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Publication Date

2013-09-09

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Large-area lanthanum hexaboride electron emitter

D. M. Goebel, Y. Hirooka, and T. A. Sketchley

University of California, Los Angeles, School of Engineering and Applied Science, Los Angeles, California 90024

(Received 2 April 1985; accepted for publication 10 June 1985)

A large-area cathode assembly which is capable of continuous, high-current electron emission is described. The cathode utilizes an indirectly heated lanthanum hexaboride (LaB_6) disk as the thermionic electron emitter. The LaB_6 cathode emits over 600 A of electrons at an average of 20 A/cm² continuously with no observable lifetime limits to date after about 400 h of operation in a plasma discharge. Proper clamping of the LaB_6 disk is required to avoid impurity production from chemical reactions with the holder and to provide adequate support if the disk fractures during rapid thermal cycling. Modification of the LaB_6 surface composition due to preferential sputtering of boron by hydrogen and argon ions in the plasma discharge has been observed. The surface appearance is consistent with the formation of LaB_4 as a result of boron depletion. The electron emission capability of the cathode is not significantly altered by the surface change. This surface modification by preferential sputtering is not observed in hollow cathodes where the ion energy from the cathode sheath voltage is typically less than 50 V. The electron emission by the cathode has not been affected by exposure to both air and water during operation. Utilizing thick disks of this intermediate temperature cathode material results in reliable, high-current, long-lifetime electron emitter assemblies.

INTRODUCTION

Large-area cathodes capable of high current density electron emission are required for many applications such as in ion sources,¹ plasma generators,² electron beam accelerators,³ and ion lasers.⁴ In these and many other applications, continuous operation with long cathode lifetimes are desirable. Refractory metal cathodes require high heater power in most geometries, and have lifetimes limited by high evaporation rates at high current densities. Barium oxide impregnated tungsten dispenser cathodes are susceptible to poisoning,⁵ especially during vacuum accidents. Lanthanum hexaboride exhibits many of the desirable cathode qualities⁶ such as high emission current densities, low evaporation rates, and resistance to poisoning in vacuum accidents. The use of LaB_6 has increased in recent years as techniques to hold the very reactive material are developed, resulting in LaB_6 filaments⁷ and hollow cathodes.⁸ Lanthanum hexaboride is somewhat fragile, however, and is easily fractured by thermal shock if heated or cooled rapidly.⁹ Problems with fracturing and impurity production have previously limited the use of LaB_6 electron emitters in high-current applications where large areas are needed.

In this paper, we discuss the characteristics of lanthanum-boron thermionic electron emitters, and describe a large-area, continuously operating cathode assembly and heater. Impurity production and structural problems with support of the LaB_6 have been eliminated in the present cathode configuration. The performance of the cathode in a plasma discharge, where surface modification by ion sputtering occurs, is presented. Problem areas which affect lifetime and emission current capability are discussed.

I. ELECTRON EMITTER CHARACTERISTICS

Lanthanum hexaboride was first described by Lafferty⁶ for use as an electron emitter. The thermionic emission of lanthanum-boron compounds, as a function of the surface stoichiometry, has been extensively studied by Storms and Mueller,¹⁰ and several others.¹¹⁻¹³ The La-B system can consist of LaB_4 , LaB_6 , LaB_9 , or combinations of these compounds. The color of the cathode surface is characteristic of the composition,¹⁴ with LaB_6 having the classical purple color, LaB_4 being grey, and LaB_9 appearing blue. The effective work function of La-B compounds has been reported to range from 2.52 to 3.35 eV,¹⁰ with the higher values corresponding to an increase in the boron content of the surface layer. The large range in the work functions reported has been attributed to both a composition variation of the emitters tested^{10,13} and the presence of surface impurities.¹⁵ The electron emission from single crystals and various crystallographic planes is not discussed here because polycrystalline press sintered LaB_6 is used.

