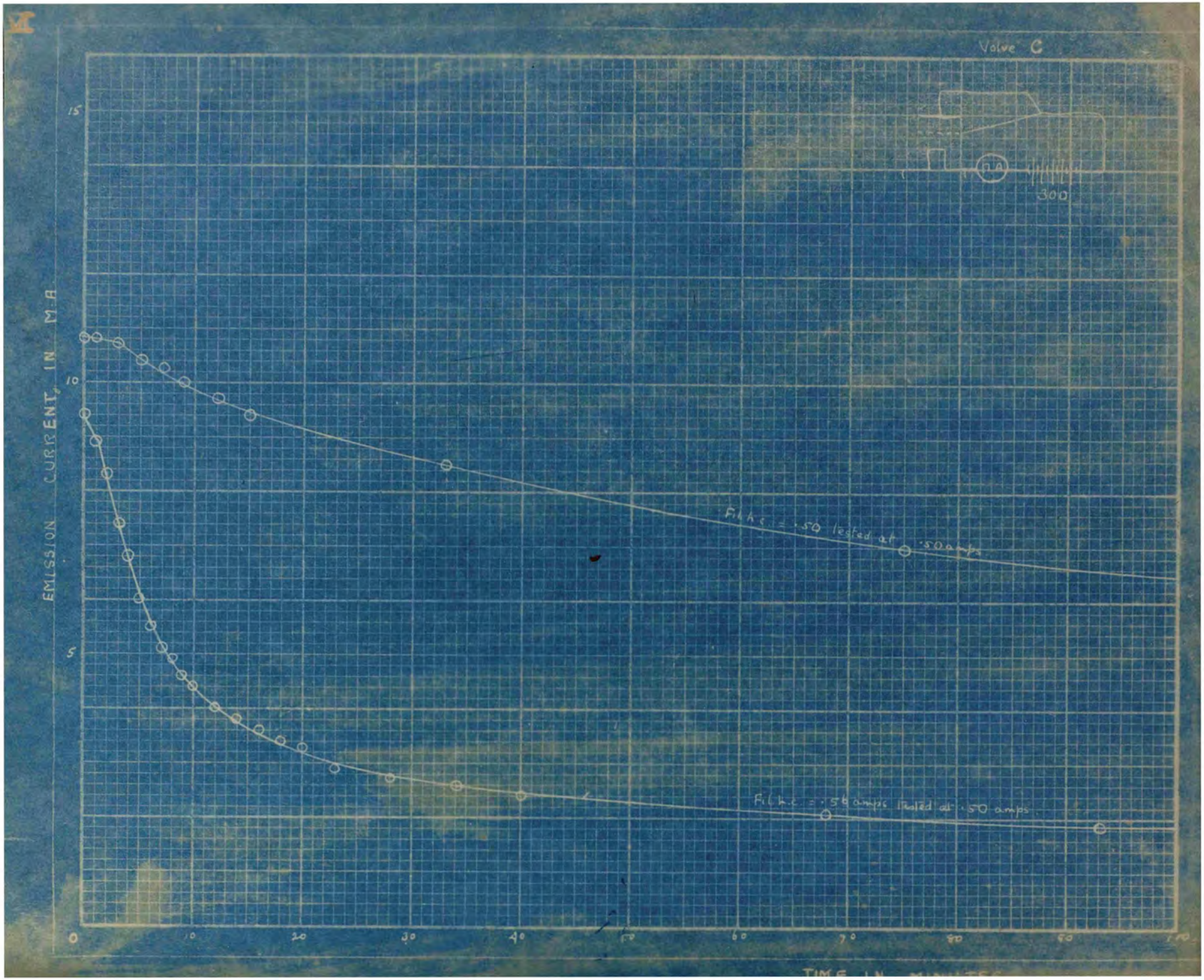
200 volts at .60 amps h.c.

5.0 amps

Valve A.

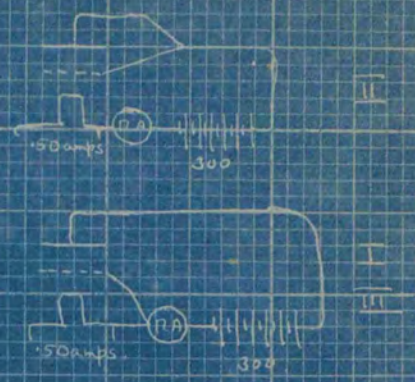
ALLIANCE SERIES


EMISSION CURRENT IN MA
TIME IN MINUTES.
Volve A.
300


Valve C
300
EMISSION CURRENT, IN M.A
15
10
5
0
10
20
30
40
50
60
70
80
90
100
Fil h.c. = .50 tested at .50 amps
Fil h.c. = .56 amps tested at .50 amps
TIME IN MINUTES

VII

Valve C



EMISSION CURRENT IN M.A

15

10

5

0

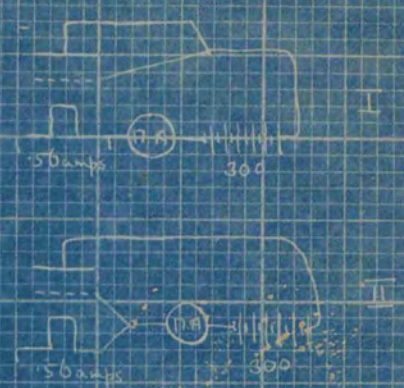
60

120

TIME IN MINUTES

VIII

Valve C



EMISSION CURRENT IN M.A.

