

ON A NEW METHOD OF DETERMINING THE THERMIONIC CONSTANTS OF MOLYBDENUM—I

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1. INTRODUCTION.

Nearly all the investigations on the thermionic constants of metals are based on the pioneer work of O. W. Richardson in which the electronic emission, i , from a filament of the metal under investigation heated in vacuum to a temperature T , is measured and the constants A and ϕ of the equation

$$i = AT^2 \exp(-\phi/kT)$$

are determined. Such determinations suffer from certain defects.

According to the simple thermodynamic theory of thermionic emission by Dushman, A in the above equation is equal to $A_0(1-r_e)$ where $A_0 = 4\pi m\epsilon k^2/h^3$, r_e is the external reflection coefficient for the electrons incident on the metal, and ϕ is the heat of evaporation of the electrons at the absolute zero. According to the Fermi-Dirac statistical derivation of Sommerfeld, which is more rigorous, $A = A_0(1-r_i)$, where r_i is the internal reflection coefficient for electrons, but ϕ now becomes the work function at the temperature T . In the latter case however ϕ depends on the temperature but if this temperature dependence is ignored, the two formulae become identical if $r_i = r_e$, a relation which actually holds. As has been shown by Becker and Brattain (1934), the reflection coefficient is less than 0.1 and the experimental value of A is not usually accurate to within 10%. In principle, however, the filament type of experiments serve to determine $A_0(1-r)$ or $A_0(1-r) \exp[-(e/k)(\partial\phi/\partial T)]$ if ϕ is assumed to be a function of T and r independent of T (Herring and Nichols, 1949). From this the exact value of A_0 cannot be determined as, theoretically, the value of r is almost incapable of being determined under the experimental conditions.

Then, from the experimental point of view also, some difficulties arise. Formerly, as a result of the rigorous process of degassing by heating with d.c., the filament used to get pitted so that the cross-section varied from place to place and the exact area of the emitting surface was difficult of determination. Now heating is done by a.c. and the surface remains smooth. But, nothing can prevent evaporation of the metal from the surface of the filament so that the cross-section may yet vary from place to place. This may result not only in a non-uniform temperature and emission, but also in complications in the corrections for lead losses, Schottky effects, etc.

Further, the measurement of the filament temperature has its own difficulties. Although tables of emissivities are now available, a temperature scale for each metal to be investigated has to be set up under conditions of heat treatment as nearly identical as possible with those used in the measurement of the electron emission. Since conditions of heat treatment vary from sample to sample and, in the event of crystallisation of the metal occurring, from point to point even in the same sample, an element of uncertainty in the value of the temperature creeps in.

In the correction for the Schottky effect we have to use the equation

$$\log_{10} i_1 = \log_{10} i_0 + 1.905(F_c)^{\frac{1}{2}}/T$$

where F_c is the external field at the emitter in volts/cm. F_c can be calculated in terms of the applied voltage provided that the space charge distribution is accurately known. The accurate calculation of the space charge distribution for any given disposition of the electrodes is a difficult problem. For the simple case of a heated straight filament surrounded by a coaxial cylindrical anode the theoretical investigations, as pursued by Langmuir (1913), Langmuir and Blodgett (1923), and others, could give the space charge distribution only for the idealised case of the velocity of the electrons emitted being zero—an assumption unrelated to facts, since the emitted electrons are known to have a Maxwellian velocity distribution.

The present report deals with a new method of determining the thermionic constants of metals, and the constants of molybdenum have been found. A further communication will deal with the results obtained on Iron and Graphite by the same method. It will be seen from the present report that the new method has certain distinct advantages and is free from the defects mentioned above in the usual filament type of experiments.

A. S. Bhatnagar (1944) has determined the thermionic work function of graphite from the rate of effusion of electrons through a tiny hole in an otherwise closed hollow cylinder made from a rod of graphite heated electrically in vacuum to various high temperatures.

It is obvious that if we make a similar cylinder of molybdenum and heat it, then, for any given temperature, the electron gas produced in the metal chamber will attain a saturation pressure in equilibrium with the walls of the chamber and that the rate of effusion of the electron gas through the hole will be proportional to this pressure. If the effused electrons are collected by a Faraday cylinder with a limiting diaphragm of radius, r , placed at a distance, d , from the effusion hole of area, S , the saturation current, i_0 , has been shown by B. N. Srivastava (1938) to be given by

$$p = (2\pi mk)^{\frac{1}{2}} \cdot (r^2 + d^2) \cdot i_0 \cdot T^{\frac{1}{2}} / \epsilon Sr^2 \quad \dots \quad (1)$$

where p is the equilibrium pressure of the electrons within the cavity and ϵ , m , k , and T are the electronic charge and mass, the Boltzmann constant, and the absolute temperature, respectively.