The electron emission current density as a function of temperature for LaB_6 , tungsten, tantalum, lanthanum-molybdenum (LaMo),¹⁶ and barium-oxide dispenser¹⁷ cathodes is shown for comparison in Fig. 1. The curves for W and Ta are calculated from the Richardson-Dushman equation for emission constants found in Kohl.¹⁸ The values for LaMo and BaO impregnated tungsten dispenser cathodes are from the experimental data in the references. The emission current density for LaB_6 is calculated assuming standard values of the work function of 2.7 eV and a Richardson constant of 30 A/cm² K². At intermediate temperatures of up to 1800°C, emission current densities of over 40 A/cm² are

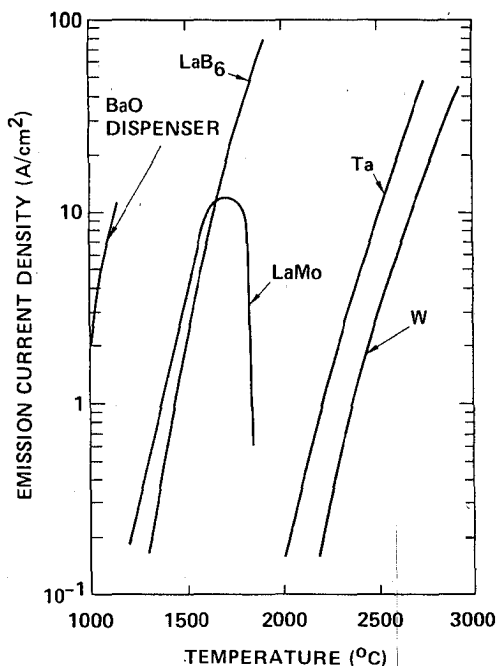


FIG. 1. Electron emission current density vs temperature for LaB_6 , tungsten, tantalum, lanthanum-molybdenum, and barium-oxide impregnated tungsten dispenser cathodes.

available from LaB_6 . The other moderate temperature cathodes, LaMo and BaO dispenser, have maximum emission current densities of about 12 A/cm^2 . The BaO dispenser cathodes can be damaged by repeated exposure to air, so they were not selected for use in our experiments. The LaMo cathode material has been utilized in hollow cathodes,^{1,19} and is further described in another article.²⁰ Of these familiar cathode materials, LaB_6 provides the highest emission current density at the lowest temperature.

The evaporation rate of LaB_6 can be found in the literature.^{6,7,14} Compared to standard refractory metal cathodes, the evaporation rate of LaB_6 is significantly lower at equal emission current densities. Freely evaporating LaB_6 is congruently vaporizing. At the high temperatures above 1600°C used in this work, this results in a surface composition of about $\text{LaB}_{6.05}$, independent of the bulk stoichiometry over the range of $\text{LaB}_{5.3}$ to $\text{LaB}_{8.7}$.¹⁰ A surface of LaB_6 results after sufficient time at high temperatures with free vaporization for the surface and vapor composition to come into equilibrium.

Lanthanum hexaboride is very reactive with refractory metals which might be used to support the cathode at emission temperatures. Typically, boron from LaB_6 diffuses into the refractory metal lattices and forms interstitial boron alloys. Tungsten-boron and molybdenum-boron compounds were detected in our early cathode geometries which supported the LaB_6 with these refractory metals, and appeared as a thick white powder on the surface of the emitter. Utilizing supports made of rhenium or tantalum-carbide minimizes these chemical reactions, and the use of graphite totally eliminates reaction problems. Studies of ternary systems of LaB_6 and refractory metal borides²¹ indicate that the work function of LaB_6 increases as the concentration of the refractory boride is raised. Molybdenum has been identified

as particularly poisonous to LaB_6 , forming molybdenum borides on the surface and increasing the work function. However, the high electron-emission current densities for LaB_6 in Fig. 1 are calculated with the same work function as that associated with a surface that was slightly poisoned by molybdenum.¹⁵ A clean LaB_6 surface, with the molybdenum removed by dissociated halogens, was reported in this reference to have a work function of 2.36 eV and a Richardson constant of $120 \text{ A/cm}^2 \text{ K}^2$. This corresponds to a calculated increase in the emission current density compared to Fig. 1 by over a factor of 5 at 1700°C . The low work function observed^{15,21} despite the presence of some surface impurities might explain the ability of LaB_6 to perform well in environments which might be expected to poison the surface. Electron emission ultimately fails at temperatures above 1600°C when thick impurity layers are formed. Oxidation of LaB_6 raises the work function,¹⁰ and can produce white La_2O_3 on the surface.¹³ Heating of the material to over 1500°C in a vacuum of 10^{-6} Torr or better restores the characteristic purple color and the electron emission.¹²

