Dynamic behavior of thermionic dispenser cathodes under ion bombardment

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We have investigated the surface coverage and electron emission of thermionic dispenser cathodes during 3 keV Ar+ ion bombardment, thereby simulating the bombardment of the cathodes by residual gases that takes place in cathode-ray tubes as used in television sets. During the ion bombardment at the operating temperature of 1030 °C, a dynamic equilibrium is established between the sputter removal and resupply mechanisms of the Ba and O atoms that form the dipole layer on the cathode substrate. We demonstrated that the performance of the cathodes under ion bombardment is governed by the O removal and resupply rates. It was found that the Ba resupply rate is almost an order of magnitude higher than the O resupply rate, but that the Ba can only be present on the surface bound to O atoms. Therefore, the Ba/O ratio is approximately equal to unity during the ion bombardment. Based on the investigations of the removal and resupply processes, we proposed a model that accurately describes the surface coverage and electron emission during the ion bombardment, including the dependence of the ion flux and cathode temperature. © 2001 American Institute of Physics. [DOI: 10.1063/1.1356433]

I. INTRODUCTION

Oxide cathodes consisting of the oxides of earth alkali elements (Ba, Sr, and Ca) have been employed for many years as electron sources in cathode-ray tubes used in television sets.1,2 The cathodes consist of a thick oxide layer with a low work function of approximately 1.3 eV, and can deliver continuous electron emission densities of the order of 1 A cm−2 at an operating temperature of 700–800 °C.

With the introduction of large high-quality tubes and high-definition television, the demand arose for higher emission densities exceeding the capabilities of the oxide cathodes. Emission densities of 1 to 2 orders of magnitude higher than the limit of the oxide cathodes can be obtained from thermionic Ba dispenser cathodes, which are also used in high-power microwave tubes. These dispenser cathodes consist of a porous W matrix impregnated with a BaO containing compound.3 Figure 1 shows a secondary electron microscope image of the porous matrix, showing the individual W granulates and the impregnated pores. During cathode operation a Ba–O monolayer is formed on the substrate with a Ba/O ratio approximately equal to unity.4 This electropositive dipole layer results in a work function of approximately 2 eV. Although this work function is higher than the extremely low work function of the oxide cathode, the metallic conductivity of its body enables cathode operation at temperatures of 1000 °C to compensate for the higher work function. Note that there is thus a fundamental difference between the oxide and dispenser cathode: the low work function of the dispenser cathode is obtained by a monolayer Ba–O complex, while the low work function of the oxide cathode is due to a thick oxide layer.

The Ba–O complex responsible for the low work function can be removed from the surface by thermal evaporation or through ion bombardment by residual gases in the tube. Residual gas atoms, ionized by the high density electron beam, are accelerated onto the cathode surface due to the electric fields present in the electron gun. The impinging rest gas ions sputter the Ba and O atoms from the surface. In order to maintain the near monolayer Ba–O complex essential for the emission properties, the loss of the Ba–O complex is replenished by surface diffusion from the pores. However, because the required diffusion length is of the order of 10 μm for the dispenser cathodes, ion bombardment can strongly influence the emission densities. In addition to removal of the Ba–O complex, the ion bombardment can also damage the substrate of the cathode. Substrate damage is especially important for top-layer dispenser cathodes, where the emission is further enhanced by coating the W matrix by an additional metal (e.g., Os, Re, and Ir) or metal/oxide (e.g., Re/Sc2O3) layer of the order of 300–500 nm.5 While the removal of the Ba–O complex can be restored by resupply from the pores, damage to the coating leads to gradual and permanent long-term emission degradation.

The low resistivity of the Ba dispenser cathodes to the ion bombardment is an important drawback of these cathode...
The importance of ion bombardment resistivity is most evident for the scandate cathode which has a coating containing a $\text{Sc}_2\text{O}_3$ complex. Although the scandate cathode has an enormous emission potential of up to 400 A cm$^{-2}$, its breakthrough for applications has not been possible so far due to its strong susceptibility to ion bombardment damage and emission degradation.\(^5\)

In order to solve the problem of the low ion bombardment resistivity of the dispenser cathode, and especially for scandate cathodes, more insight is needed in the behavior of the cathodes during the bombardment. Although several studies have been reported on this topic, most of the studies have focussed on studying merely the influence of the bombardment on the electron emission.\(^6\) From these emission measurements, one often indirectly draws conclusions about the surface composition. Here, we have combined surface analysis techniques with emission measurements in order to investigate the surface composition and electron emission during the ion bombardment of thermionic dispenser cathodes. We will show that the generally accepted belief that the Ba removal and resupply rates determine the behavior of dispenser cathodes during ion bombardment is incorrect. Instead, we show that if an ample supply of Ba is present in the pores, the O removal and resupply rates actually determine the Ba–O surface coverage and electron emission under bombardment.

