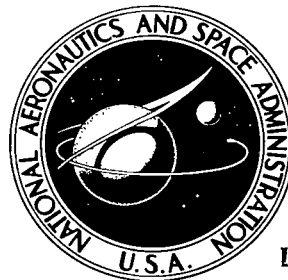


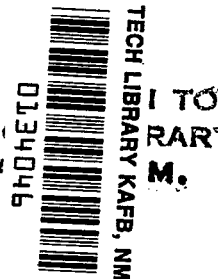
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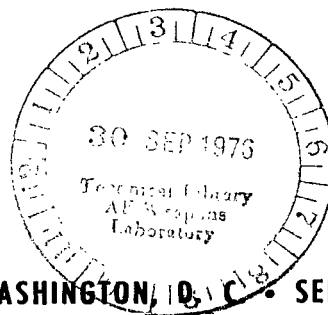


SURFACE STUDIES OF THERMIONIC CATHODES AND THE MECHANISM OF OPERATION OF AN IMPREGNATED TUNGSTEN CATHODE

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SUMMARY

Auger spectroscopy, work function measurements, and residual gas analysis are the techniques used in this study of active cathodes operated at nominal values of emission and temperature. The surface properties of conventional impregnated cathodes are investigated by the use of Auger spectroscopy and work function measurements, and these are compared with a synthesized barium or barium oxide coated tungsten surface. The barium and barium oxide coated tungsten surfaces are prepared by evaporating barium onto a tungsten surface which can be heated to elevated temperatures. Multilayer or monolayer coverages can be investigated using this technique. The results of this study show that the surface of an impregnated tungsten cathode is identical to that observed for a synthesized monolayer or partial monolayer of barium on partially oxidized tungsten, using the criteria of identical Auger patterns and work functions.

Desorption measurements of barium from a tungsten surface were also made. These results in conjunction with Auger and work function data have been interpreted to show that throughout most of its life an impregnated cathode operating in the range of 1100° C has a partial monolayer rather than a monolayer of barium on its surface.

INTRODUCTION

The space program is responsible for many unique accomplishments in modern technology, and one of the most interesting is that associated with space communications. In recent years one space communications technology program has been directed toward obtaining high-powered (100 to 1000 W), high-frequency microwave tube amplifiers. A basic requirement for such an amplifier is that it be long-lived since replacement in most cases is very costly and difficult and in others impossible. Since the thermionic cathode would be responsible for the ultimate failure of the tube and other space commun-

ication equipment, the real need is to develop long-lived, reliable thermionic cathodes for space-power amplifier tubes. The first phase of such a program is to obtain a basic understanding of the mechanism of operation of conventional cathodes used in high-power microwave amplifiers. The second phase is to develop improved long-lived cathodes based on the information obtained from the first part of the program. This report presents work accomplished in the first phase of the program.

Conventional cathodes, which are used in modern microwave power amplifiers, can be divided into two categories: oxide cathodes and impregnated tungsten cathodes. Oxide cathodes are generally limited to microwave devices operated at relatively low current densities and low power and are not currently being considered for high-powered, space applications. Impregnated tungsten cathodes can be operated at current densities in the range of amperes per square centimeters, are therefore of current interest in NASA space communication devices, and are the major subject of this report.

The impregnated tungsten cathode is an outgrowth of the L-cathode (ref. 1) developed in the early 1950's. These thermionic cathodes are mechanically rugged, have good thermal properties, and are capable of delivering high electron current densities. The cathode, invented by Levi (refs. 2 to 4), is made by impregnating a porous tungsten body with a suitable molten barium compound, this body having one or more surfaces that emit electrons at elevated temperatures. Levi (ref. 4) preferred a barium compound having the composition $5\text{BaO} \cdot 3\text{CaO} \cdot 2\text{Al}_2\text{O}_3$. This mixture in an impregnated tungsten matrix is known as the type B cathode. Variations of the mixture (e.g., $4\text{BaO} \cdot \text{CaO} \cdot \text{Al}_2\text{O}_3$, the S-cathode developed by Semicon Associates, Inc.) have been proposed to improve the device, but the basic principle of impregnating a porous tungsten body with an appropriate barium compound mixture is common to all conventional commercial impregnated tungsten cathodes. The most recent innovation in the art is the M-cathode developed by Philips (ref. 5). This consists of a conventional B-cathode whose surface is covered with a thin layer of osmium. Philips claims that osmium lowers the work function of the device and enables the user to operate it at a lower temperature than conventional impregnated cathodes for the same emission current, thereby leading to longer life.

Impregnated tungsten cathodes have been finding increasing use in the microwave power tube industry because of their ruggedness, good thermal properties, and especially high emission capabilities. The interest in obtaining more reliable and durable cathodes of this type has been responsible for generating many research studies in this area in the last 20 years. The objective of most of these studies was to obtain a better understanding of the basic mechanism of thermionic emission from the devices. It was generally agreed, in the early work, that free barium is generated in the interstices of tungsten, by a reaction between tungsten and the barium compound mix and that the barium diffuses through the pores to the tungsten surface (refs. 6 to 8). However, differing mechanisms (refs. 9 and 10) were offered for the surface reaction responsible for lowering the work

function of the tungsten surface of the cathode.

In recent years, with the development of Auger electron spectroscopy (AES), a major breakthrough has occurred in techniques for studying surface reactions. Auger electron spectroscopy permits one to study and identify chemical elements existing on a solid surface where the coverage can vary from a fraction of a monolayer to many monolayers. It is a valuable technique for studying the surface phenomena responsible for the copious electron emission from thermionic cathodes. Two studies (refs. 11 and 12) of tungsten impregnated cathodes using AES have recently appeared. Both reach conclusions that differ from those obtained in the present investigation; the reasons for these differences are discussed later in this report.

Auger electron spectroscopy work function measurements, and residual gas analysis are the techniques used in this study of an active cathode operated at nominal values of emission and temperature. The experimental program was divided into two parts: The first was an investigation of the surface properties of conventional impregnated cathodes using Auger and work function measurements. The second was a basic study of the surface properties of barium or oxidized barium on a clean tungsten surface. A thorough investigation of the second part enables one to interpret the AES and work function data of the first.

EXPERIMENTAL TECHNIQUES

Figure 1 illustrates the experimental apparatus used for these measurements. The equipment was all contained in a bakeable ultrahigh vacuum system, using Vac-ion pumping and capable of attaining base pressures in the range of 1.33×10^{-9} newton per square meter (10^{-11} torr). The sample is either a thermionic cathode or a pure tungsten pellet in these experiments. It is connected to the micromanipulator, which enables the rotation or three-dimensional placement of the sample so that it faces either the anode, for electron emission measurements, the Auger spectrometer, or the barium dispensing getter. The temperature of the sample is controlled by passing current through the heater, and its temperature is measured with an optical pyrometer through a sighting hole. The residual gas analyser is used to monitor the gases evolved during the experiment and the purity of any gas introduced into the system. High purity gas (e. g., oxygen) is introduced into the vacuum system by means of a variable leak valve and can be controlled down to the 1.33×10^{-7} N/m² (10^{-9} torr) level. Figure 2 is a photograph of a cathode and the getter mounted on the micromanipulator flange.

Temperature measurements on commercial conventional thermionic cathodes were made by optical pyrometry using the sighting hole (fig. 1). To obtain the true temperature, the optical readings were corrected for the emissivity of the cathode sleeve. However, in the barium and barium oxide on tungsten measurements, the pure tungsten pellet

temperature was measured by a tungsten-rhenium thermocouple in addition to pyrometric comparative measurements.

The emission capability of cathodes at normal operating temperatures was obtained by placing the cathode approximately 1.5 millimeters away from the anode and obtaining current-voltage characteristics. Very high current density measurements, up to 5 to 6 amperes per square centimeter, could be made CW. (continuous duty) because the oxygen-free, high-conductivity (OFHC) copper anode surface was cooled with water flowing at the rate of 76 liters per minute (20 gal/min). At low temperatures, in the range of 1000 K, emission measurements were made by measuring the current obtained when 200 volts was applied between the cathode and anode. This current was always less than 10 percent of the space-charge-limited current at that voltage and was assumed to be the zero-field, temperature-limited current value normally used in the Dushman-Richardson emission equation. This procedure was originally adopted to make work function measurements of barium films on tungsten. Because the film coverage changed with time even at temperatures in the range of 1000 K or lower, we arbitrarily chose to measure the emission current at 200 volts and use the electron-current value obtained to calculate the effective work function. This parameter ϕ was calculated from the Dushman-Richardson relation

$$\phi = -kT \ln(J/120 T^2). \quad (1)$$

where k is Boltzmann's constant in electron volt per kelvin, T is the temperature in kelvins, and J is the current density in ampere square centimeter.

The Auger spectrometer (fig. 1) is essentially a surface probe that identifies the chemical constituents from the top atomic layers of a surface (ref. 13). It operates on the principle that the energy of the Auger secondary electrons, which are emitted from a surface bombarded with a high-energy electron beam, are related to the atomic levels that are present in the chemical species existing on the surface. An analysis of this spectrum of secondary energies permits one to determine the atomic species on the surface. This technique became practical when Harris (ref. 14) using a method of electronic differentiation was able to demonstrate the high sensitivity attainable and the practicality of this procedure for surface analysis. The coaxial cylindrical Auger analyzer used in this program is commercially available and is one of the latest developments in Auger equipment. The output one obtains from the spectrometer is a curve of dN/dE versus E , where N is the number of secondary electrons emitted at energy E . It has been shown (refs. 13 and 14) that dN/dE is a more sensitive parameter than N for detecting the Auger peaks and that the position of the sharp minimums in the dN/dE versus E curves is the best technique for determining the Auger transition energies of atomic species on the surface. The Auger spectral curves that appear in this report have the ordinate labeled dN/dE .