Large lanthanum hexaboride pieces are fabricated by press sintering powder in molds. The material can be ground after fabrication with ceramic tools, but is extremely brittle and tends to crack and break. Fracturing of the material during use is quite common if it is clamped too strongly or thermally stressed by rapid heating or cooling. In hollow cathodes,^{8,9,22} rods and well rounded crucibles were used to decrease breakage by using shapes with low stress. In addition, supports with spring clamps are desirable to minimize stress during thermal expansion. Graphite has a thermal expansion similar to that of LaB_6 and maintains spring at the temperatures of interest, making it a good clamping material. Directly heated filaments of LaB_6 cut from solid rods and clamped by rhenium supports have recently been developed.⁷ Large area LaB_6 cathodes must be indirectly heated by radiation or electron bombardment because the resistance of the material is too low for direct heating of practical thicknesses and shapes other than filaments. In addition, LaB_6 has an emissivity of about 0.8, and fairly large heater powers are required to offset the radiated power at the electron emission temperatures.

II. CATHODE GEOMETRY

The cathode configuration consists of electron emitting disks²³ 7.6 cm in diameter and 0.64 cm thick which are indirectly heated by tungsten filaments. The LaB_6 disk cathode is shown in Fig. 2. The disk is clamped between concentric rings of Ta with springs to provide for thermal expansion. A 6.3-cm-i.d. ring is electron beam welded to a 0.5-mm-thick Ta skirt of about 0.5-mm-larger diameter than the disk, forming a loose hood. The second ring slips inside the skirt and presses the disk into place inside the hood. Graphite paper 0.25-mm thick is sandwiched between all Ta pieces and the LaB_6 . This small amount of graphite protects the disk against chemical reactions with the Ta, provides some space for radial thermal expansion, and is shielded from plasma or high-voltage electrodes by the Ta hood during operation of the cathode. In addition, the graphite carbur-

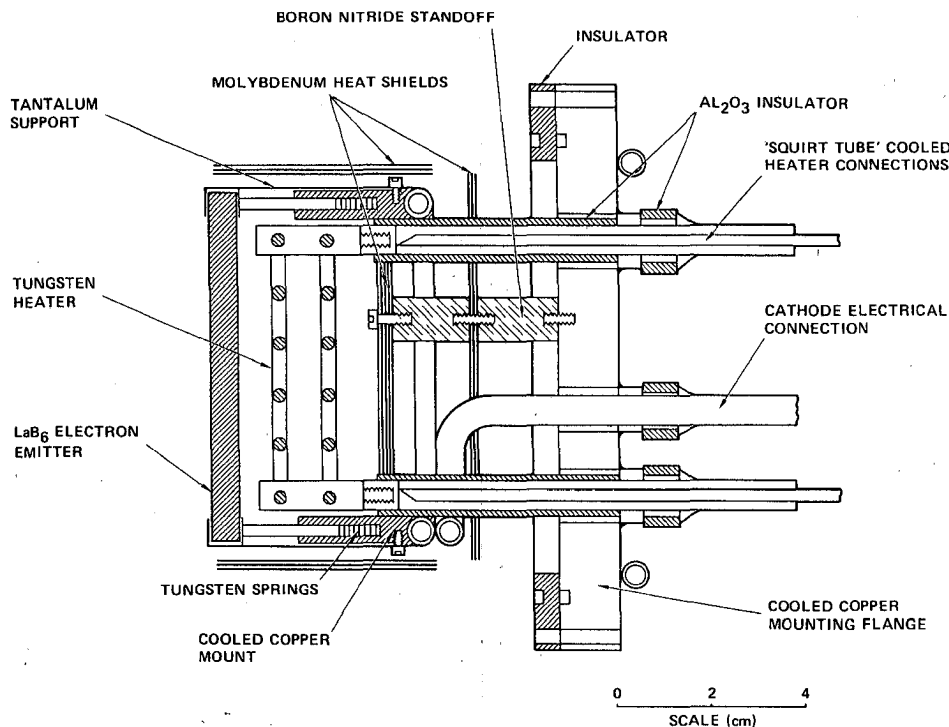


FIG. 2. LaB_6 cathode assembly.

izes the inner Ta surfaces that face the LaB_6 , resulting in further protection against chemical reactions. The LaB_6 disk and second Ta ring are pushed against the hood by eight Mo rods and tungsten springs inserted into holes in the copper mounting ring. The rods and springs are machined such that when the hood is slipped over the water-cooled Cu support and clamped by screws, the LaB_6 is held firmly in place. The exposed area of the LaB_6 emitter is 31.6 cm^2 .