30

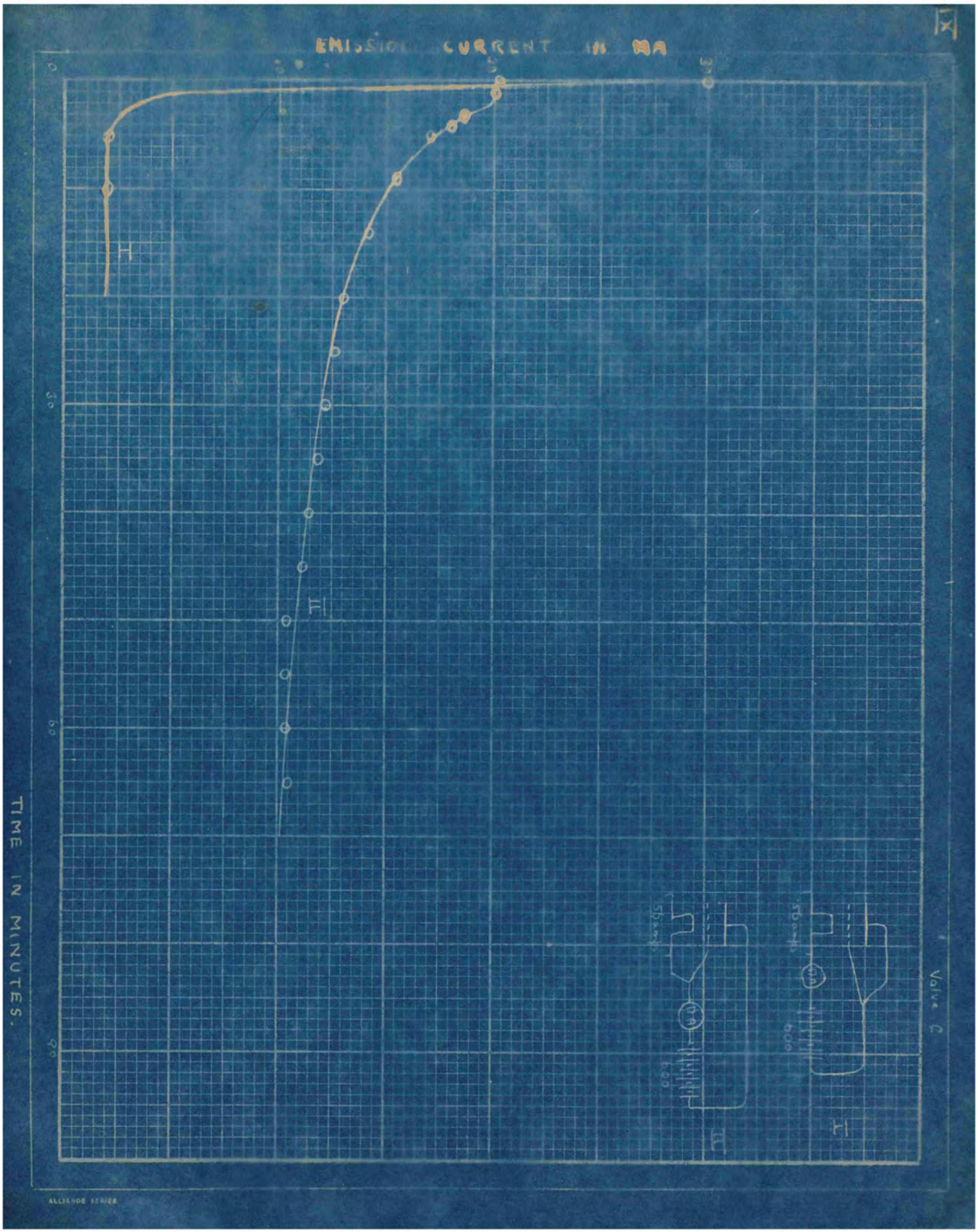
20

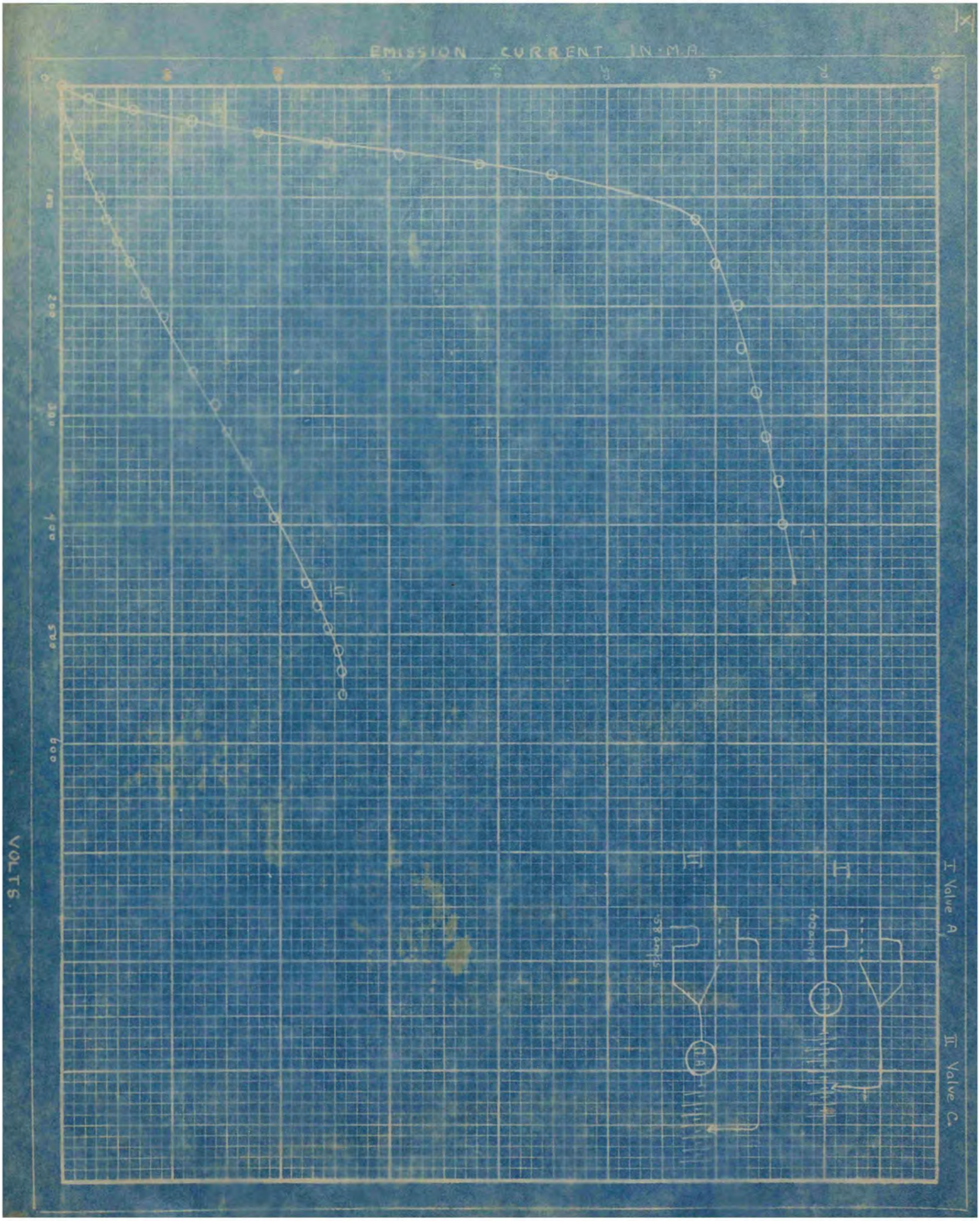
10

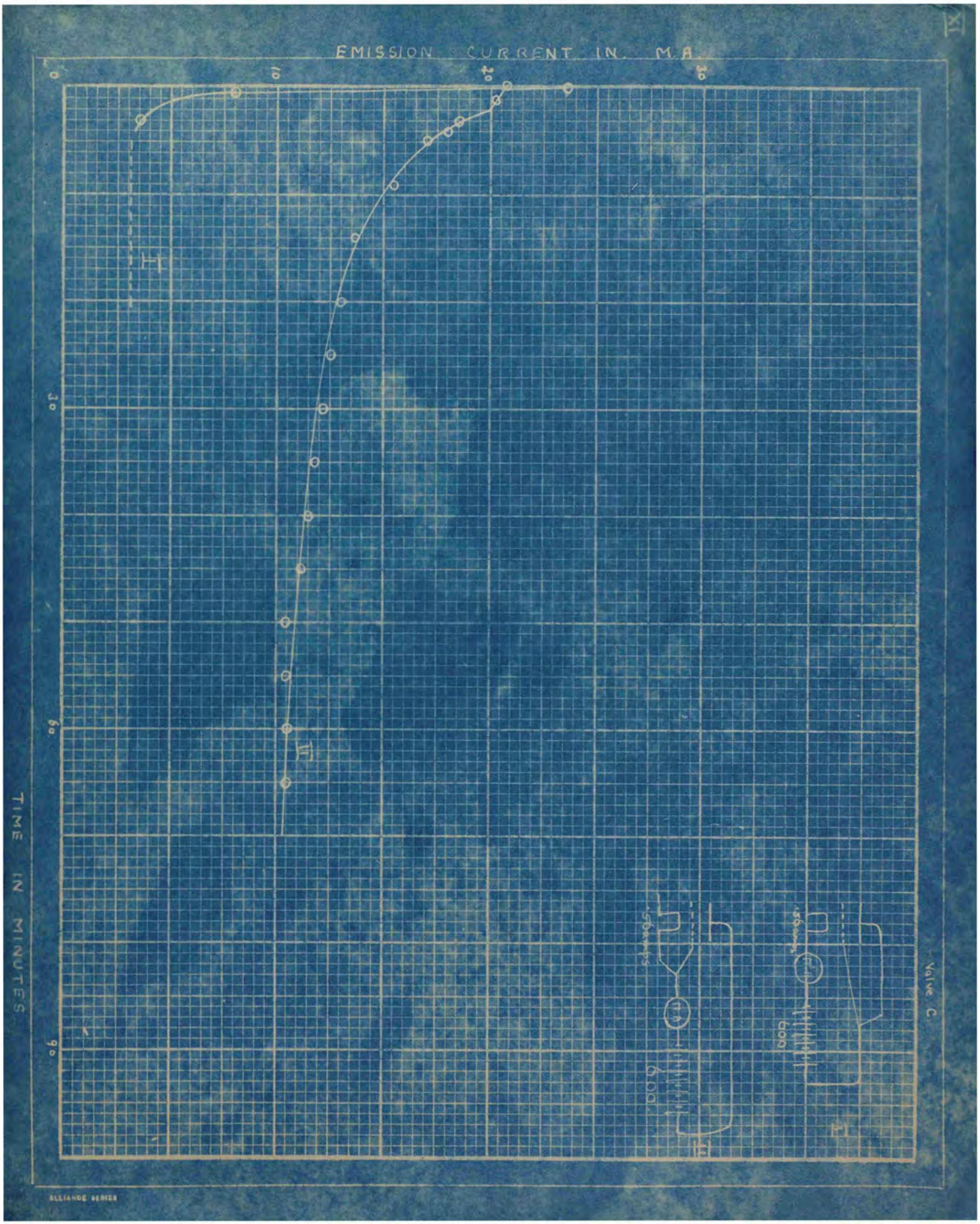
0

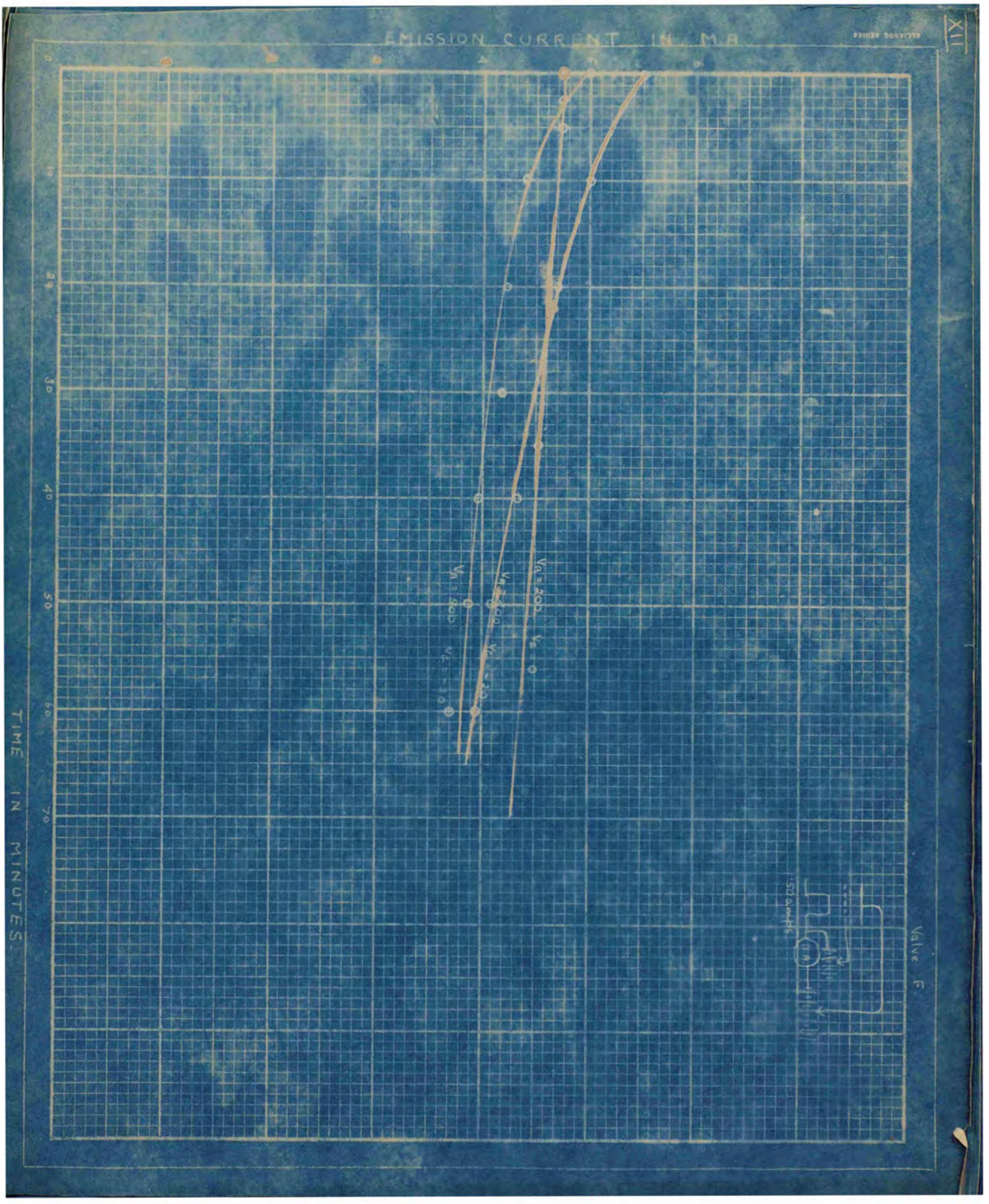
30

TIME IN MINUTES


EMISSION CURRENT IN MA
TIME IN MINUTES.
H
Fl
Valve C
ALLIANCE SERIES


X
EMISSION CURRENT IN M.A.
VOLTS.
I Valve A.
II Valve C.


EMISSION CURRENT IN M.A.
0
10
20
30
TIME IN MINUTES
60
90
I
II
Valve C.
ALLIANCE SERIES


EMISSION CURRENT IN MA
XII
TIME IN MINUTES
Vg1 = 200 Vg2 = 0
Vg1 = 200 Vg2 = 200
Vg1 = 200 Vg2 = 30
150 OHMS
Valve F

XIV

EMISSION CURRENT IN M.A.