The barium dispenser (fig. 1) is a KIC barium getter that consists of barium enclosed in a cylindrical iron container. A small hole or opening is filed into the getter tube, as shown in figure 1, before the getter is mounted in the vacuum station. A drop of kerosene is then placed over the hole to protect the barium inside the hole from the outside atmosphere during the time it takes to get the equipment (fig. 2), into the vacuum station and pumped down. After the vacuum system is baked out and pumped down, the barium is deposited on the sample by rotating the sample to the getter and heating the barium getter to 400° to 500° C by passing current through it.

EXPERIMENTS ON IMPREGNATED TUNGSTEN CATHODES

The two types of conventional impregnated cathodes tested were the Philips B and the Semicon S. They correspond to a $\text{BaO} \cdot \text{CaO} \cdot \text{Al}_2\text{O}_3$ ratio in the impregnant of 5:3:2 and 4:1:1, respectively. Figures 3 and 4 illustrate the Auger spectra obtained from a B-cathode before, during, and after activation. The data of figure 3 cover the high-energy range of 150 to 650 electron volts and those of figure 4, the low-energy spectra from 40 to 140 electron volts.

In figure 3(a), taken before the cathode was activated, one sees the presence of barium, calcium, and oxygen in the high-energy spectrum and no tungsten lines. This indicates that the impregnant essentially covers the surface. The low-energy spectrum (fig. 4(a)) has several energy minimums that correspond to barium and, possibly, aluminum lines.

Parts (b) of figures 3 and 4 illustrate the data obtained during activation with the cathode at 1175° C (1448 K). It should be noted in figure 3(b) that tungsten now appears prominently and the calcium line has disappeared. The low-energy spectrum (fig. 4(b)) shows more detailed spectral lines than figure 4(a).

After final activation the Auger spectrum given by figures 3(c) and 4(c) result. These curves represent the fully activated, impregnated tungsten cathodes operating at 1100° C (1373 K) and occur on both S- and B-cathodes. Figure 3(c), the high-energy data, is similar to 3(b) except for minor variation in intensity for both tungsten and oxygen lines and the absence of carbon in 3(c). After activation, the carbon spectral line never appears in the spectra during high-temperature operation, as has been previously reported (refs. 11 and 12). The low-energy spectrum (fig. 4(c)) for the activated cathode, however, now shows very detailed spectral lines and a distinctive shape, which will later be shown to be characteristic of monolayer or partial monolayer coverage of barium on tungsten as distinct from barium oxide on tungsten. Springer and Haas (ref. 11) have reported that sulphur was always present in their experiments on activated B cathode. It was not, however, present in our tests on either the B- or S-cathodes.

The space charge and temperature limited characteristics of the B-cathode at 940° C (1213 K) are illustrated in figure 5. Its effective work function was measured as 2.0 electron volts at 1030 K by the technique described in EXPERIMENTAL TECHNIQUES using equation (1).

The activated cathode Auger data of figure 3 indicate that the cathode has a surface layer containing both oxygen and barium and that this surface layer can only be monolayers thick because the underlying tungsten comes through strongly on the Auger pattern. The spectrum, which has been discussed previously (refs. 11 and 12), indicates that barium, oxygen, and tungsten are present on the surface. The fact that oxygen is always present with barium in this Auger spectrum is one of the major reasons that led researchers to conclude that a BaO surface exists on the active impregnated cathode (refs. 11 and 12). However, the low-energy spectrum (fig. 4(c)) is even more revealing. It contains most, but not all, of the published spectral lines for barium (ref. 15). If the surface is BaO as reported, should one expect this spectrum to be different? This is a reasonable question because the Auger spectra of some metal oxides are different from the unoxidized metal (refs. 16 to 18). This is particularly true for the low-energy Auger electrons because these involve transitions between upper lying energy states of the chemical species. If the oxide formation changes the metallic model of the metal to one containing a valence band, one could expect this change to be observed in the low-energy Auger transitions. A study of the surface properties of deposited barium and oxidized barium films was conducted to resolve this problem.

EXPERIMENTS WITH BARIUM AND BARIUM OXIDE

ON TUNGSTEN AND DISCUSSION

This part of the program consists of studies of the surface properties of absorbed barium and barium oxide films on a pure tungsten surface. An Auger spectrum of the bare tungsten surface (the reference surface) is shown in figure 6. This spectrum is identical to that reference 15. If barium is deposited on the surface to a sufficient depth, the tungsten Auger pattern disappears, and the bulk barium lines are obtained (fig. 7). The accuracy of the electronic energy transitions is ± 1 electron volt. The spectra in figure 7 have slightly more detail at low energies than those published by Palmberg, et al. (ref. 15), recently, but they are consistent with their data. To obtain BaO data, oxygen is introduced into the vacuum system in the pressure range of 1.33×10^{-6} newton per square meter (10^{-8} torr). The pressure was measured with a nude Bayard-Alpert gage, and no attempt was made to correct the readings for oxygen ion formation (ref. 19). The introduced oxygen gas was analyzed by the residual gas analyzer and showed very minor impurities of atomic masses 40, 20, 28, and 2 listed in their order of magnitude. The

curves shown in figure 8 illustrate what happens to the low-energy barium spectrum when oxygen is introduced at a pressure of 6.65×10^{-6} newton per square meter (5×10^{-8} torr). The identifying parameter on each curve is the degree of exposure to oxygen given in Langmuirs (L) where $L = 1.33 \times 10^{-4}$ newton second per square meter (10^{-6} torr sec). The pattern displayed by the curve for 366 langmuirs remained stable with any additional oxygen exposure and is assumed to be the pattern for BaO. The final pattern for oxidized barium (or BaO) is shown in figure 9. The high-energy spectrum differs only from the pure metal by the prominent oxygen peak, but the low-energy spectrum is quite different. There are eight energy level transitions in the barium spectrum and three in the BaO spectrum. This experimental evidence proves that one can differentiate between barium and BaO films by the use of low-energy Auger spectroscopy.

Although one can differentiate for heavy deposits of barium and BaO on tungsten, can one differentiate for monolayer film coverage? This was verified experimentally, and the data are illustrated in figure 10. The lower curves of figures 10(a) and (b) illustrate the Auger spectrum that results from a monolayer of barium on tungsten scan. The monolayer was obtained by using a procedure described by Moore and Allison (ref. 20) for their experiments with barium on tungsten filaments. They showed that bulk barium films, deposited on tungsten, can be converted to a monolayer by heating the tungsten substrate to 1000 K for 1 to 3 minutes. Such a procedure was followed in this investigation. First, a bulk barium film was deposited on a cleaned tungsten disk containing a small, partial coverage of oxygen. Because the maximum operating temperature was limited to about 1500 K, oxygen could never be completely removed from the tungsten surface. The bulk deposition of barium resulted in the Auger spectrum shown in figure 7. The tungsten surface was then heated to 1000 K for 1 minute; the patterns shown on the bottom curves of figures 10(a) and (b) were the result. This pattern is characteristic of a barium monolayer on tungsten. It should be noted, however, that there is a measurable oxygen peak in the spectrum, which means that the surface is predominantly barium on tungsten but that there are some surface areas which probably contain barium on oxygen on tungsten. When this monolayer is exposed to oxygen at 6.65×10^{-6} newton per square meter (5×10^{-8} torr) for 11 minutes (33 L), the upper spectra of figure 10 were obtained from the oxidized sample. The major differences observed occur in figure 10(a). The detailed spectrum of six energy peaks for the barium monolayer becomes three broad energy peaks in the oxidized barium layer, a similar phenomenon to that observed in the bulk barium experiments (fig. 7 to 9). The most interesting point about the monolayer barium data of figure 10 is that they are almost identical to those obtained for the activated impregnated cathode in figures 3 and 4. In addition, work function measurements on the barium monolayer surface, made at 1000 K, show it to be 2.0 ± 0.5 electron volts, which is also the same as that measured for the impregnated tungsten cathode. The 0.05 electron volt is a maximum deviation in the measurement. The similar Auger spectra and effective work functions show that the surface of an activated impregnated cathode is

very similar to the surface of a synthesized monolayer of barium on tungsten.

The effective work function of the oxidized monolayer surface of figure 10 was 2.3 ± 0.05 electron volt, much higher than that for the barium monolayer. This is the obvious poisoning mechanism, that is, the deleterious effect of oxygen on an impregnated cathode. However, bulk BaO films such as those shown in figure 9, have very different electrical properties. Their effective work functions, at 900 K, were 1.75 ± 0.05 electron volts, much lower than even the monolayer barium films. This is consistent with the model normally suggested for an oxide cathode such as a BaO matrix cathode (ref. 21). Auger spectroscopy on an oxide cathode, discussed later in this report, confirms this conclusion.

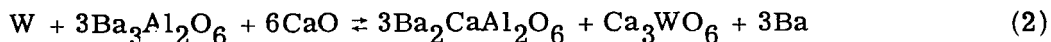
As the one monolayer of barium data of figure 10(b) show, residual oxygen is always present when barium is deposited. We were not able to produce barium films on tungsten without some residual oxygen. Since we were limited to heating the samples to temperatures of about 1500 K because of possible heater problems, we were not able to eliminate oxygen completely from even a "clean" tungsten surface. This is illustrated in figure 6 where one can see some residual oxygen in the Auger spectrum. Other possible sources of oxygen are (1) the getter wire, which may give off oxygen when it is heated, and (2) the tungsten sample from which oxygen may diffuse when the sample is heated to produce a monolayer. We suspect the tungsten because no oxygen peaks were observed by the residual gas analyser (hydrogen was mainly observed) when the getter wire was heated to evaporate barium.