The heater for the cathode, shown in Fig. 3, has two 0.32-cm-diam tungsten filaments clamped in parallel into squirt tube water-cooled Mo connectors. Several layers of 0.13-mm-thick Mo or Ta foil are utilized to heat shield the back and sides of the cathode structure. The heater operates

typically at 17 V, 510 A to heat by radiation the LaB_6 electron emitter to about 1650°C , as determined by an optical pyrometer. This heater arrangement is very reliable and has a long lifetime because the tungsten surface temperature is only about 2100°C . Attempts to utilize electron bombardment heating by biasing filaments negative with respect to the cathode were not very efficient because the electrons also heated the support structure which could not be easily insulated. The radiative heating has worked perfectly for hundreds of hours to date, and has not required a single filament change.

III. EXPERIMENTAL PERFORMANCE

The cathode is intended for use in a continuously operating plasma generator,² and is tested in this device as shown in Fig. 4 for electron emission performance in the presence of a hydrogen or argon plasma. The cathode is mounted to the end of a cooled copper tube 15 cm in diameter and 50 cm long which is insulated and coupled to a 22-cm-diam stainless-steel vacuum system. An axial magnetic field of up to 2 kG can be applied by external coils if desired. The copper tube acts as the anode in these tests, with only the electron emitting disk and its support biased to cathode potential. The experimental procedure is to outgas the cathode, apply the discharge voltage, inject Ar or H_2 gas, and monitor the discharge voltage, discharge current, pressure, and current to several Langmuir probes in the plasma. The discharge current from the cathode during continuous operation is then increased by turning up the discharge power supply, up to a maximum current of about 650 A.

The LaB_6 disks usually have a purple-blue color when delivered from the manufacturer, characteristic of a B/La ratio greater than six. The cathode is heated above 1600°C

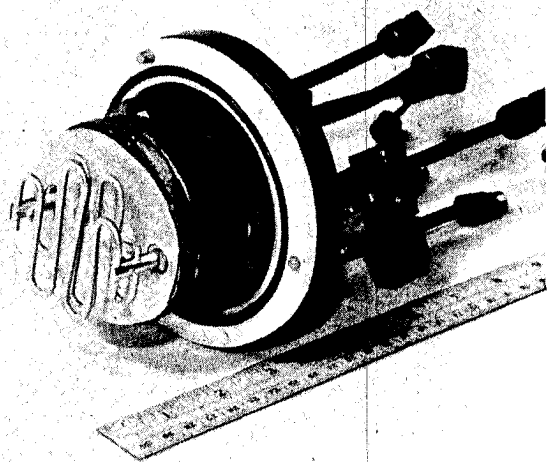


FIG. 3. Cathode heater consisting of two serpentin tungsten filaments. Uniform heating of the disk was achieved by indirect heating from these large filaments.