0 10 20 30

20 40 60 80 100 120

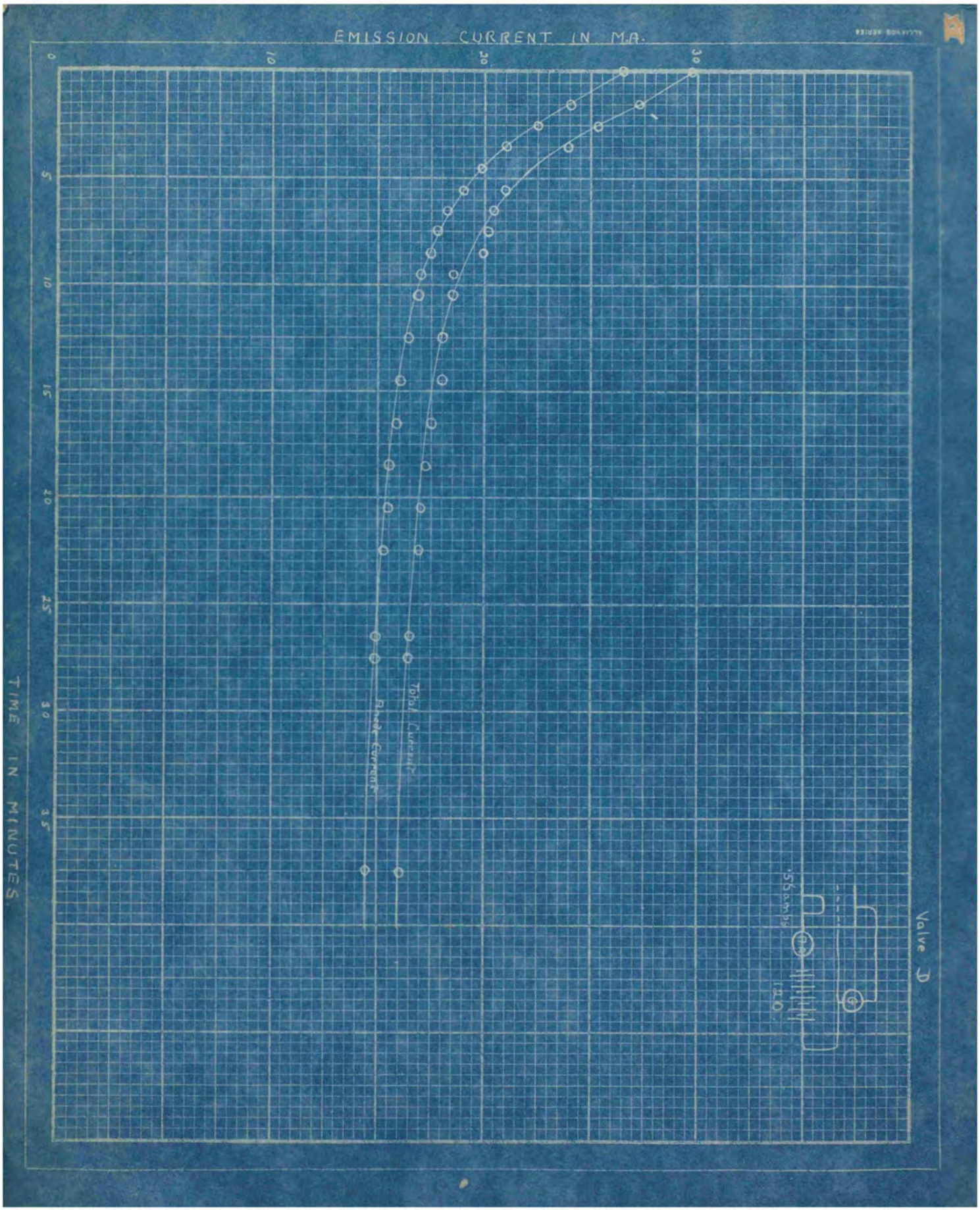
VOLTS

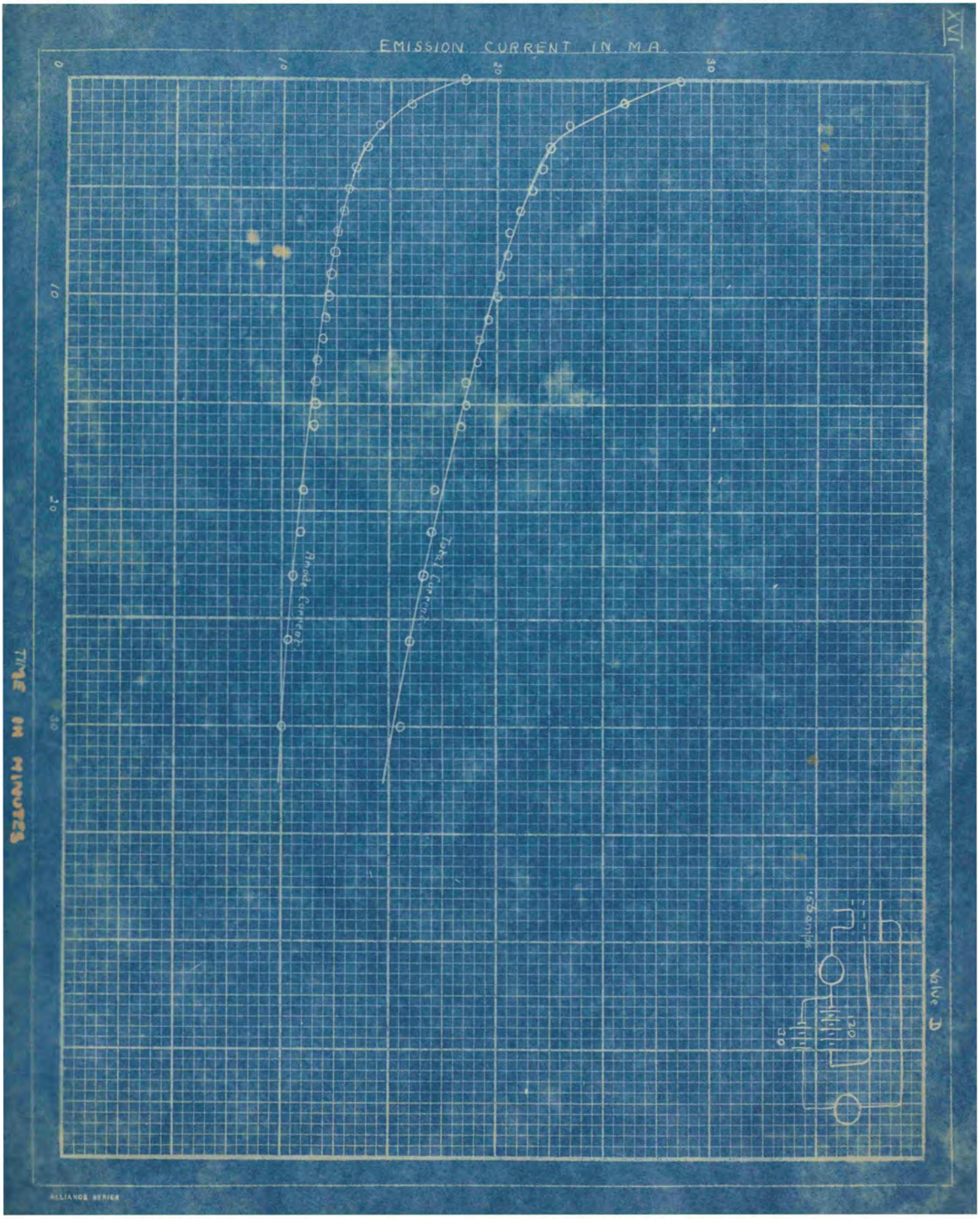
Negative current

Positive current

Valve C.

POSITIVE CURRENT IN AMPS X 10^{-8}


EMISSION CURRENT IN MA.
0
10
20
30
5
10
15
20
25
30
35
TIME IN MINUTES.
Total Current
Anod Current
.56 amps
120
Valve D


EMISSION CURRENT IN M.A.
0
10
20
30
40
50
TIME IN MINUTES
Anode Current
Total Current
Valve A
Grid
Filament
Anode

XVII

EMISSION CURRENT IN M.A.

0 10 20 30

TIME IN MINUTES

0 10 20 30 40

Anode Current

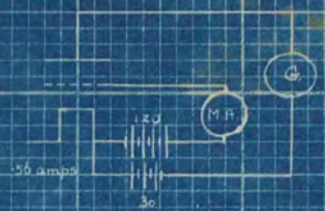
Total Current

Valve F1

ALLIANCE SERIES

XVIII

Valve E



EMISSION CURRENT IN M.A.