Ritner, Ahlert, and Rutledge (ref. 10) have found, with hot tungsten wire techniques, that barium on clean tungsten has a higher work function and lower sticking time than barium on oxidized tungsten. We have not been able to duplicate their results because of our inability to prepare a barium on tungsten surface without some residual oxygen. However, we were able to measure the effect of increased oxygen coverage on the surface reaction of barium on oxygen on tungsten. This was done in the following experiment. A clean tungsten surface, similar to that shown in figure 6, was exposed to oxygen (120 langmuirs) until the oxygen peak in the Auger pattern was saturated. Barium was then deposited on the surface, and the surface was heated to 1000 K for 1 minute. The Auger pattern that resulted is shown in figure 11. It is very similar to the monolayer of barium pattern in figure 10, except for the larger oxygen peak. This larger oxygen peak probably corresponds to monolayer coverage. The resulting Auger spectrum is a monolayer of barium on oxygen on tungsten where each barium atom overlies an oxygen atom. The effective work function for this surface agreed with that for the surface monolayer of barium within experimental error. These measurements indicate that the work function of a monolayer of barium on oxygen on tungsten is relatively independent of oxygen coverage for a relatively wide range of oxygen coverage.

The Auger spectrum and work function measurements on an activated impregnated cathode, early in life, have shown that the surface responsible for copious electron emission is a monolayer of barium on "slightly oxidized" tungsten. This assertion is mainly based on the proposition that the two different surfaces, which have almost identical Auger spectra (e.g., activated data of fig. 3 and the one-monolayer-of-barium curves of fig. 10), are equivalent. The equality of work functions confirms this conclusion. Since the tungsten matrix of the cathode is also never heated in vacuum above 1500 K, it is not surprising that it also shows a residual oxygen peak. Monolayer coverage is the preferred surface condition because it leads to the lowest possible work function (refs. 20 and 22). But does it exist on the surface of an impregnated cathode throughout life? To answer this question one has to consider the mechanism of operation of an impregnated cathode and design an experiment to either prove monolayer or partial coverage during life.

DISCUSSION OF THE MECHANISM OF OPERATION FOR AN IMPREGNATED TUNGSTEN CATHODE AND DESORPTION EXPERIMENTS

The Auger pattern for an activated cathode shows that its surface, at normal operating temperatures, is initially a monolayer of barium on oxidized tungsten. This is an equilibrium condition where the rate of barium evaporated from the surface is equal to the rate of barium arriving at the surface from internal barium diffusing to the surface. The internal barium is being generated by a chemical reaction, which is thought to be (ref. 5)



The barium generated in the cathode pores has to flow to the surface at a high enough rate to assure an adequate supply of barium at the surface. If the arrival rate is greater than that needed to maintain a monolayer, the excess barium evaporates. Multilayer barium on tungsten is unstable at normal operating temperatures, and the excess barium would probably evaporate, leaving the more stable barium monolayer. If the arrival rate is less than that needed to maintain a monolayer, partial coverage will result and, as will be shown later, lead to a higher work function.

Assuming that this discussion explains the mechanism for cathode operation, one can then set up an experiment to determine whether monolayer coverage exists during life. If a synthesized barium monolayer on tungsten surface, having an Auger spectrum identical to that of the activated cathode of figure 3, is heated to some operating temperature and the change in barium coverage measured as a function of time, one can determine

the desorption rate of barium as a function of coverage. By multiplying the value of the desorption rate at monolayer coverage with the lifetime of an impregnated cathode at this temperature, one can obtain the total barium evaporated from the surface of such a cathode during its lifetime, if it maintained monolayer coverage throughout its life. Showing that this figure for total barium evaporation is either equal to, less than, or greater than the total available barium in the cathode impregnant will allow us to evaluate whether monolayer coverage persists during the life of a cathode. Lifetime data on commercial B- and S-cathodes, operating at approximately 1100⁰ C (1373 K) and at 2 amperes per square centimeter, have recently been obtained (ref. 23). The latest results show that these cathodes can operate for more than 2 years at these operating conditions. Therefore, it would be desirable to design an experiment to measure the rate of evaporation as a monolayer of barium from partially oxidized tungsten at 1100⁰ C (1373 K) and to determine whether this rate of evaporation is compatible with the life test data.

When desorption experiments were attempted at 1100⁰ C (1373 K), the initial desorption rate was so high that major changes in coverage developed before the operating temperature could be reached and reliable quantitative data could not be obtained. Therefore, we decided to determine desorption at lower temperatures, where desorption rates were more stable, and to extrapolate these values to the higher temperature. Measurements were made at 1000 and 1100 K. (The data are shown in fig. 12.) Auger peak to peak height as a function of time at a given temperature was the technique used in these measurements (ref. 24). The experimental data of figure 12 could best be fitted by a curve of the form suggested by Langmuir (refs. 20 and 25)

$$\theta^4 = \frac{1}{1 - 4qt} \quad (3)$$

where θ is the coverage that varies from 1 to 0, t is the time, and q the curve fitting parameter at a given temperature. To extrapolate the coverage function to higher temperatures, one has to have a relation for θ as a function of temperature T . This is given by this equation from reference 26,

$$\frac{d\theta}{dt} = Kf(\theta)\exp \frac{-E}{kT} \quad (4)$$

where t is time, K is a velocity constant, $f(\theta)$ is the number of sites for which desorption is possible, k is Boltzmann's constant in electron volts per kelvin, and E is the activation energy for desorption. The value of E can be determined by taking the ratio of $d\theta/dt$ for the same coverage at temperatures T_1 and T_2 , for example,

$$\frac{d\theta(T_1)/dt}{d\theta(T_2)/dt} = \exp \left[-\frac{E}{k} \left(\frac{1}{T_1} - \frac{1}{T_2} \right) \right] \quad (5)$$

From the result

$$\frac{d\theta}{dt} = q\theta^5 \quad (6)$$

and from the values of $d\theta/dt$ at 1000 and 1100 K in figure 12, one obtains from equation (5)

$$E = 2.06 \text{ eV} \quad (7)$$

A coverage function θ_3 for the desorption of barium from an oxidized tungsten surface at 1100° C (1373 K) can then be extrapolated from equations (3), (5), (6), and (7) as

$$\theta_3^4 = \frac{1}{1 + 13.8t} \quad (8)$$

and

$$\frac{d\theta_3}{dt} = -3.45 \theta_3^5 \quad (9)$$

where t is expressed in minutes. As discussed earlier, no reliable quantitative experimental verification of equation (8) was obtained. However, order of magnitude agreement could be obtained. When a monolayer of barium was heated to 1100° C (1373 K) for about 5 to 10 minutes, the barium coverage dropped to about 0.3, which is in semiquantitative agreement with equation (8). This validates the method of using equation (8) to calculate rates of evaporation with the understood limitation that the answers are only reliable for order of magnitude agreement.

If one calculates the rate of evaporation of barium on tungsten from equation (9) as a function of coverage by multiplying the value of $d\theta_3/dt$ by the number of barium atoms per square centimeter in a monolayer (5×10^{14} , ref. 20) and the mass of a barium atom, one obtains the 1100° C curve in figure 13. The figure is a plot of barium evaporation rates as a function of coverage with temperature as the variable parameter. The curves for 1000° and 1200° C (1273 and 1473 K) were obtained by using a similar procedure to that used for obtaining equation (9). The rate of evaporation for a monolayer is shown to be 2.4×10^{-5} grams per square centimeter per hour at 1100° C (1373 K) in figure 13. If this rate were to persist for 2 years, the total barium evaporated would be 0.42 gram per square centimeter. This is about an order of magnitude higher than the amount of barium available for activation in the B- and S-cathode used in the life tests of reference 23 (see

table I) and indicates that the coverage has to be less than a monolayer on an impregnated cathode for most of its life. If we look at the 1100°C (1373 K) curve in figure 13, we see that the evaporation rate drops very rapidly with coverage. In fact, at a coverage of 0.6, the evaporation rate is down by an order of magnitude, in the range of 10^{-6} gram per square centimeter per hour. At this evaporation rate the impregnated cathode could operate satisfactorily for 2 years at 1100°C (1373 K) if the work function of the cathode at $\theta = 0.6$ is low enough to sustain a current density of 2 amperes per square centimeter, which is the operating current under discussion. To determine the work function of the barium on oxygen on tungsten surface as a function of coverage, the emission from the surface (initially a monolayer coverage) at a given temperature as a function of time was measured. Typical results of such measurements are illustrated in figure 14. If the data of figure 14 are compared with the 1100°K curve of figure 12, one obtains the curve of work function against coverage (fig. 15). At a coverage of 0.6, the work function is 2.18 electron volts. Assuming that this value can be extrapolated to the higher temperature (1100°C or 1373 K), its insertion into equation (1) predicts a current density of 2.3 amperes per square centimeter, which is in the right order of magnitude.

As pointed out earlier, the purpose of this discussion is not to quantitatively predict life test experimental data with the proposed theoretical model. The experimental data obtained in these measurements of barium on tungsten are not accurate enough to predict that the cathode at 1100°C (1373 K), runs at a coverage of 0.5, 0.6, 0.7, etc., for most of its life. The main goal of this discussion is to show that, because of the high rate of evaporation of a monolayer of barium on oxidized tungsten, it is much more likely that, during most of its life, the impregnated cathode has a partial monolayer rather than a monolayer of barium on its surface. Assuming that this model is valid, one can propose a mechanism to explain the life characteristics of impregnated cathodes. Early in life when the barium arrival rate at the surface is high, because barium is being generated in pores close to the surface, one would expect the evaporation rate to either be larger or equal to that needed to maintain monolayer coverage. As the pores near the surface are depleted of barium, both the arrival rate and the evaporation rate will decrease. If the rate of arrival of barium at the surface is less than that needed to maintain a monolayer, a partial monolayer will result such that the evaporation from the partial monolayer equals the rate of arrival of barium at the surface. If the work function of the partial monolayer is lower than that needed to sustain emission, the cathode will still be satisfactory. Eventually, a condition will be reached where the rate of arrival of barium at the surface gives a partial monolayer whose work function is too high to sustain the electron emission, and the emission will drop resulting in the end of useful life.