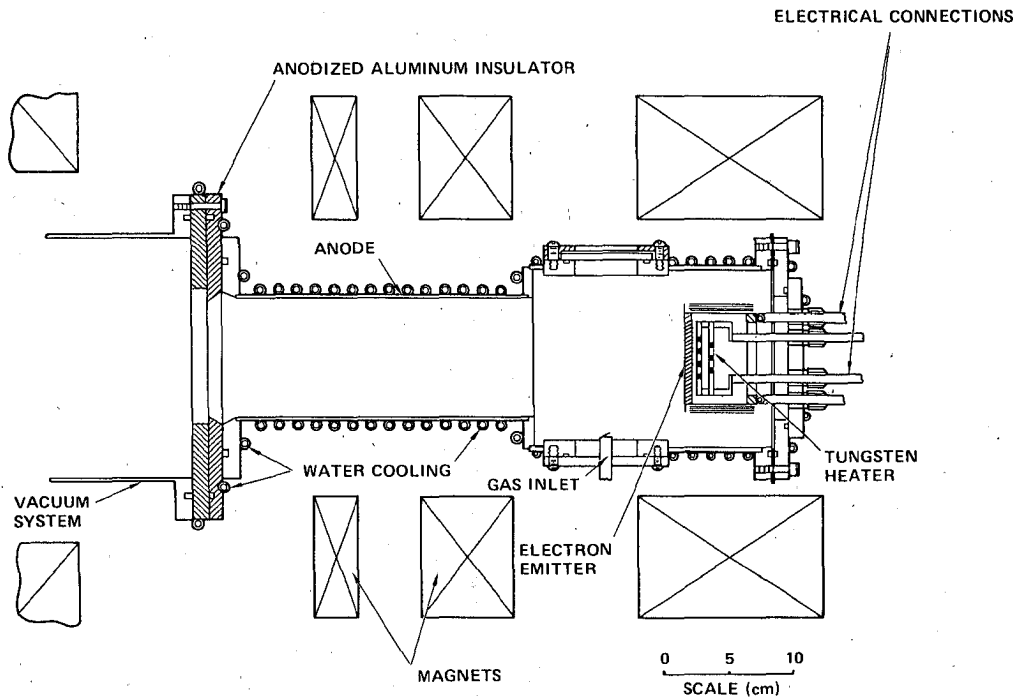


FIG. 4. Plasma generator test chamber for cathode performance study.

and outgassed for several hours after assembly in a separate vacuum chamber until the pressure returns to the 5×10^{-7} Torr base pressure obtained before heating. The disk develops the purple color of LaB_6 during this initial heat treatment. After mounting in the test chamber, the LaB_6 cathode is then heated to about 1650°C over a period of 15 min. The chamber pressure returns to the base pressure in a matter of minutes after the operating temperature is reached for a previously outgassed cathode. The current-voltage performance of the cathode is shown in Fig. 5 for two different fill pressures indicated in hydrogen. A maximum discharge current of 670 A has been achieved. The performance in Ar at 2×10^{-4} Torr is similar with a slightly lower discharge voltage at a given current. The discharge voltage is determined by the gas pressure, discharge power supply level, and magnetic field strength at the anode. During these tests, the gas pressure is varied by up to a factor of 4 and the magnetic field varied from 0 to 1000 G to ensure that the electron emission is not space charge limited. This will be discussed further in Sec. V. The maximum electron emission for the LaB_6 cathode is ultimately limited by the discharge power supply capability in these experiments.

The actual emission current density must be corrected for ion current to the cathode, and this will also be discussed in the Sec. V. At discharge currents above 400 A, the heater power can be reduced to below 7 kW without altering the discharge level. Heating of the cathode by ion bombardment from the plasma provides additional power to raise the temperature of the LaB_6 to over 1700°C necessary for up to 20 A/cm^2 of emission. Measurements of the actual surface temperature after plasma bombardment by the optical pyrometer are not reliable because of a possible emissivity change associated with surface modification. The heater powers are selected based on temperature measurements made before plasma discharges. This will be discussed further in Sec. IV.

The discharge power level was cycled several times from 0 to 60 kW over a period of 15 min each without fracturing the disk. The mounting assembly and clamping system permit adequate thermal expansion to avoid breakage. The discharge power supply was then interrupted at the 50-kW power level, resulting in a termination of the discharge in less than 5 ms. The ion current accelerated through the 100 V sheath to the cathode before the interruption was about 50 A, as estimated from the Langmuir probes. The interruption of the discharge decreased the power to the cathode by about 5 kW. The disk immediately developed four radial fractures

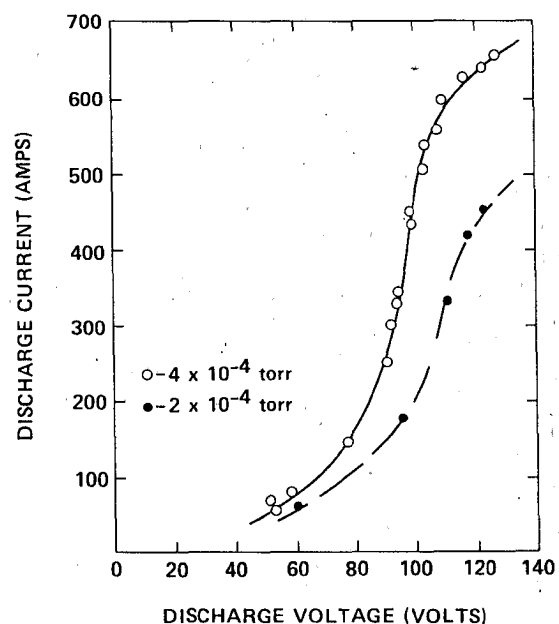


FIG. 5. Discharge current vs voltage for the LaB_6 cathode at two gas pressures measured in the vacuum system.