The effect of the space charge on the electronic current under such an arrangement of apparatus has been worked out theoretically by Srivastava and Bhatnagar (*a*, 1944) and the conclusions derived have also been experimentally verified by them (*b*, 1944).

Now, applying the Clausius-Clapeyron equation to the equilibrium vapour pressure of a monatomic gas, the electronic gas pressure within the cavity of the molybdenum cylinder is given by (Jones, 1936).

$$p = e^c T^{\frac{5}{2}} e^{-\phi/kT} \quad \dots \quad (2)$$

where ϕ is the work function, and c the chemical constant of the electron gas.

Hence $\log_{10} p/T^{\frac{5}{2}} = -\epsilon\phi/kT \log_{10} e + c \log_{10} e$.

Substituting the value of p from (1), we get

$$\log_{10} i_0/T^2 = -\epsilon\phi/kT \log_{10} e + c \log_{10} e - \log_{10} (2\pi mk)^{\frac{1}{2}}(r^2 + d^2)/\epsilon Sr^2 \quad \dots \quad (3)$$

so that, as usual, if the experimental values of $\log_{10} i_0/T^2$ are plotted against the corresponding ones of $1/T$, the slope of the curve gives the value ϕ of the work function.

Now, since according to Sackur and Tetrode

$$e^c = 2(2\pi)^{\frac{3}{2}} k^{\frac{3}{2}} m^{\frac{3}{2}} / h^3,$$

$$\therefore c \log_{10} e - \log_{10} (2\pi mk)^{\frac{3}{2}} (r^2 + d^2) / \epsilon Sr^2 = \log_{10} 4\pi mk^2 \cdot \epsilon Sr^2 / h^3 (r^2 + d^2).$$

But $4\pi mk^2 / h^3 = A_0$, the thermionic constant in Richardson's T^2 equation. Hence, the above becomes equal to

$$\log_{10} A_0 Sr^2 / (r^2 + d^2).$$

Whence, (3) becomes

$$\log_{10} i_0 / T^2 = -e\phi / kT \log_{10} e + \log_{10} A_0 Sr^2 / (r^2 + d^2) \quad \dots (4)$$

from which, knowing ϕ , A_0 can be calculated. If, however, ϕ be regarded as dependent on T , the slope will be equal to $\phi - T(\partial\phi/\partial T)$ and the intercept will be

$$\log_{10} A_0 \exp [-(e/k)(\partial\phi/\partial T)] \cdot Sr^2 / (r^2 + d^2).$$

2. EXPERIMENTAL ARRANGEMENT.

The details of the method by which the thermionic constants of molybdenum were determined in accordance with the above ideas are briefly given below.

(a) The Molybdenum Cylinder:

A spectroscopically pure cylindrical rod was bored throughout its length so as to form a tube. This tube was provided with a tightly fitting plug of the same sample of molybdenum at each end. One of the plugs was solid, while the other, facing the Faraday Cylinder, was hollow with a wall at the end lying within the molybdenum tube (Fig. 1). There was a central hole of diameter 0.127 cm. in this

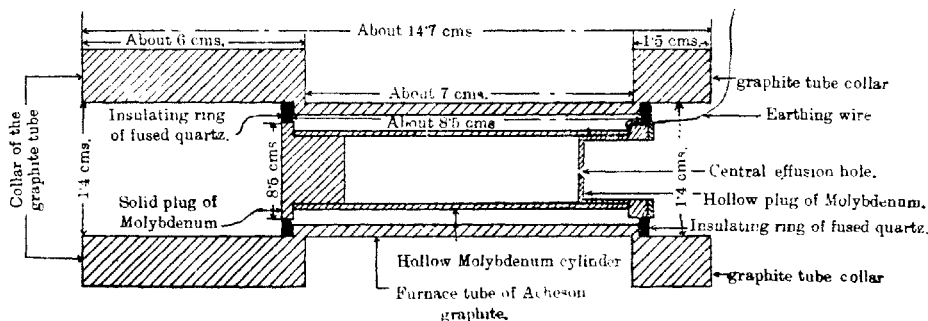


Fig. 1.—Furnace tube of Acheson Graphite with hollow Cylinder of Molybdenum.

wall. It was through this hole that the electrons produced within the cavity of the molybdenum tube, when heated, effused out to be collected by the Faraday cylinder. The overall length of the molybdenum cylinder thus prepared was 8.5 cm., and the outer diameter was 8.5 mms.