Published results on barium evaporation during life from an impregnated cathode are consistent with the predicted evaporation rates from the proposed model. Brodie, Jenkins, and Trodden (ref. 27) measured the variation of evaporation rate during life from

an impregnated cathode having an impregnant composition $3\text{BaO} \cdot \text{Al}_2\text{O}_3 \cdot \frac{1}{2}\text{CaO}$. This was an accelerated life test where the cathode was run at 1500 K. Periodic observations were made of the evaporation rate and cathode temperature. These investigations found that the evaporation rate decayed considerably during life. Initially, the evaporation rate at 1100°C (1373 K) was about 1.2×10^{-5} gram per square centimeter per hour 60 hours later it was 2.3×10^{-6} , and 2200 hours later it was 4×10^{-7} gram per square centimeter per hour. Comparing these values with the 1100°C (1373 K) curve of figure 13 shows that they correspond to about a monolayer down to approximately 0.4 of a monolayer near the end of life. Rittner, Tutledge, and Ahlert (ref. 8) measured the variation of evaporation rate during the life of an impregnated cathode (impregnant composition, $5\text{BaO} \cdot 2\text{Al}_2\text{O}_3$) operating at 1190°C . The initial evaporation rate was 6×10^{-5} gram per hour per square centimeter, and the rate near the end of life (about 4000 hr) was 6×10^{-7} gram per hour per square centimeter. Once again, the values correspond to a monolayer coverage at the start of life and about 0.4 of a monolayer at the end. (See the 1200°C curve of fig. 13.)

Rittner, Rutledge, and Ahlert also included the electron emission data of their cathode throughout its life. Their findings do not agree with the model proposed herein. Although the drop-off in current at the end of life is consistent with the model, in that a calculated coverage of 0.4 of a monolayer predicts an effective work function of 2.3 electron volts (extrapolated from fig. 15), the fact that their temperature-limited emission data were constant during life is not. According to the model proposed herein, temperature limited emission should have decayed with life. However, their measurements were made on very early impregnated cathodes which are not at all representative of the commercial cathodes measured in this study. The cathode was difficult to activate and had very poor electron emission characteristics compared with modern conventional cathodes. The authors attributed these problems to either contamination or some poisoning agent on the surface. Their results on emission with life were probably determined by this effect, rather than any effects due to barium surface coverage.

In concluding this section a comment should be made on the desorption of oxidized barium, or BaO, from the tungsten surface. Although no quantitative measurements were made on the desorption of oxidized barium, as distinct from those reported here for barium on oxygen on tungsten, the qualitative measurements showed that desorption from oxidized films (e.g., figs. 9 and 10) was much more difficult than desorption for unoxidized films. Similar results have been reported by other investigators (ref. 10).

EXPERIMENTS ON THE M-TYPE PHILIPS CATHODE

In recent years there has been considerable interest in a new type of impregnated cathode developed by Philips called the M-cathode (refs. 5 and 28). This cathode is sim-

ilar to the $5\text{BaO} \cdot 3\text{CaO} \cdot 2\text{Al}_2\text{O}_3$ impregnated tungsten cathode discussed previously except that the tungsten surface is coated with osmium and ruthenium. Philips claims this treatment lowers the work function so that it can be operated 100° cooler than the normal B-cathode and still deliver the same temperature limited current density. This obviously would lead to cathodes with much longer life and have important implications for applications to high-powered microwave space tubes. Since life extension is a major objective of this program, the study was extended to this cathode.

The M-cathode was activated by (a) heating it to 1200°C (1473K ; 1100°C brightness on the molybdenum sleeve) for 10 minutes, (b) then heating it at 1150°C (1423K) for about 3 hours, and (c) aging it at 1 ampere per square centimeter for 2 hours at 1150°C (1423K). The Auger spectra taken after activation are shown in figure 16. The high-energy spectrum shows the identifiable osmium and ruthenium transition levels in the 150 to 300-electron-volt range as well as the oxygen and barium lines. The other transition levels between 200 and 300 electron volts are combinations of osmium and ruthenium transitions. The low-energy spectrum is particularly interesting in that it has the same transition energies and almost the same shape as that shown in figure 4(c) for an activated conventional cathode, early in life. This indicates that the active surface is probably either a barium monolayer or partial barium monolayer on the oxygenated osmium-ruthenium surface, a situation similar to that of the conventional cathode. However, the barium coverage on the M-cathode leads to a much lower work function value than that on conventional cathodes (ref. 5). This result can be obtained from the Schottky plot (fig. 17) on the initial diode data from the M-cathode. The data taken at 1010K show the zero field emission current density to be 0.126 ampere per square centimeter, and the experimental Schottky slope exceeds the theoretical value by a factor of 2.1. From the zero field emission value one calculates an effective work function of 1.8 electron volts, a significant improvement of 0.2 electron volt from the normal impregnated cathode. To illustrate the advantages of this lower work function, the simple calculations shown in table II were made. The results are derived by assuming that the work function is constant over the range of temperature considered, being 1.8 for the M-cathode and 2.0 for the conventional cathode, and by inserting these values into equation (1).

Lewis has been interested in the life potential of conventional cathodes operating at space-charge current densities of 2 amperes per square centimeter. Reference 23 shows that the conventional impregnated tungsten cathode operating at 1100°C (1373K) and delivering space-charge-limited currents of 2 amperes per square centimeter has a useful life of 2 years or more. An examination of the values in table II indicates that an M-cathode would only have to operate at 975°C (1248K) for the same results. If the desorption curve for barium on osmium is similar to that for barium on tungsten, operation at the lower than normal temperature should reduce the evaporation of barium by a factor of 5.8 (from eq. (5)). This seems to indicate that an M-cathode operation at

975^o C (1248 K) and delivering 2 amperes per square centimeter of electron current could have a useful life of 10 years or more. This conclusion has to be tempered by the fact that the useful life of the M-cathode may not be determined by barium depletion but rather by the alloying action of osmium and tungsten, which would result in a much higher work function cathode surface (ref. 28). If this mechanism is dominant for even a short time, the useful life of the M-cathode could be considerably less than this optimistic value. Finally, it should be noted that (1) Van Stratum and Kuin (ref. 28) found no appreciable alloying action after 27 000 hours at 950^o C and (2) even though an M-cathode may degenerate, it eventually ends up identical to a conventional impregnated tungsten device at the end of its useful life. As a consequence, it can then still be used as a conventional device at a higher operating temperature until all the barium is depleted.

EXPERIMENTS ON COATED POWDER CATHODES

The coated powder cathode (CPC) is a modified oxide cathode originally developed by Maurer and Pleass (ref. 29) at Bell Laboratories. Tests were run on this cathode to verify a conclusion stated in this report that the electron emission characteristics of a multilayered oxidized barium (BaO) surface resemble those of an oxide cathode. After activation, Auger data were obtained on the CPC's (fig. 18). The high-energy spectrum (150 to 600 eV) shows the characteristic calcium, oxygen, and barium lines. The cathode studied was a triple carbonate, and strontium was observed in the detailed spectrum, not shown here, in the range of 1300 to 1750 electron volts. The low-energy spectrum (fig. 18 (a)) is particularly interesting. It shows that the barium lines between 50 and 75 electron volts resemble the oxidized barium lines observed in figure 9(a). This resemblance substantiates earlier statements about the oxidized multilayer barium surfaces.

CONCLUSIONS

The copious electron emission characteristics of a conventional impregnated tungsten cathode arise from the existence of a monolayer or partial monolayer of barium on a partially oxygenated tungsten surface. Barium coverage at any temperature is determined by an equilibrium condition such that the rate of evaporation of barium from the surface is equal to the rate of arrival of barium at the surface from the barium generated in the pores of the cathode by the impregnant reacting with the tungsten matrix. Early in the life of an activated impregnant tungsten cathode, when the barium arrival rate to the surface is high because barium is being generated in pores close to the surface, the arrival rate is high enough to sustain a barium monolayer coverage. As the pores near the

surface are depleted of barium, the arrival rate of barium decreases and only a partial monolayer of barium can be sustained on the surface. The cathode will continue to function satisfactorily as long as the partial monolayer has a low enough work function to maintain the required current density. The end of life will occur when the arrival rate of barium is too low and the work function is too high to support the required electron emission.

The active chemical constituent on the surface of an impregnated tungsten cathode is barium on oxygen on tungsten and not barium oxide on tungsten. Oxygen is always present in an activated cathode, but the results obtained in this report indicate that it is not chemically combined with the barium.

Auger studies on the M-cathode show that barium on oxygen on the osmium-ruthenium surface is responsible for its low-temperature operation as a copious electron emitter.

A final conclusion of this program is that an oxidized monolayer of barium on oxygen on tungsten has a much higher work function than its unoxidized counterpart (Ba on O on W) and that a multilayered barium oxide surface resembles an oxide cathode surface and has a much lower work function than a monolayer of barium on oxygen on tungsten.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, April 29, 1976,
506-20.

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TABLE I. - CHARACTERISTICS OF B- AND S-CATHODES^a

Type of cathode	Emitting area, cm ²	Thickness, cm	Porosity of tungsten pellet, percent	Impregnant	Impregnant weight, percent of total	Barium available, mg/cm ²
B	0.32	0.2	20	5BaO · 3CaO · 2Al ₂ O ₃	4.5	27
S	.32	.2	18	4BaO · CaO · Al ₂ O ₃	5	36

^aCathodes used in life test studies at the Watkins-Johnson Co.