associated with this rapid change in the heating of the cathode and the resultant thermal stress of the material. These fractures do not change the cathode performance, however, because the clamping system on the edge of the disk still holds the pieces in place. The cathode has been used over 300 h since this event, and did not develop additional cracks. A second LaB₆ disk performed identically, except to form five fractures when thermally stressed. The pieces of the first disk have been reassembled and used again with identical emission performance. It appears that the fractures relieve the stresses in the disk and no further damage occurs. As long as only radial cracks occur, the clamping system maintains the cathode integrity. It has been observed that during heating the disk expands and closes any openings at the cracks, presenting a complete surface to the plasma again.

IV. SURFACE MODIFICATION

When examined after about 10 h of use in the plasma generator, the LaB₆ disk had a grey-silver color over the majority of the center region, and the normal purple color at the edge near where it was clamped. It appeared as if refractory metals from the cathode supports and probes, or stainless steel from the vacuum vessel, had coated the center of the disk where the plasma flux is a maximum. However, Auger analysis indicated that the LaB₆ disk has no significant metal impurities on the surface. The Auger spectrum for the normal purple colored edge region is shown in Fig. 6(a), and the spectrum from the grey-silver center region is shown in Fig. 6(b). The Auger spectra indicate that the B/La ratio has decreased in the region of plasma bombardment. No significant quantities of metal impurities are observed. The boron depletion and color change in this center area suggest¹⁰ the formation of LaB₄. During ion bombardment in these plasma tests, the cathode surface is not freely vaporizing to produce the steady state LaB₆ surface. Ions striking the cathode surface in our tests are 100-V protons, which preferentially sputter the boron atoms with a similar mass. The surface modification of the cathode required a fluence of over 10²³ ions/cm² in these experiments. Oxidation or hydrogen reduction of the LaB₆ surface cannot be responsible for the formation of LaB₄ because the edge region of the disk with a lower ion flux was not modified.

The LaB₄ region is much harder than LaB₆, and cannot be easily removed from the bulk material by peen blasting or grinding. We reproduced this boron depletion and grey-silver color on the purple edge region of the disk with a long exposure of 5 kV argon ions from the Auger depth profiling analysis. The formation of LaB₄ has not been observed on LaB₆ electron emitters used in hollow cathodes in the plasma generator. The ion energy striking the emitter surface in these hollow cathodes is typically only 20–30 V,⁸ which is below the threshold for preferential boron sputtering to occur. The hollow cathode serves to protect the emitter surface from sputtering by high-energy ions. The characteristic refractory metal-boron compounds associated with boron reactions with support materials have not been detected on the disk cathode described here.