The outline of this article is as follows. First, we give a brief description of the experimental techniques used to determine the surface composition and electron emission (Sec. II). Subsequently, the results are presented and discussed (Sec. III). A demonstration of the dynamic behavior of the dispenser cathodes during the bombardment is given in Sec. A. In order to come to a full description of the dynamic behavior, we then separately discuss the Ba–O removal and resupply processes (Sec. B and C, respectively). The surface coverage and electron emission at the dynamic equilibrium as a function of the incident ion flux and cathode temperature are subsequently discussed (Sec. D). Furthermore, the experimentally determined coverage and emission are compared to a model that describes the Ba–O coverage during the ion bombardment, and is based on the separately determined removal and resupply rates. In addition to the influence of the ion bombardment on the Ba–O adlayer, we also investigated the damage to the cathode substrate that leads to unrecoverable long-term emission degradation (Sec. E). Finally, an ion bombardment experiment in an $\text{O}_2$ background pressure is performed in order to emphasize the importance of the O resupply during the ion bombardment (Sec. F). The main conclusions of this work are summarized in Sec. IV.

II. EXPERIMENT

The two main constituents of the residual gas present in the cathode-ray tubes are Ar and $\text{N}_2$. When ionized, the ions are directed onto the cathode surface with an average kinetic energy of the order of 3 keV due to the electric fields in the electron gun. Thus, in order to simulate the ion bombardment that takes place in cathode-ray tubes, the dispenser cathodes are bombarded by 3 keV $\text{Ar}^+$ ions (similar behavior for $\text{N}_2$). The ion bombardment is performed in a ultrahigh vacuum (UHV) apparatus using a differentially pumped ion gun. The ion beam is directed perpendicular onto the cathode, delivering a uniform ion dose over its surface. The dispenser cathodes used in the investigations were experimental Philips Os/Ru-I cathodes, where the surface of the W matrix with a diameter of 1 mm is coated with Os (80%)/Ru (20%) top layers of 20 to 500 nm. (The coating on which the Ba–O complex is adsorbed and will further be referred to as the substrate.) The matrix is impregnated with a 4:1:1 mixture of BaO, CaO, and $\text{Al}_2\text{O}_3$. The Os/Ru-I cathode is capable of emission densities of the order of 10–20 A cm$^{-2}$ at its operating true temperature of 1030°C.

The surface composition during the ion bombardment is analyzed by two different methods. The composition of the outermost atomic layer of the surface was analyzed using low-energy ion scattering by detecting the $\text{Ar}^+$ ions that have backscattered from the cathode surface.\(^7\)–\(^10\) Because of the high electron affinity of the $\text{Ar}^+$ noble gas ions, the neutralization probability during an interaction with the surface atoms is very high. Ions that penetrate into the cathode beyond the outermost atoms, have multiple interactions and are therefore almost certainly neutralized. Here, we have measured the $\text{Ar}^+$ ions that have backscattered over an angle of 136°. These ions are energy analyzed by a cylindrical mirror analyzer (CMA) and detected using a microchannelplates based detector.\(^11\) The energy spectrum thus obtained represents a mass spectrum of the outermost atomic layer during the bombardment.

In addition to analyzing the backscattered $\text{Ar}^+$ ions, Auger electron spectroscopy (AES) was used to determine the Ba and O coverage during the ion bombardment. For the Auger analysis a grazing incidence electron gun is used, where the electron beam is directed onto the surface at an angle of approximately 10° with the cathode surface. The grazing incidence minimizes the contribution from the pores, and thus enables a more accurate investigation of the emitting surface compared to perpendicular incidence. The Auger electrons are analyzed by the same CMA as used for the ion scattering, but with a reversed polarity of the pass energy.