TABLE II. - COMPARISON OF EMISSIONS OBTAINED FROM B- AND M-CATHODES

Type of cathode	Cathode operating temperature, °C					
	850	900	950	1000	1050	1100
M-cathode	1.3	3.1	6.9	15	29	56
Tungsten impregnated cathode	.16	.42	1.0	2.4	5.1	10

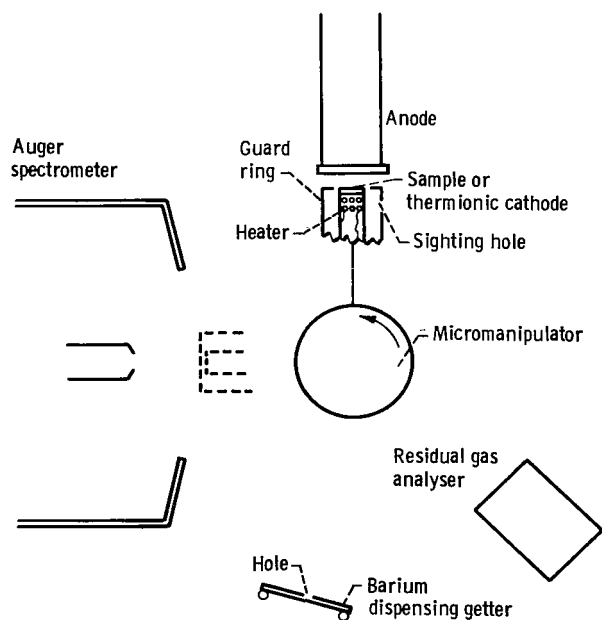
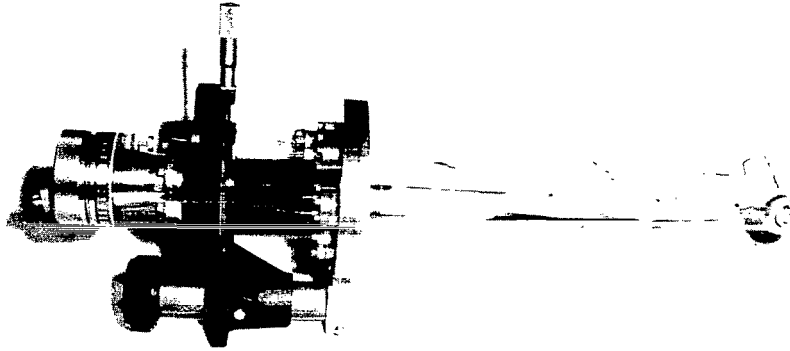


Figure 1. - Schematic view of experimental apparatus.



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Figure 2 - Impregnated cathode and getter mounted on micromanipulator.

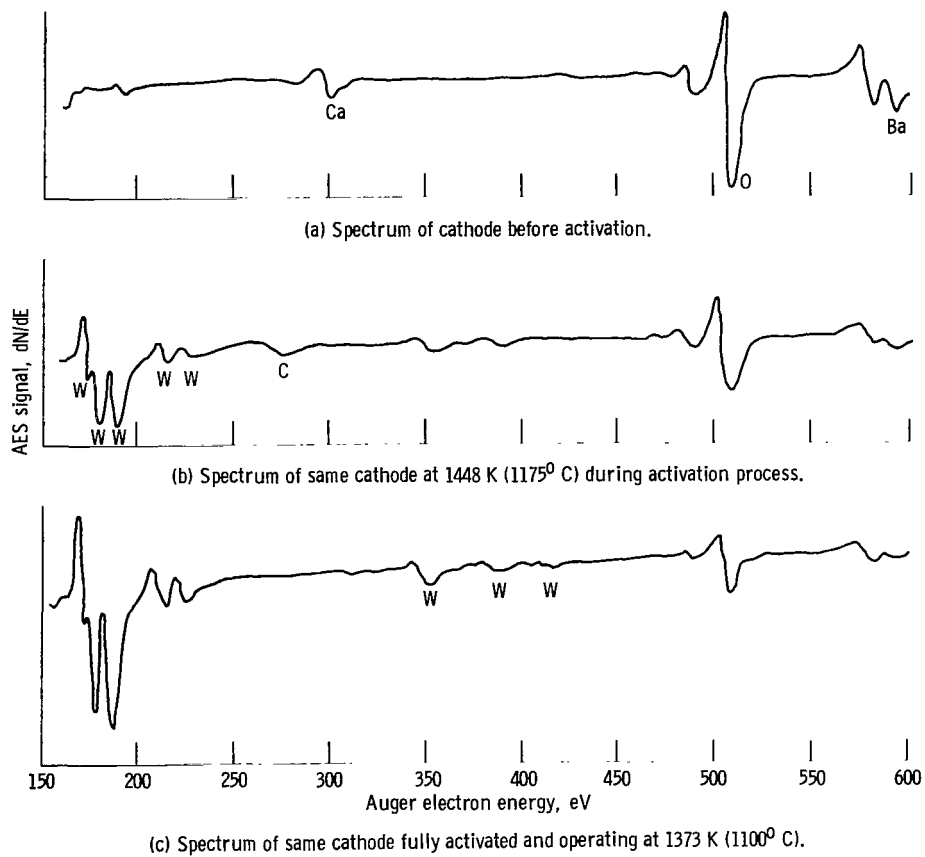


Figure 3. - Auger spectrum of type B impregnated cathode in energy range 150 to 600 electron volts.

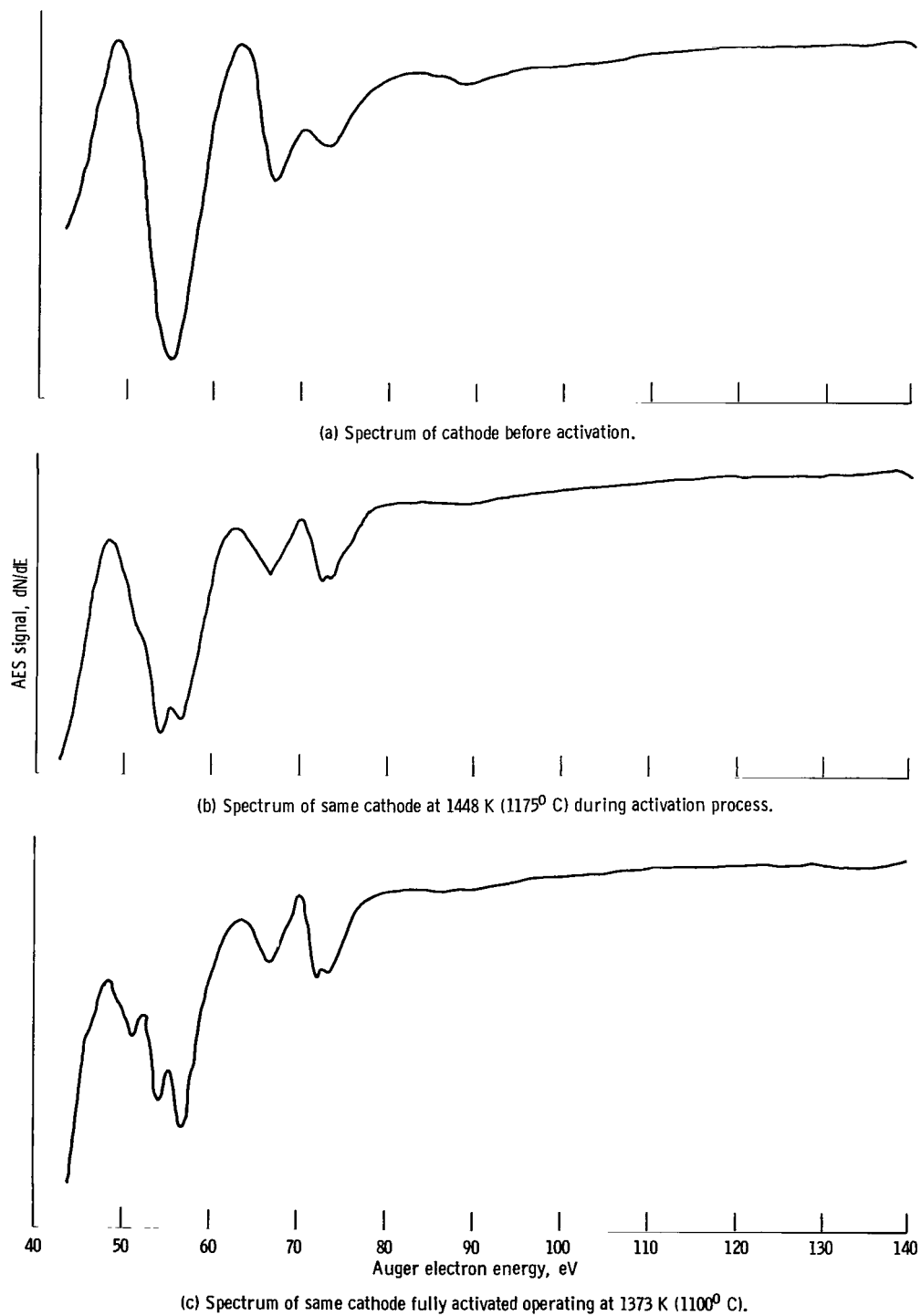


Figure 4. - Auger spectrum of type B impregnated cathode in energy range of 40 to 140 electron volts.