The surface of the molybdenum cylinder and plugs was cleansed of all greasy matter with carbon tetrachloride after which a new emitting surface was formed by etching with dil. HCl.

This molybdenum cylinder was heated to various high temperatures in vacuum by being enclosed within, but insulated from, a graphite furnace tube which was heated by a heavy electric current from a low-tension transformer. The insulation

was accomplished by means of loosely fitting fused quartz rings on which the plugs of the molybdenum cylinder rested just outside of the furnace portion of the graphite tube. The hollow plug of molybdenum was earthed by means of a molybdenum wire.

(b) *The graphite furnace tube:*

The graphite furnace tube was made out of a cylindrical rod of Acheson graphite and had the shape shown in Fig. 1, which gives also the disposition of the molybdenum cylinder in the furnace tube. The two collars at the ends of the furnace tube acted as leads to the heating current. The diameter of the fused quartz rings was slightly larger than that of the furnace portion of the graphite tube so that they could not enter the furnace accidentally. As will be seen from the figure, the entire length of the cavity of the molybdenum cylinder as well as most of the lengths of the plugs lay within the furnace portion of the graphite tube.

(c) *The water-cooled vacuum chamber:*

Figures 2 and 3 show the vertical and horizontal sections of the vacuum chamber which housed the furnace tube and its fittings. A double walled water-

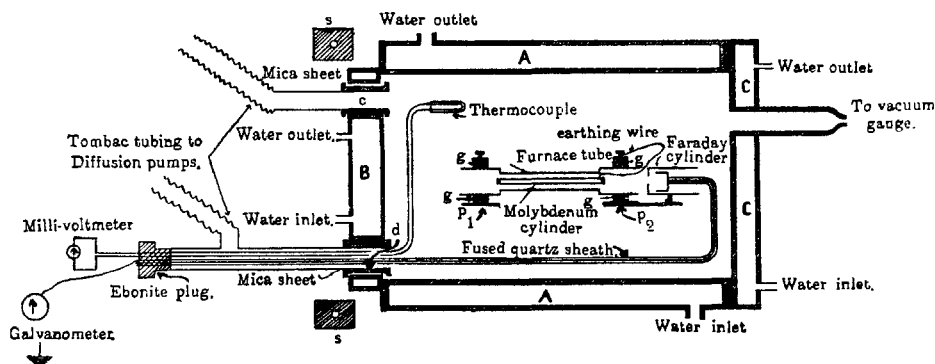


Fig. 2.—Vertical section of the Water-Cooled Vacuum Furnace Chamber.

cooled cast iron drum, *A*, could be made to slide on wheels so as to make an accurate contact with a water-cooled cast iron vertical plate, *B*, fixed to the bed of an old lathe by supports, *SS*, and thus seal the contents against the atmosphere. In the beginning, a gasket of para rubber was kept between the area of contact of the edges of *A* and the plate *B*. But this practice was given up as it was found unnecessary. The vertical plate, *C*, of the drum had a tube leading to a vacuum gauge. The drum was furnished also with a side tube (Fig. 3) on which a glass window was sealed. An optical pyrometer sighted through the window enabled the temperature of the graphite furnace to be measured.

The plate, *B*, had 4 holes, *a*, *b*, *c* and *d*. The holes, *a* and *b*, each carried an annular water-cooled tube, *t*, of copper insulated from *B* by means of mica sheets. On the water-cooled ends within the chamber the tubes had exactly fitting copper collars ending in thick copper plates, *p*₁ and *p*₂, provided with slots. The upper surfaces of *p*₁ and *p*₂ made an intimate electrical contact with rectangular graphite blocks, *g*, between which the collars of the graphite furnace tube were clamped to *p*₁ and *p*₂. The holes, *c* and *d*, carried two brass tubes, each soldered to a length of tombac tubing leading to a 4-stage mercury diffusion pump. The brass piece in *d* had

another hole closed with an ebonite plug which carried through it the wires of a Pt-Pt 10% Rh thermocouple as well as the well-insulated copper lead from the