The electron emission of the cathodes during the ion bombardment was determined in a separate setup employing a close spaced diode configuration,\(^5\)\(^12\)\(^13\) with a distance of

![FIG. 1. Secondary electron microscope image of the dispenser cathode, showing the porous matrix and the impregnated pores. The black and white bars at the bottom have a length of 10 µm.](image)
approximately 300 µm between the cathode and the anode. The ion beam was directed onto the cathode through a circular hole in the anode with a diameter of 2 mm. A high-voltage saw-tooth pulser was used to apply a diode voltage of 800 V, and the electron emission was determined in the saturation regime. The emission current was continuously monitored before, during, and after the ion bombardment.

III. RESULTS AND DISCUSSIONS

A. The dynamic equilibrium

A demonstration of the dynamic behavior of the dispenser cathodes during the ion bombardment is given in Fig. 2. This example shows the emission of an Os/Ru cathode bombarded at operating temperature of 1030 °C by a flux of $6 \times 10^{13}$ ions cm$^{-2}$ s$^{-1}$ Ar$^+$ ions. The emission is normalized to the emission during operation in an UHV in the absence of ion bombardment. Three stages can be identified in Fig. 2: the formation of the dynamic equilibrium (I), the dynamic equilibrium (II), and the recovery after the ion bombardment (IB) is stopped (III). At the equilibrium, the emission complexity removal rate equals the resupply rate, and the surface coverage and emission are constant.

The removal rate depends on the incident ion flux $F$ and the number of atoms removed per incident ion. The resupply is governed by surface diffusion of Ba and O atoms, and depends on the cathode temperature. The equilibrium coverage under ion bombardment thus depends on the ion flux to which the surface is subjected, and the temperature at which the cathode is operated. This is illustrated in Fig. 3 which shows ion scattering spectra under the dynamic equilibrium for different ion fluxes $F$ at a fixed operating true temperature of 1030 °C. At a very low ion flux (a), no substrate atoms are observed in the outermost layer, indicating a complete coverage of the substrate. With increasing ion flux the removal rate increases, while the resupply rate determined by the temperature remains constant [(b) and (c)]. Consequently, the substrate is no longer completely covered by the Ba–O complex and substrate peaks due to Ru, W, and Os appear in the ion scattering spectra. These spectra serve as qualitative examples to demonstrate the change in surface coverage with increasing ion flux. Quantitative analysis is not straightforward due to the dependence of the neutralization probability of the Ar$^+$ ions on the work function, as is discussed elsewhere.14,15

B. The sputtering process

Here, we will determine the probability that the Ba and O atoms constituting the emission complex are removed by the impinging ions. Only a small fraction of the adatoms are sputtered directly from the surface due to a collision between the adatom and an incident ion. Most incident ions do not have a hard collision with the outermost Ba atoms, but rather penetrate into the cathode substrate beyond the adlayer. The collisions of the ions with the substrate atoms initiate linear collision chains of the substrate atoms as sketched in Fig. 4.16,17 If a chain of collisions is directed towards the surface, an atom in the outermost layer may be emitted from the surface since it cannot transfer its energy to a subsequent atom. Thus, the actual sputtered atom is only indirectly coupled to the incident ion. The sputter yield $Y$, which is defined as the number of atoms removed per incident ion, depends on the details of the bombardment, such as the incident energy and the incident angle of the ions.18,19

To determine the sputter yield for the removal of the Ba and O adatoms, the cathode surface is bombarded by 3 keV Ar$^+$ ions at room temperature and thus no resupply of the Ba–O complex is possible. Figure 5 shows the Ba and O coverage as a function of the applied ion dose measured by the ion scattering and Auger analysis, where the coverages are normalized to the initial coverages of the undamaged cathode. We emphasize that the initial increase in the ion scattering signal is caused by a decrease in the neutralization
The decrease of the Ba density signal decreases as a function of the applied dose. The decrease is investigated here for the Ba–O complex in the absence of IB (at $t = 0$). The sputter yield can be deduced from the decay constant of the ion scattering and Auger signal (solid curve). For the maximum Ba density, we used $n_{\text{max}} = 6 \times 10^{14}$ atoms cm$^{-2}$, according to investigations of Ba adsorption of W substrates.\textsuperscript{20,21} The obtained sputter yield is $Y = 0.62 \pm 0.07$. This is in good agreement with previously determined sputter yields by van der Gon et al.\textsuperscript{14} ($Y = 0.63$) and Gartner et al.\textsuperscript{22} ($Y = 0.58$), which were deduced directly from ion scattering investigations and indirectly from emission measurement, respectively.