30
25
20
15
10
5

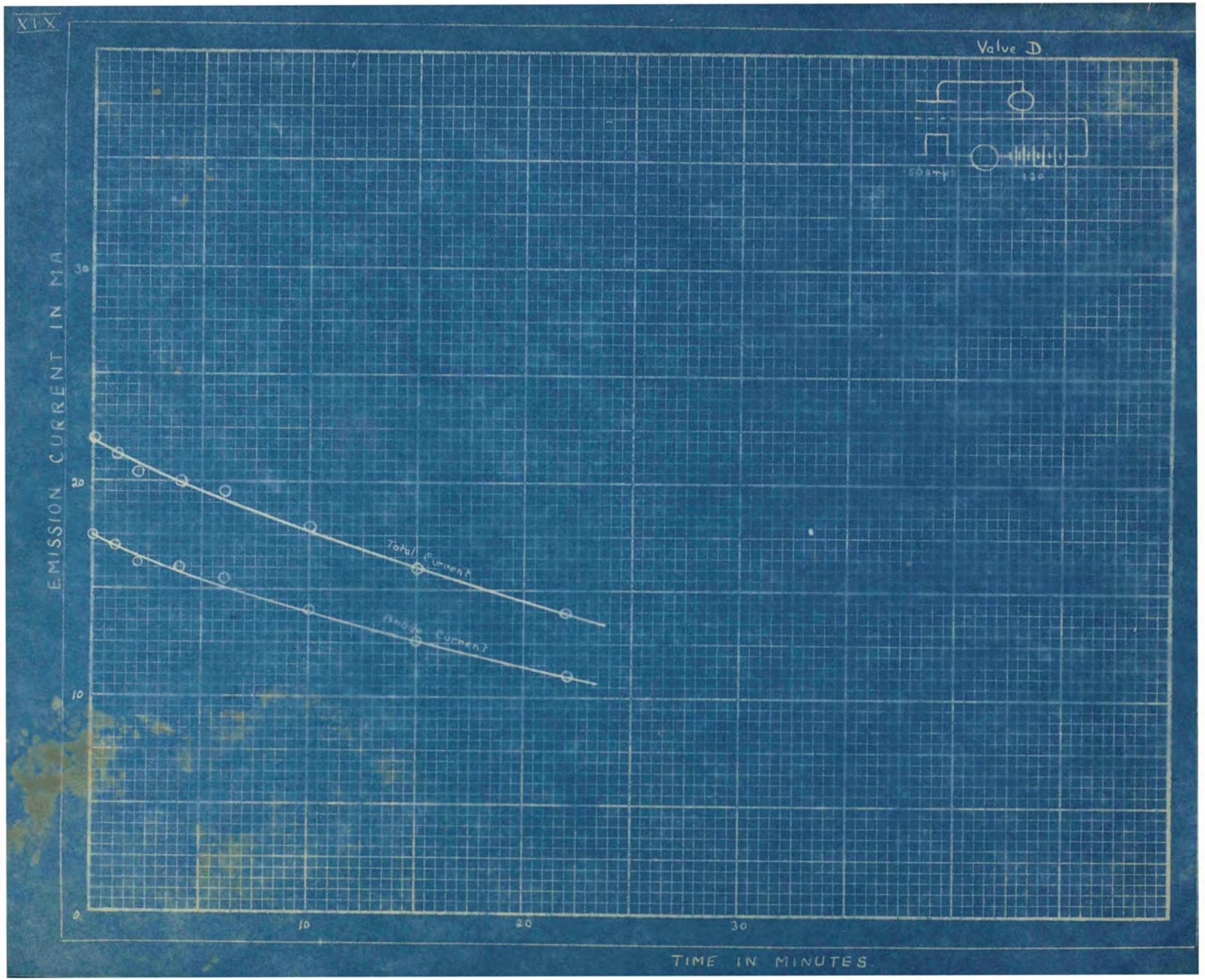
negative current

positive current

0
10
20
30
40

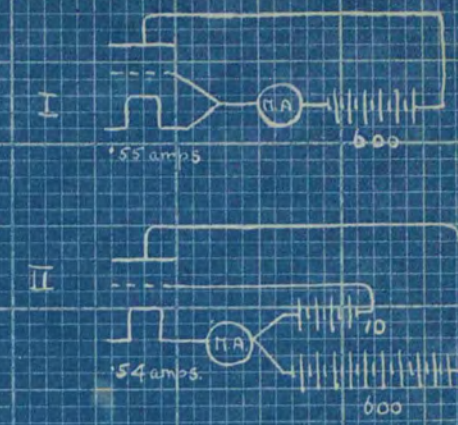
TIME IN MINUTES

XIX


Valve D
50 amps
+30
EMISSION CURRENT IN MA
30
20
10
0
Total Current
Anode Current
10
20
30
TIME IN MINUTES

XX