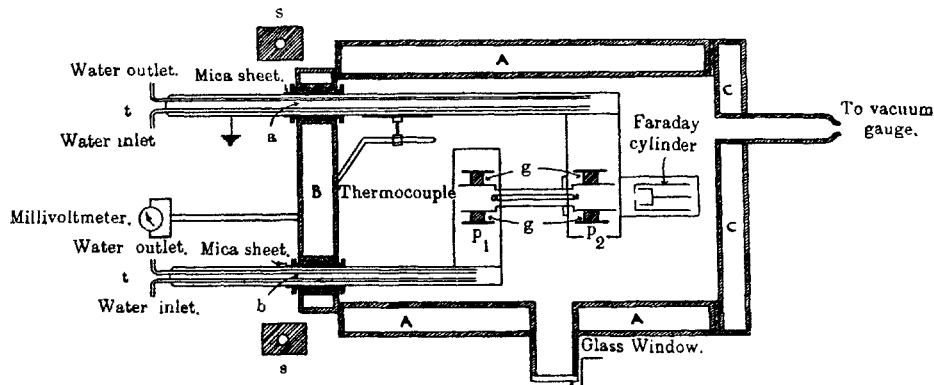


Fig. 3.—Horizontal Section of the Water-Cooled Vacuum Furnace Chamber

central electrode of the Faraday cylinder. The thermocouple wires were connected to a millivoltmeter and the lead from the Faraday cylinder to a sensitive galvanometer kept outside the vacuum chamber. The electrode carrying p_2 was earthed from outside the chamber.

(d) *Temperature measurement:*

Since the molybdenum cylinder was enclosed within the graphite furnace tube, which practically formed a closed cavity, it was reasonable to suppose that the radiations within the cavity were those of a black body so that, after some time, the temperature of the interior of the molybdenum cylinder and, hence, of the emitting surface, would become the same as that of the outer surface of the graphite furnace tube. An optical pyrometer sighted from outside through the glass window in the drum *A* at the graphite furnace tube should, therefore, give the temperature also of the emitting inner surface of the molybdenum cylinder. To see if it was so, the solid plug of the molybdenum cylinder was removed and the thermojunction of the Pt-Pt 10% Rh element was inserted so as to be in contact with the centre of the molybdenum tube. The following data will show that, considering that the smallest graduation in the optical pyrometer was 5°C ., the agreement between the readings of the two instruments was satisfactory.

TABLE 1.

Temp. in $^\circ\text{C}$. of the inner surface of the molybdenum tube as measured from the thermojunction.	Temp. in $^\circ\text{C}$. of the outer surface of the graphite furnace tube as read from the optical pyrometer.
975	980
1260	1260
1440	1435
1500	1500

In the light of the above, it was considered reasonable to assume that the temperature of the inner emitting surface of the molybdenum cylinder was the same as that read on the optical pyrometer sighted at the graphite furnace tube.

It was noticed that, except at its ends where it met its collars the furnace tube glowed uniformly along its length. Readings of the optical pyrometer at different portions of the furnace confirmed the conclusion that the temperature was almost constant along the central portion of the furnace and, hence, must have been still more so within the cavity of the molybdenum cylinder.

(e) *Effect of fused quartz rings:*

Before any measurements on emission from molybdenum could be taken it was necessary to determine whether the two rings of fused quartz themselves emitted any thermionic current.

To test this the molybdenum cylinder was removed and the fused quartz rings were kept in position. No appreciable electron emission could be found even when the temperature of the furnace was kept at 1640°C. for an hour. The temperature of the fused quartz ring near the plate *B*, as measured by the thermoelement in contact with it was only 260°C. The comparatively low temperature of the ring must have been due to the fact that the temperature of the graphite furnace collar was not allowed to rise by the thick graphite blocks, *g*, in intimate contact with the water cooled thick copper plates p_1 and p_2 . To be sure that even under the experimental conditions with the molybdenum cylinder placed in position there was no spurious electronic current reaching the Faraday cylinder, the molybdenum cylinder was reversed and kept in its allotted place on the rings. The solid plug, therefore, now faced the diaphragm of the Faraday cylinder. There was, again, no appreciable electronic current reaching the Faraday cylinder.

This blank experiment established definitely that not only did the fused quartz rings produce no appreciable emission but also that no spurious electronic current was reaching the Faraday cylinder. Hence if, subsequently, on replacing the molybdenum cylinder in its proper position, any current entered the Faraday cylinder it could come only from within the molybdenum cylinder.