The decrease in the O coverage is slower than that of Ba coverage, indicating that the Ba–O complex is not removed as a whole. This is not surprising since the O atoms are situated in a plane below the Ba atom,\textsuperscript{14,15,23,24} and are predominantly removed after the Ba atoms in the outermost atomic layer have been removed by previous sputtering events. However, it is not unlikely that a small fraction of the O atoms are removed via sputtering of the Ba–O complex as a whole. The middle of the dashed curves in Fig. 5 shows the expected signal decrease assuming an identical sputter yield for Ba and O, and a step-by-step removal of the complex. The other dashed curves indicate the influence of the value of the sputter yield for O on the fit. When the Ba is almost completely removed, the O becomes more difficult to remove by the ion bombardment because the O atoms are more tightly bound to the substrate.

We conclude that the Ba and O adatoms are removed step by step with approximately the same sputter yield. Although these sputter yields were determined at room temperature, the same values apply during the operating temperature since the thermal energy is negligible compared to the energy of the ions.

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C. The resupply process

The Ba–O resupply during the IB is investigated here by studying the reactivation of the cathodes after the Ba–O complex has been removed by sputtering at room temperature. The reactivation procedure is studied at various temperatures between 800 °C and 1200 °C. The surface coverage during the reactivation is monitored by AES. Figure 6 shows the Ba coverage (filled circles) and O coverage (open circles) as a function of the reactivation time for cathode temperatures of 920 °C [Fig. 6(a)] and 1030 °C [Fig. 6(b)]. At low reactivation temperatures, of the order of 900 °C, the Ba resupply is faster than the O resupply in the initial reactivation [see Fig. 6(a)]. However, as the coverage increases, the Ba resupply approaches and follows the O resupply. When the temperature is increased, the initial faster Ba resupply becomes less significant [see Fig. 6(b)]. At high reactivation temperatures, of the order of 1100 °C, the Ba and O resupply curves are identical.

To investigate the role of the oxygen in the resupply process, the reactivation procedure was also performed on preoxidized substrates. After the removal of the Ba–O layer by sputtering, the substrates were exposed to oxygen prior to reactivation. Figure 6 also shows the reactivation of the preoxidized substrates for an initial coverage of $n_0 = 100\%$.
The clean substrate is attributed to the fact that the desorption coverage of approximately 50%. The limited Ba coverage of diffusion across the clean substrate results in a maximum mutual depolarization between the Ba adatoms.\textsuperscript{29}

\[ a \text{ and } b \] given by\textsuperscript{25–27} the Ba adatoms is limited by the residence time, and is represented by the time the Ba spends on the surface before it is lost at approximately the same rate as the Ba resupply takes place.

These observations can be explained as follows. The initial fast resupply of Ba at low reactivation temperatures is caused by Ba diffusion across the clean cathode substrate. The average diffusion length \( \langle l \rangle \) of the Ba atoms is determined by its diffusion rate \( D \) and residence time \( \tau \), which represents the time the Ba spends on the surface before it is thermally desorbed. The substrate area that can be covered by the Ba adatoms is limited by the residence time, and is given by\textsuperscript{25–27}

\[ \langle l \rangle^2 = D \times \tau = D_0 \tau_0 \exp\left[ (Q - E_d)/kT\right]. \]

Here, \( Q \) and \( E_d \) are the activation energies for desorption and diffusion, respectively, and are further referred to as desorption and diffusion energy, respectively. The effective diffusion length for Ba on the clean substrate is evidently insufficient to cover the substrate completely.\textsuperscript{28} This is most clearly seen in Fig. 6(a), where it is observed that the Ba diffusion across the clean substrate results in a maximum coverage of approximately 50%. The limited Ba coverage of the clean substrate is attributed to the fact that the desorption energy \( Q \) strongly decreases with increasing coverage due to mutual depolarization between the Ba adatoms.\textsuperscript{29}

Surface coverage beyond the limit of the diffusion length for Ba on the clean substrate requires the presence of oxygen on the surface. The interaction between the Ba and O atoms increases the desorption energy due to the stronger Ba–O–substrate bond compared to the Ba–substrate bond.\textsuperscript{30–34} Consequently, the diffusion length for Ba is higher in the presence of O, but the Ba resupply rate depends on the arrival of O on the substrate. On the preoxidized substrates, the Ba resupply rate is determined by the diffusion rate of Ba on the oxidized surface, and is not limited by the slower O resupply.