Valve E



EMISSION CURRENT IN M.A.

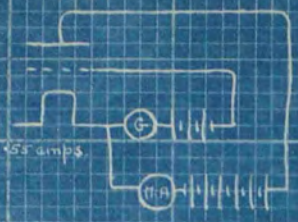
20
15
10
5
0

20
40
60
80
100
120
140
160
180
200

TIME IN MINUTES.

XXI

Valve F.



EMISSION CURRENT IN M.A.

0 2 4 6 8 10 12 14

100 200 300 400 500 600

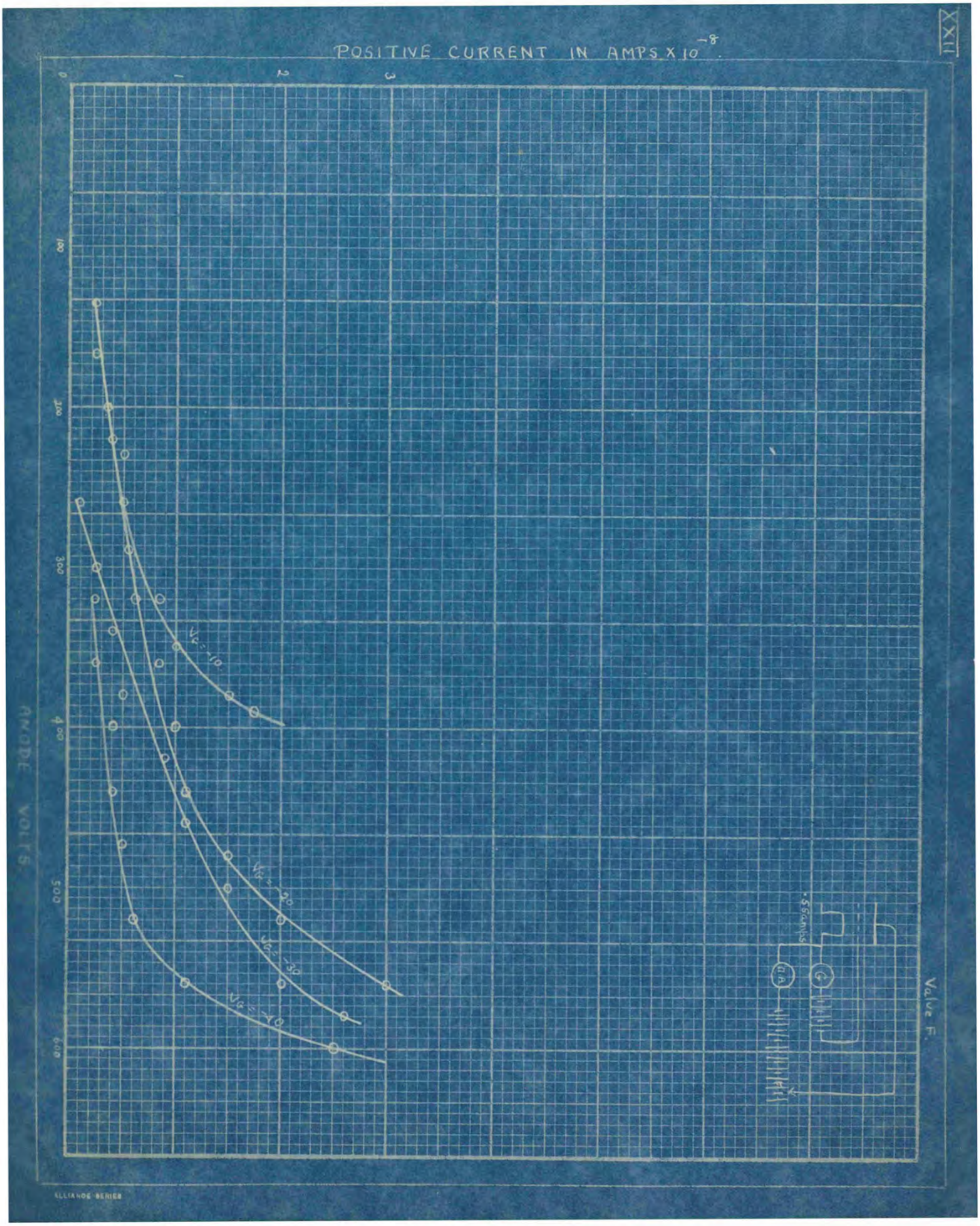
ANODE VOLTS.