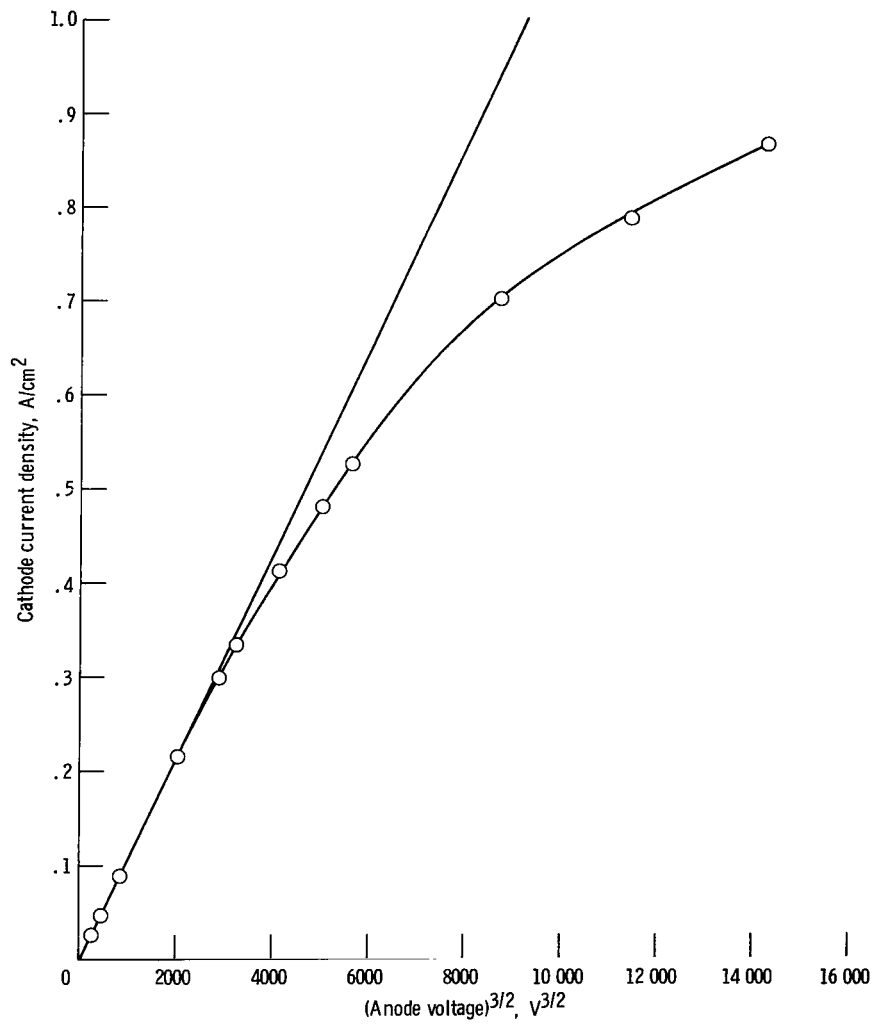


Figure 5. - Cathode current as function of (anode voltage)^{3/2} for B-cathode operating at 1273 K (940° C).

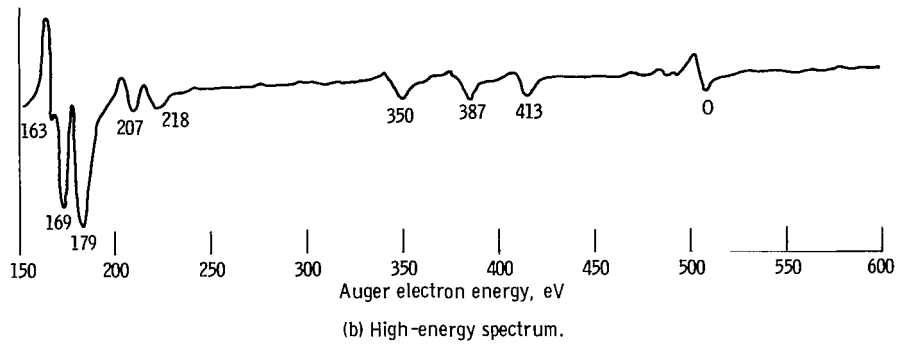
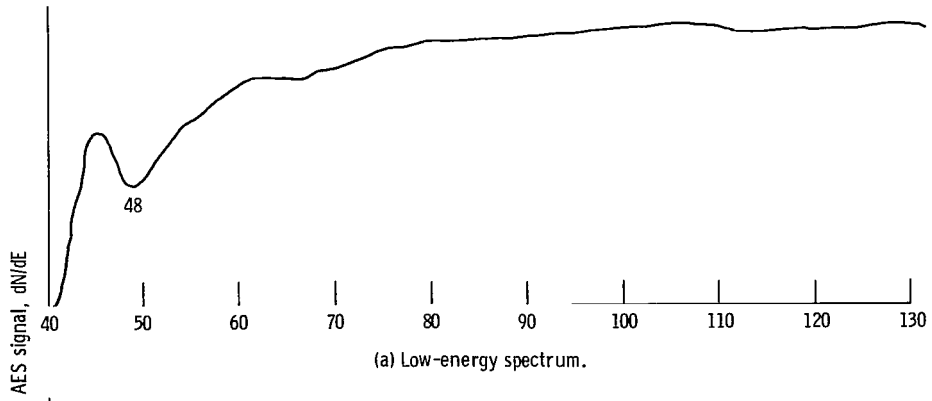


Figure 6. - Auger spectrum of tungsten.

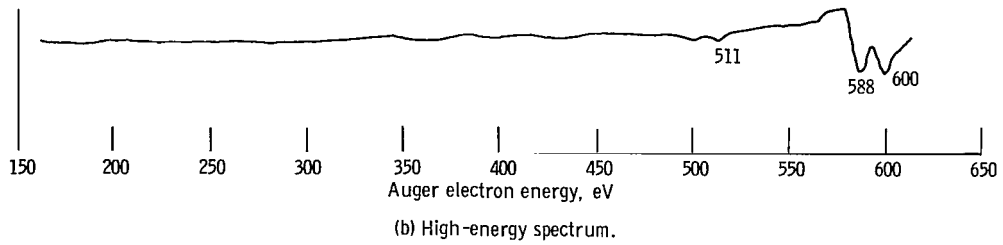
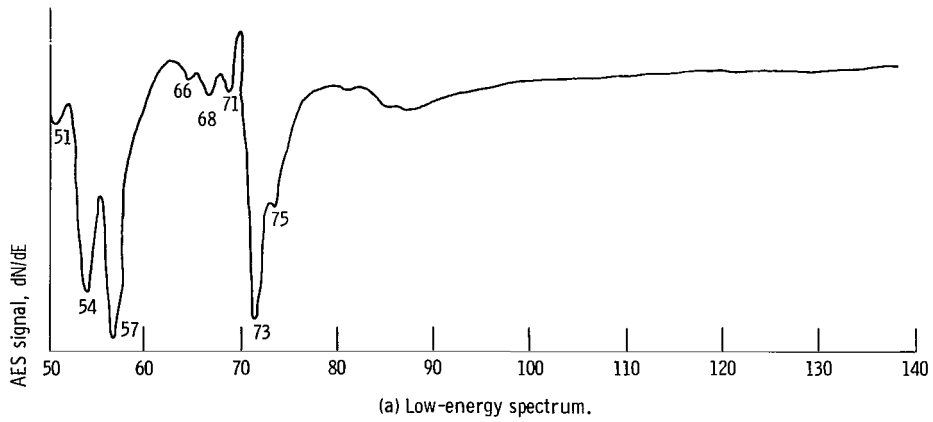


Figure 7. - Auger spectra of barium.

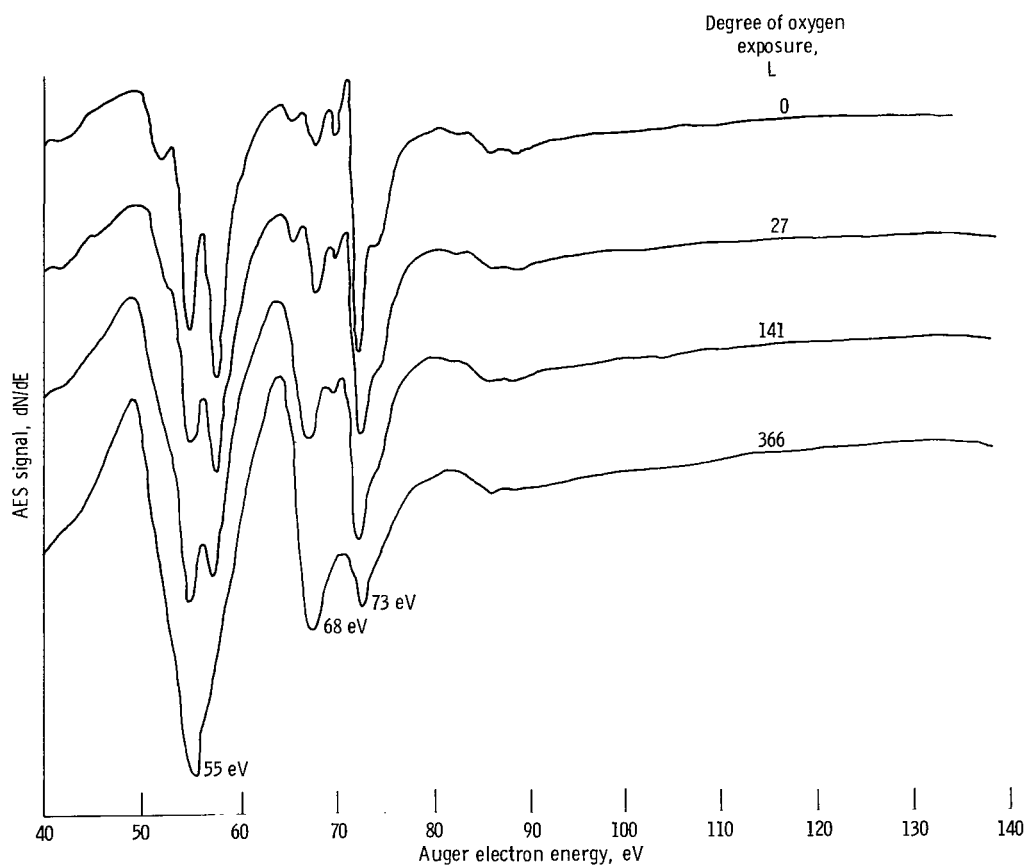
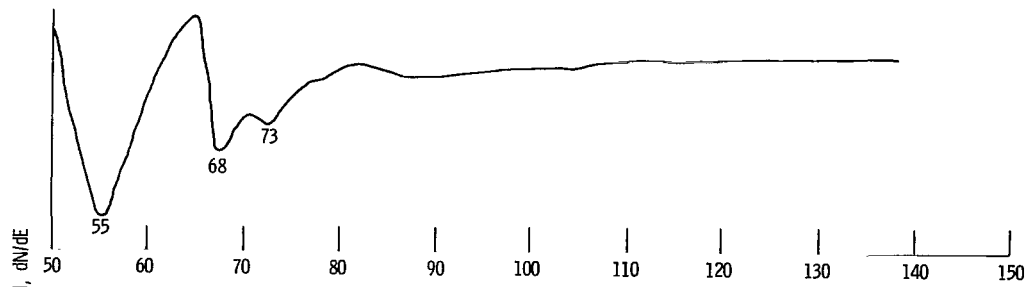
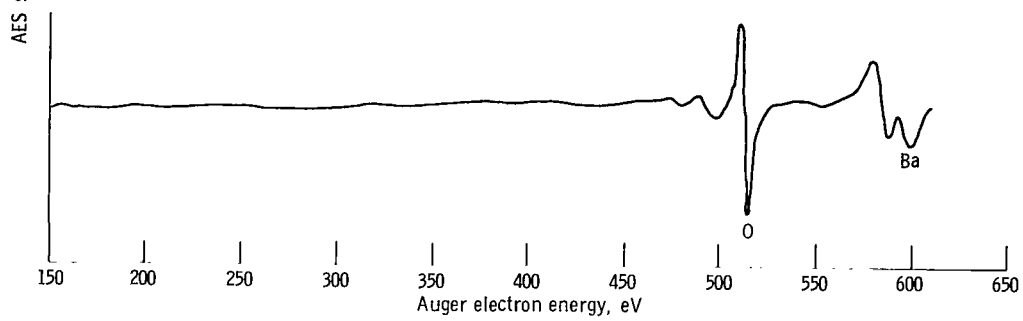