The disk cathode with the modified surface of LaB₄

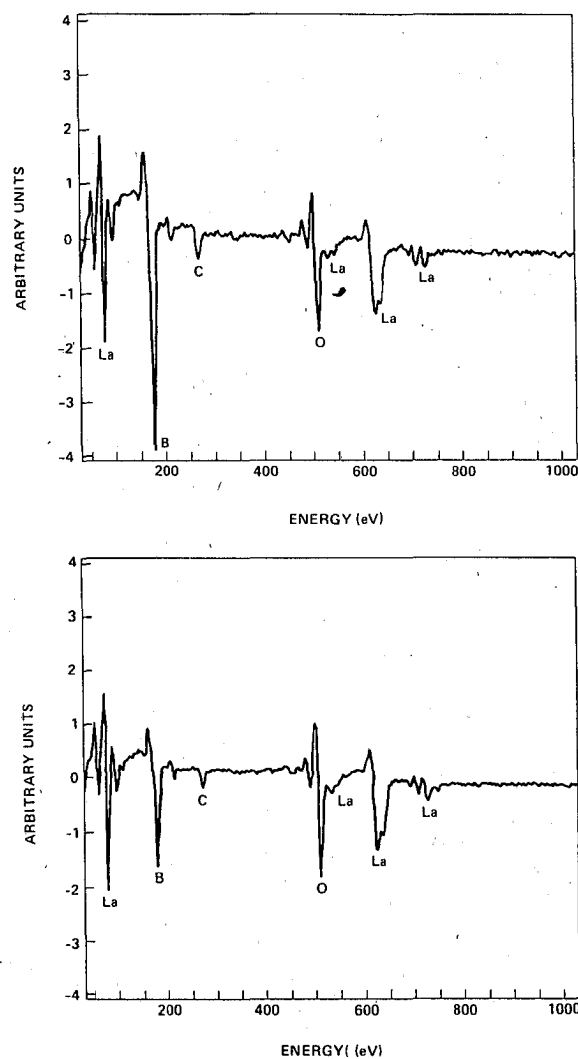


FIG. 6. Auger spectrum from the normal purple-colored edge region (a), and from the metallic center region (b). Preferential sputtering of boron by the plasma discharge and the formation of LaB₄ is observed from the decreased boron concentration and color change in the center region of the disk where the ion flux is a maximum.

+ LaB₆ is still an excellent electron emitter. Measurements indicate that the work function actually decreases as the B/La ratio decreases.^{10,11} During the 10 h over which the damage occurred, the heater power had to be raised in our experiments from the initial level by about 15% to 9 kW to maintain a constant discharge current. The new temperature of the disk could not be determined accurately with the optical pyrometer because of the possibility of an unknown emissivity change in the modified surface. However, the highest currents reported here were achieved with the modified surface and fractured disk.

DISCUSSION

The actual cathode electron emission current can be considered to be the discharge current minus the plasma ion current incident on the cathode surface. For space-charge neutralization, the ion current must be at least the square root of the mass ratio times the electron current, but can be more than this value. In hydrogen at 700 A of electron cur-

rent, for example, this corresponds to about 16.3 A of total ion current to the emitting surface area. A cooled Langmuir probe inserted into the plasma indicated that an ion current density of 1.6 A/cm² was actually incident on the emitting surface for the 670 A discharge. This corresponds to about 50 A of ion current, which is well in excess of that required for space-charge neutralization. For positive plasma potentials relative to the anode measured by the probe, ion current is also lost to the anode. This loss might offset any quoted emission current error resulting from ion current to the cathode. Neglecting anode ion current and taking the case of discharge current minus ion cathode current as the emission current, then the LaB₆ cathode emits 620 A at an average current density of 19.7 A/cm². The discharge current did not increase with heater powers above 9 kW, and the heater could be decreased during operation. The neutral gas density, heater power, and anode area were increased at full current to ensure that the maximum emission current was not limited by space-charge effects. The cathode produces discharge currents at the maximum of the power supply capability, and it is anticipated that more electron current is available from this cathode.

The cathode was exposed to air and water during operation at discharge currents of 500 A. Subsequent outgassing for about 30 min after reestablishing the base pressure restored the cathode operation again without any observable change in the emission capability. The modification of the LaB₆ surface, due to preferential sputtering of boron to form LaB₄, occurred after a high fluence had been accumulated. It is possible that this preferential sputtering has not been previously observed because of impurity erosion or acculimation in other experiments that might have had the necessary fluence of relatively low-energy light ions. Surface analysis detected no impurities on the cathode surface which might sputter the material, inhibit electron emission, or eventually appear in the plasma. The LaB₆ disk has been used for an estimated time of about 400 h in the plasma generator without requiring any cleaning or maintenance. During this time, the vacuum system was vented to air over one hundred times with the cathode cold, and several times with the cathode hot, without observable change in the cathode performance.

The LaB₆ cathode has been extremely reliable in spite of the large fractures. The spring clamps on the edge of the disk hold the pieces together very effectively. If a very flat emitting surface is needed, then a thin boron nitride backing disk

between the LaB₆ and the heater can be used.²⁴ A new LaB₆ disk with a coarse mesh of graphite or rhenium imbedded into it during the press sintering process is being ordered to provide some strength against fracturing. Tests of this new system will be reported in the near future.

ACKNOWLEDGMENTS

We would like to thank Prof. R. W. Conn for supporting this work, and K. Andrews and V. Low for technical assistance. This research is supported by U.S. Department of Energy under Contract No. P.A. DE-AT03-84ER52104.

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