$V_c = -10$

$V_c = -20$

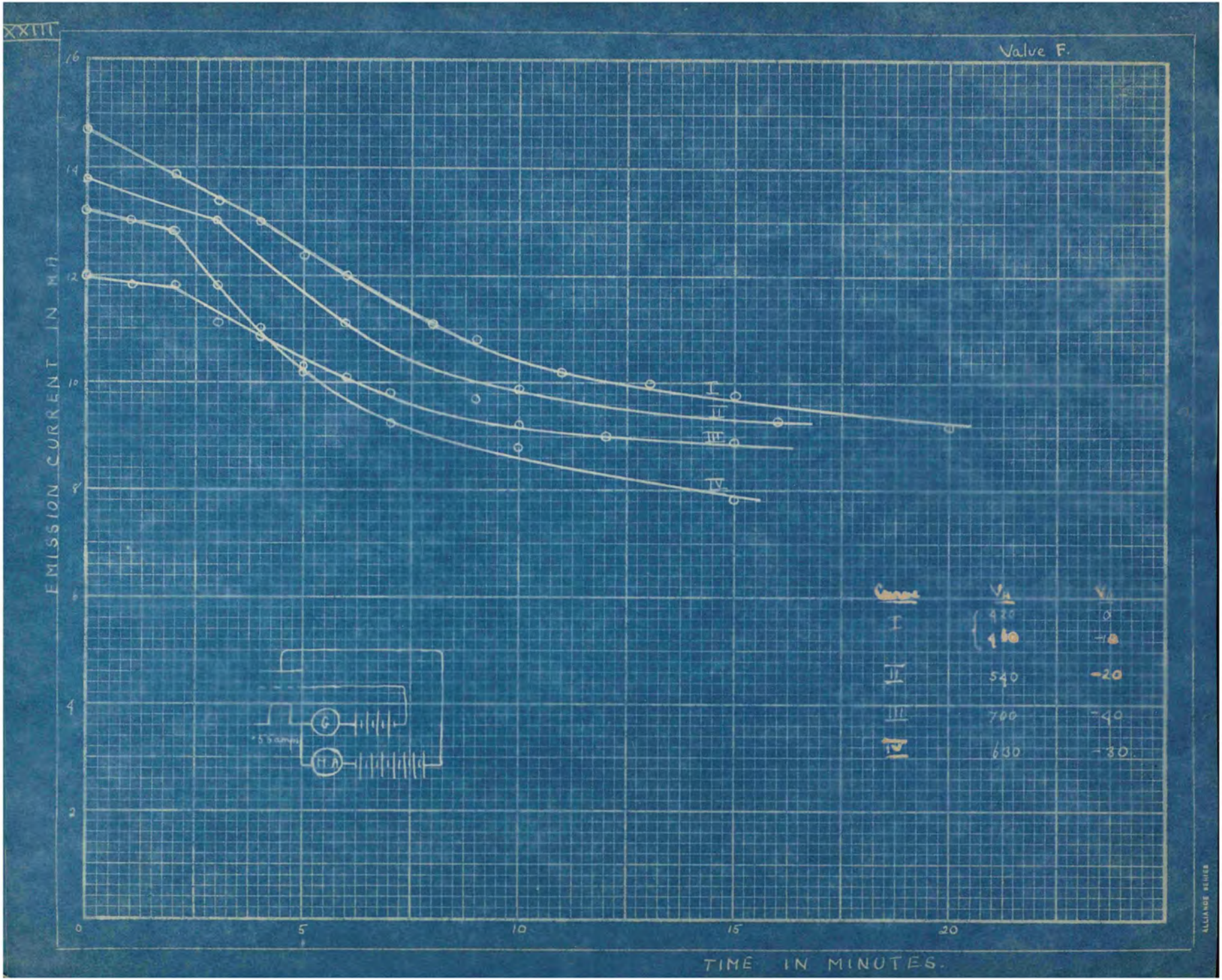
$V_c = -30$

$V_c = -40$


XXII
POSITIVE CURRENT IN AMPS X 10^-8.
0
1
2
3
100
200
300
400
500
600
ANODE VOLTS
Vg = -10
Vg = -20
Vg = -30
Vg = -40
Valve F.
ALLIANCE SERIES

XXIII

Valve F.



XXIV

Valve F

POSITIVE CURRENT IN AMPS X 10⁻⁶

10
8
6
4
2

$V_A = 700$ $V_G = -40$

$V_A = 540$ $V_G = -30$

$V_A = 540$ $V_G = -20$

$V_A = 400$ $V_G = -10$

$V_A = 400$ $V_G = 0$

5
10
15
20

TIME IN MINUTES.