Figure 8. - Low-energy Auger spectrum of barium exposed to oxygen at (5×10^{-8} torr).

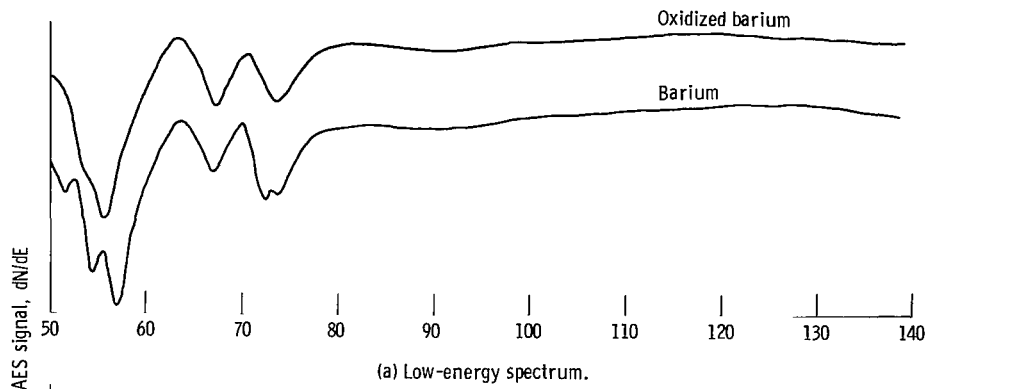


(a) Low-energy spectrum.

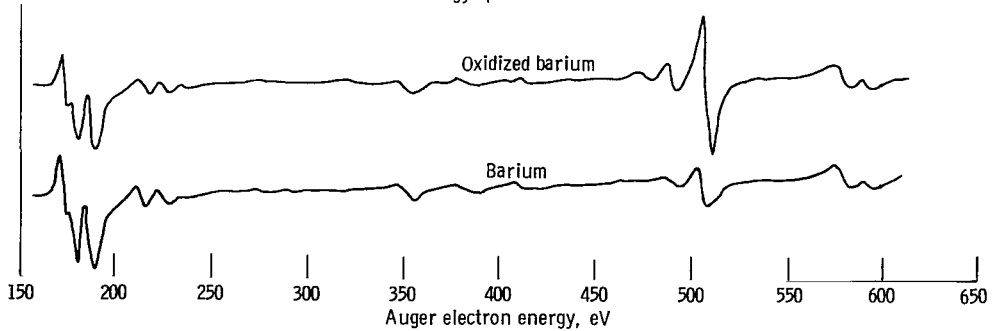


(b) High-energy spectrum.

Figure 9. - Auger spectra for oxidized barium.



(a) Low-energy spectrum.



(b) High-energy spectrum.

Figure 10. - Auger spectra for barium and oxidized barium monolayers on tungsten surface.

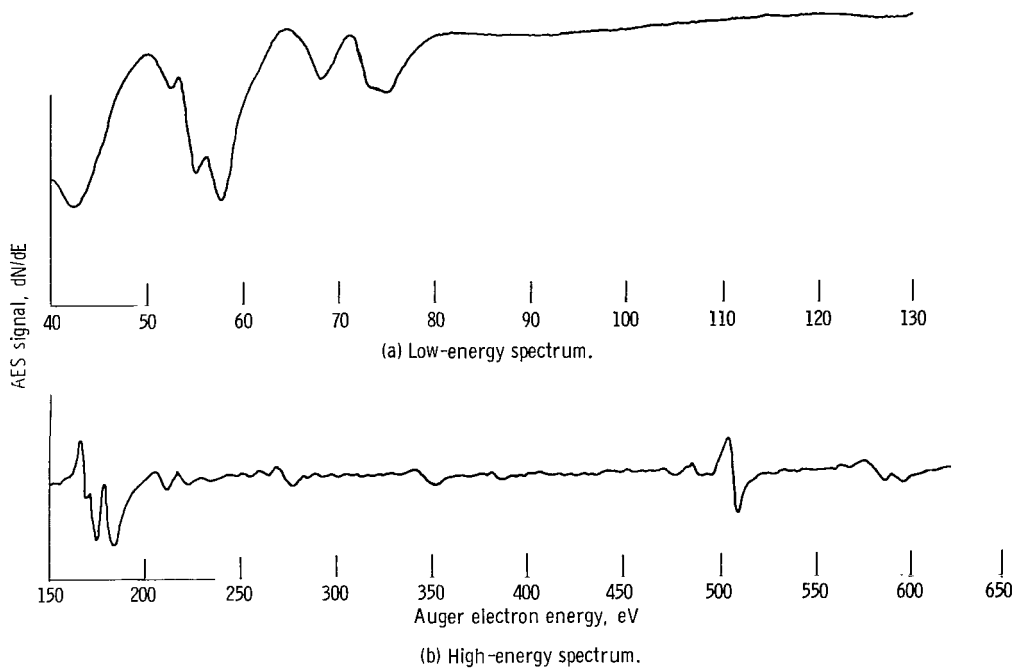


Figure 11. - Auger surface for barium on saturated oxygen on tungsten surface.

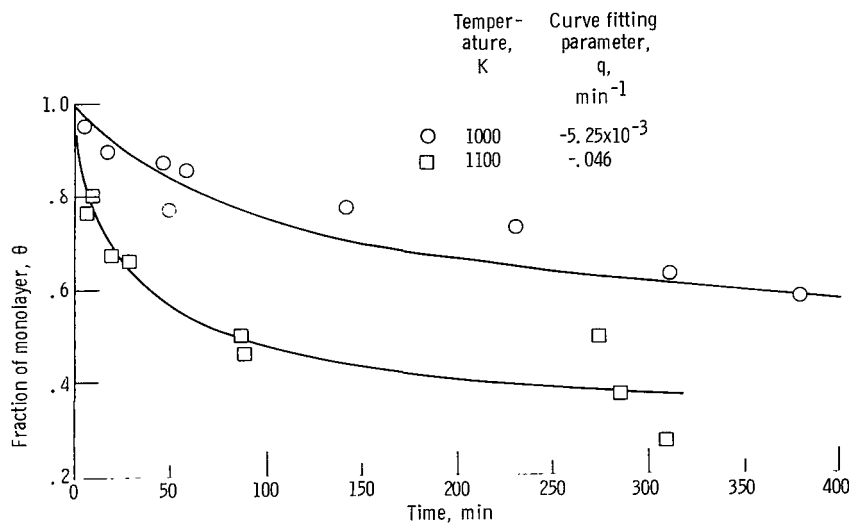


Figure 12. - Desorption curves for monolayer of barium on tungsten at 1000 and 1100 K as function of time. (See eq. (3).)