(f) *Degassing of the emitter:*

The contents of the vacuum chamber having been sealed by moving the drum against the plate *B*, the backing and then the two diffusion pumps were started. When the maximum possible vacuum had been created the heating current was gradually sent into the previously degassed furnace through the electrodes, *t*, connected to the transformer till the temperature rose to above 1850°C. at which it was kept constant for 2 to 3 hours. Then the temperature was decreased by steps till it fell to about 1650°C. in a few hours. This way the heating was carried on for weeks to condition the emitter and also to degass the contents of the vacuum chamber. When it was considered that the conditioning was probably complete, measurements on the thermionic emission were taken from a temperature of about 1650°C. to the lowest at which the electronic current was measurable by the galvanometer connected to the Faraday cylinder. In order to collect the saturation current positive as well as negative voltages up to 4 volts, in steps of 0.2 volts, were applied to the central electrode of the Faraday cylinder. Usually, about 1 to 2 volts, so applied, sufficed to bring about saturation electronic current. The positive thermionic currents were always negligible and were not considered. It was only when the value of the electronic work function attained a steady value that the final observations were taken.

3. OBSERVATIONS.

A typical day's observation is recorded in the following table. From the readings, taken on different days, of the galvanometer deflections for various temperatures and for different voltages on the Faraday cylinder a graph of

$\log_{10} i_0/T^2$ against $1/T$ for the various days on which the final observations were taken was plotted, i_0 being the saturation current. The numbers 1, 2, 3, 4 and 5, written against the various points on the graph in Fig. 4, indicate the number of the day on which the data were collected.

That the emission was reproducible for a given temperature is shown by the fact that, after the degassing was complete, all the values of $\log_{10} i_0/T^2$ plotted against $1/T$ for different working days fall on the same straight line.

TABLE 2.

Temp. °C.	Galvanometer		Galvanometer deflections in mms. of scale divisions when the voltage applied on the Faraday cylinder was +									Volts.
	Shunt	corresp. sensi- tivity in 10^{-9} amp./mm	0	0.2	0.4	0.6	0.8	1.0	2.0	3.0	4.0	
1610	1/10	17.4	20.0	34.0	49.0	62.0	68.0	72.0	72.5	73.5	74.0	
1575	1/3	5.8	20.0	47.0	82.0	110.0	120.0	127.5	128.5	130.0	131.0	
1540	1/3	5.8	10.0	24.0	42.0	56.0	67.0	73.5	74.5	76.0	77.0	
1515	1/3	5.8	2.0	12.0	25.0	37.0	48.5	49.5	50.5	51.0	52.0	
1495	1/1	1.7	20.0	58.0	95.0	108.0	119.0	119.0	119.5	120.5	121.0	
1450	1/1	1.7	7.0	22.0	37.0	55.5	55.5	56.0	57.0	58.5	60.0	
1350	1/1	1.7	2.0	6.0	8.5	9.0	9.0	9.0	10.5	11.0	12.0	

4. RESULTS.

From the slope of the straight line in Fig. 4, the value of the thermionic work function ϕ of molybdenum comes out to be 4.23 e.v.

Substituting this value of ϕ in (4), the following values of A_0 are found for the various temperatures as given below:

TABLE 3.

Temp. in °C.	1650	1800	1550	1500	1450	1400	1350
Value of A_0 in amp./cm. ² deg. ²	243	244	237	239	240	244	241

Mean 241 amp./cm.² deg.²

5. DISCUSSION.

It will be seen that the method developed in the present investigations has several distinct advantages over the usual filament type of experiments. Being dependent on the rate of effusion, which is a function only of the temperature and the value of the thermionic work function of the metal under investigation, the method is a very direct and straightforward one. There is no complication about the internal or the external reflection coefficient so that, theoretically, the method should give the value of A_0 and not that of $A_0(1-r)$ provided the temperature coefficient of ϕ is assumed to vanish. Then, since the emitting surface is completely enclosed within the graphite furnace tube, it is indirectly heated by the radiations within the graphite enclosure acting almost as a black body. Any effect of the evaporation from the surface of the metal and the consequent variation in

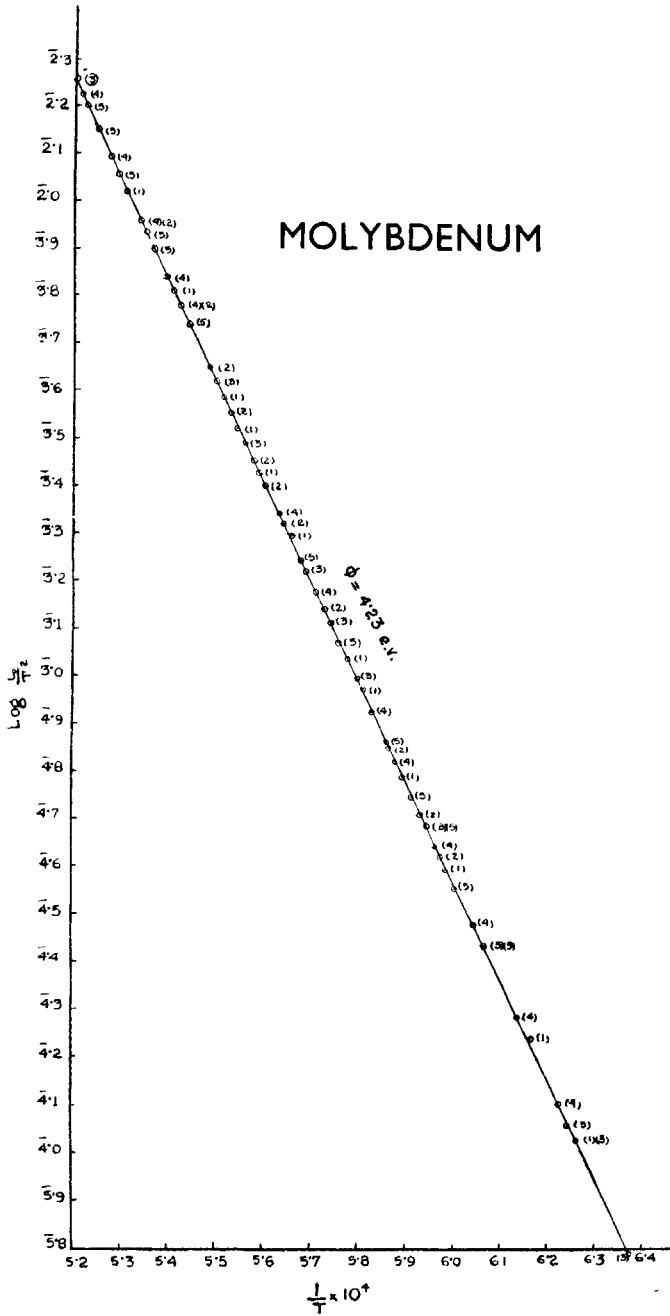


FIG. 4.

the dimensions of the sample does not affect the temperature or the emission. In other methods such a uniformity of temperature is practically impossible to attain. Since no temperature gradients are set up in the specimen no complex corrections are needed. The potential difference required to obtain saturation electronic currents from the effusion hole to the Faraday cylinder is only about 1 to 2 volts so that the difficulties about corrections for the Schottky effect do not occur here. Similarly, corrections for the lead losses, for the rise in temperature of the filament on account of the radiation received from the heated anode, and for the variation in the heating current of the filament due to the return of the space charge do not enter here.

As against the above advantages, it might be argued that the new method is liable to the following drawbacks:

Although before the introduction of the molybdenum cylinder within the graphite furnace tube the latter had been degassed, it might be said that the molecules of carbon and of its oxides, emitted during the heating of the furnace, would contaminate the emitting surface. Secondly some heat would be conducted through the molybdenum plugs and the fused quartz rings thus setting up a temperature gradient in the emitter with the consequent complications of contact potentials, etc. Lastly, although the defect is not due to any inherent fault in the method developed, but extraneous to it, the comparatively low vacuum in the present investigations may not have brought about a thorough conditioning of the emitter and may, therefore, have vitiated the results.

As regards contamination by the molecules of carbon or of its oxides, it must be remembered that the furnace was heated only after the pumps had produced the best vacuum they could. From the geometry of the apparatus it will be seen that the chance of any foreign molecule acquiring a velocity in a direction enabling it to enter the effusion hole—the only avenue possible for it to enter the molybdenum cylinder and contaminate the inner emitting surface—was very small indeed.

Similarly, the chance of any temperature gradient along the surface of the emitter being set up was small. The diameter of the fused quartz rings being larger than that of the molybdenum plugs, the area of contact between the two was small. Hence not much heat could have been lost via the non-conducting rings.