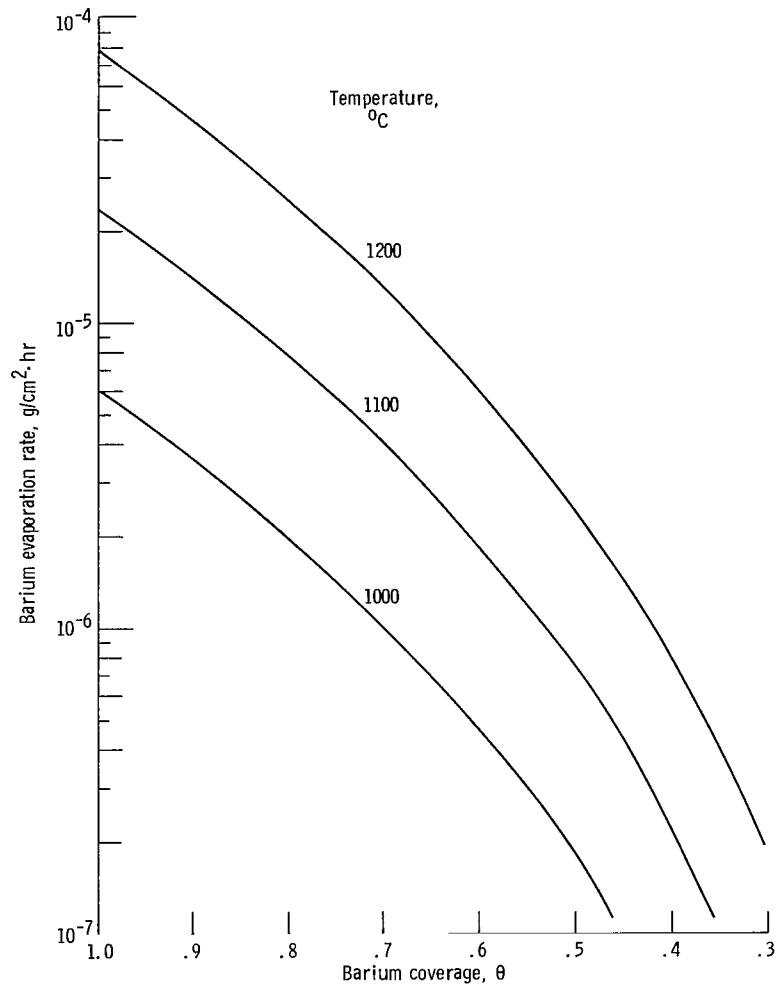


Figure 13. - Evaporation rate of barium as function of coverage.

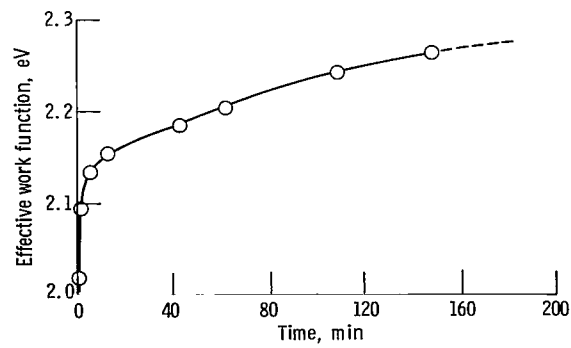


Figure 14. - Variation of effective work function of barium on oxygen on tungsten surface, initially containing monolayer of barium, with time. Surface temperature, 1100 K.

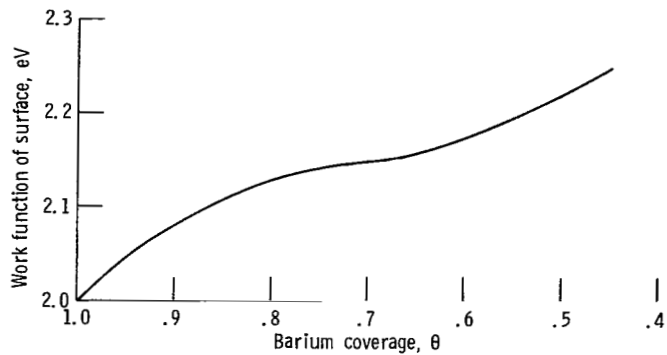


Figure 15. - Work function of barium on oxygen on tungsten surface as function of coverage at 1100 K.

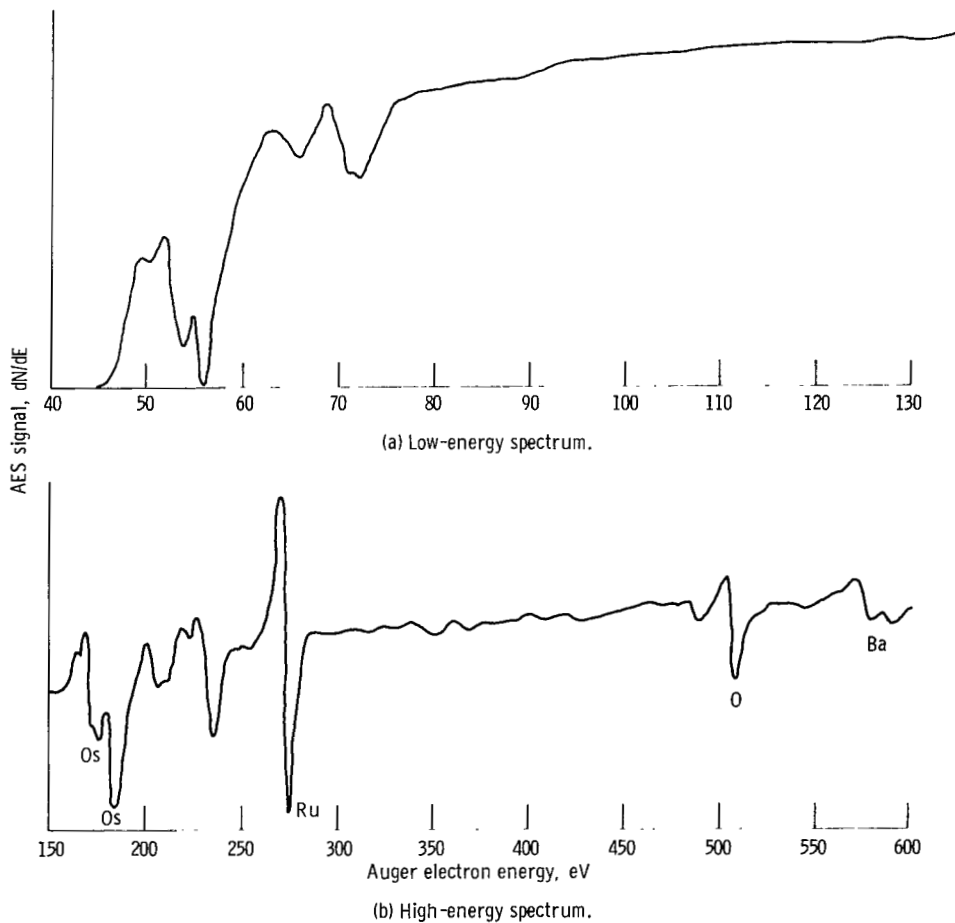


Figure 16. - Auger spectra for activated M-cathode.

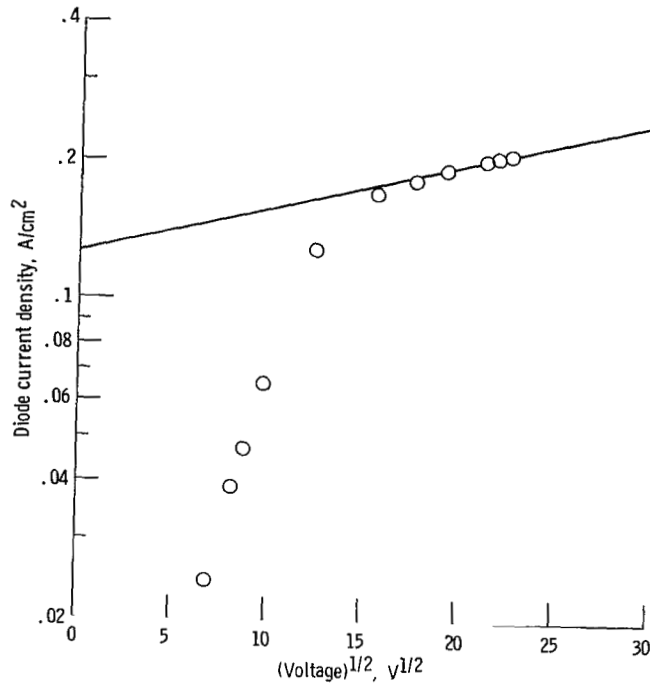


Figure 17. - Schottky plot for M-cathode after initial activation, operating at 1010 K.

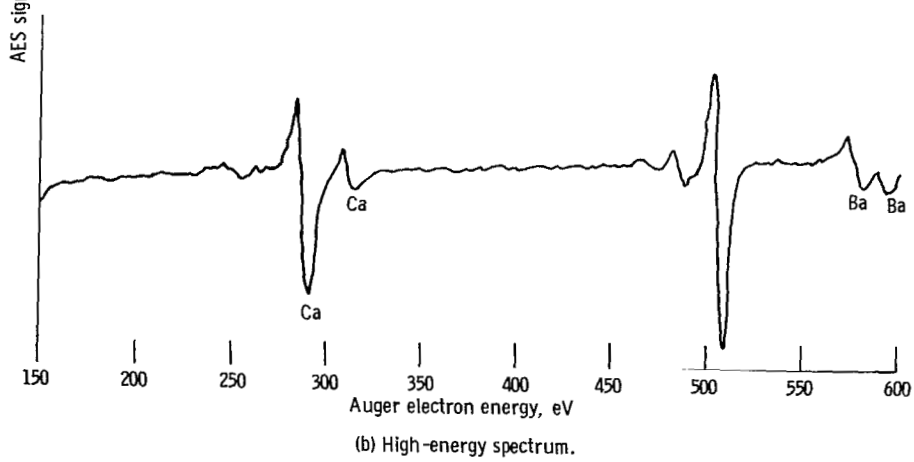
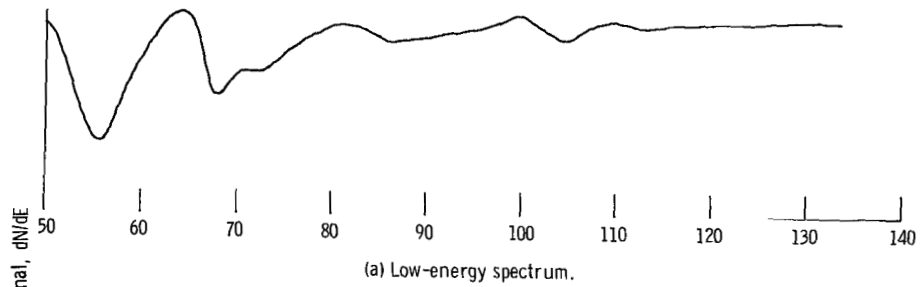


Figure 18. - Auger spectra for activated CPC cathode.



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