It has already been mentioned that the whole of the graphite furnace tube glowed uniformly except where it met the collars of the tube. Since the entire length of the cavity of the molybdenum cylinder as well as most of the lengths of the plugs lay within the uniformly glowing furnace tube, whatever temperature gradients occurred must have been produced at the ends of the plugs and not within the cavity of the molybdenum cylinder which formed the emitting surface.

It is difficult to estimate as to how far the results of the present investigations have suffered for a lack of the highest vacuum attainable these days. Table 4 shows the values of the thermionic constants of molybdenum obtained by different workers by other methods.

Although the data obtained by the author need not be accepted as final, it will be seen that the value of the work function obtained is in fair agreement with that obtained by other workers. The value of A_0 is not in equally good agreement. Theoretically, the new method should give the value of $A/(1-\tau) \approx A_0$ if the temperature coefficient of ϕ is negligible. But whether the difference is due to an insufficient degassing at high vacuum or to any natural variation in different samples of the same metal caused by the orientation of the axes of the crystals forming the emitter surface is rather difficult to say. Nichols (1940) found a difference of as much as 0.3 e.v. in the value of the work function between crystal directions (110) and (111) in a single crystal prepared of tungsten, while the value of A varied from 125 for the (112) and (001) directions to 15 for the (110) direction. Then, again, besides the above, there is what has been called a 'volume effect' caused, according to E. W. Müller, and Benjamin and Jenkins (1940), by

TABLE 4.

Thermionic Constants of Molybdenum.

Investigator	Year	Method	Value of	
			ϕ in e.v.	A in amp./cm. ² deg. ²
1. DuBridge and Roehr ..	1932	Photoelectric ..	4.15	..
		Thermionic ..	4.15	60
2. Ahearn ..	1933	Thermionic ..	4.32	..
3. Freitag and Krüger ..	1935	Thermionic ..	4.33	..
4. Krüger and Stabenow ..	1935	Cooling effect ..	4.40	..
5. Wahlin and Reynolds ..	1935	Thermionic ..	4.17	55
6. Rentschler and Henry ..	1936	Photoelectric ..	4.35	..
7. Grover ..	1937	Thermionic ..	4.2	..
8. Wright ..	1941	Thermionic ..	4.2	55 for strip and 115 for filament.
9. Mathur ..	1946	Effusion ..	4.23	241 [$A/(1-r)$].

the periodicity of the crystal lattice which results in forbidden energy regions occurring for those electron momenta which satisfy the conditions for Bragg reflection along the various crystal directions. If such a region exists at or just above the top of the surface barrier in the case of thermionic emission, or at the top of the Fermi level in the case of field emission, some of the electrons will be absent since the energy in question will lie in the forbidden region for the corresponding directions of motion inside the crystal. We should, therefore, in the light of the above, expect to obtain a sort of an average value for the thermionic constants which may vary from sample to sample depending on the disposition of the axes of the crystals forming the emitting surface. It will be seen that the values of ϕ obtained by different workers using the most up-to-date vacuum technique vary by as much as 0.25 volt for molybdenum, while the value of A varies between 55 and 115. It is possible that in the case of the present investigations a long process of high temperature treatment at moderately high vacuum may have sufficed to force the foreign molecules to quit the surface and to condition it so that the investigations of the author may not, after all, have suffered materially for lack of a very high vacuum. As Wohlfarth (1948) says, until the various disturbing effects are more fully understood, no really satisfactory comparison of theoretical and experimental results can be made.

The author wishes to thank Sir K. S. Krishnan, F.R.S., for kindly suggesting the problem and for his guidance, and Dr. B. N. Srivastava for much help. The investigation was carried out during 1945-46 in the Physics Department of the Allahabad University where the author had gone on study leave from the Lucknow University. For part of the time, the author was appointed as a research assistant to Professor Krishnan by the Council of Scientific and Industrial Research, New Delhi.

SUMMARY.

By measuring the electronic current effusing out of a small hole in an otherwise closed cylinder of molybdenum, heated to various high temperatures in a graphite furnace tube worked in vacuum, the electronic work function of molybdenum was found to be 4.23 e.v. in good agreement with some of the recent determinations by other methods. The value of the constant A , which was not in similar good agreement with the values found by other investigators, was found to be 241 amp./cm.² deg.²

The merits of this newly developed method are discussed.

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