When the temperature is increased, the Ba and O resupply rates increase [see figure 6(b)]. Because the desorption energy is higher than the diffusion energy \( E_d/Q = 0.1–0.2 \), the factor \( (Q - E_d) \) in Eq. (3) is positive. Thus, when the temperature is increased the diffusion length decreases. For reactivation temperatures higher than approximately 1000 °C, the extent to which the Ba can cover the clean substrate is negligible and the Ba resupply is only possible in the presence of O. Therefore, in Fig. 6(b) almost no initial fast Ba resupply is observed compared to the slower O resupply.

Evidence for the Ba–O bonding in the resupply process is presented in Fig. 7. Here, low-energy Auger spectra are shown for the reactivation at a temperature of 920 °C after a reactivation time of 20 s [Fig. 7(a)] and 200 s [Fig. 7(b)]. Because the desorption energy is higher than the diffusion energy \( E_d/Q = 0.1–0.2 \), the factor \( (Q - E_d) \) is positive. Thus, when the temperature is increased the diffusion length decreases. For reactivation temperatures higher than approximately 1000 °C, the extent to which the Ba can cover the clean substrate is negligible and the Ba resupply is only possible in the presence of O. Therefore, in Fig. 6(b) almost no initial fast Ba resupply is observed compared to the slower O resupply.

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supplied in the oxidic form BaO is incorrect.

From the aforementioned observations, we conclude that the resupply of the Ba–O emission complex is limited by the O resupply. The resupply of O atoms is thus equally important as the resupply of Ba atoms, since without the Ba–O bond the cathode substrate can not be sufficiently covered by the dipole complex. The role of O has been underestimated in most studies of dispenser cathodes, where one in general focuses on the Ba adatoms.

The activation energy for resupply of the Ba–O complex can now be determined from the temperature dependence of the O resupply. The diffusion rate of the adatoms is proportional to the concentration gradient (Fick’s first law). Therefore, we introduce the time constant $\tau_{\text{res}}$ for resupply as

$$
\frac{dn}{dt} = \frac{n - n_0}{\tau_{\text{res}}}.
$$

The time constant $\tau_{\text{res}}$ depends on the reactivation temperature. The O coverage during the reactivation is thus given by

$$n(t) = n_0 \left[ 1 - \exp(-t/\tau_{\text{res}}) \right].$$

The solid curves in Figs. 6(a) and 6(b) represent the results of fitting Eq. (5) to the O resupply, where $\tau_{\text{res}}$ was used as a fitting parameter. The temperature dependence of $\tau_{\text{res}}$ is shown in Fig. 8 (open circles). Figure 8 also shows the time constant from the emission recovery after the IB stop (filled circles), which will be discussed in Sec. D. The activation energy for resupply is found to be $E_{\text{res}} = (2.1 \pm 0.1) \text{ eV}$, which is in good agreement with the activation energies for recovery determined by Manenschijn et al.\textsuperscript{38} ($E_{\text{res}} = 2.2 \text{ eV}$) and Gartner et al.\textsuperscript{5} ($E_{\text{res}} = 2.0 \text{ eV}$) determined from emission measurements.

In order to investigate whether the activation energy for the O resupply is determined by the diffusion of O across the substrate, or by the formation of free O in the pores, reactivation of the cathodes was studied after $O_2$ exposure. The cathode was first exposed to 100 L $O_2$ at room temperature, and subsequently the substrate was cleaned by the IB. Howver, an ample supply of O is now present in the pores as follows from the following results. Reactivation was subsequently studied at a temperature of 1030 °C. This procedure was performed for an Os/Ru cathode, as well as unimpregnated W matrix where no BaO containing compound was present in the pores. It was found that the time constants for the O resupply for the impregnated and unimpregnated cathode were $\tau_{\text{res}} = 9 \text{ s}$ and $\tau_{\text{res}} = 7 \text{ s}$, respectively. Reactivation without $O_2$ exposure gives a time constant for the O resupply of $\tau_{\text{res}} = 21 \text{ s}$ [see Fig. 6(b)]. The O resupply is thus faster by approximately a factor of 2.5 if O is abundantly present in the pores. We therefore conclude that the limiting factor in the O resupply is the formation of free O in the pores. Moreover, the activation energy for the resupply derived here is much larger than the activation energy for O diffusion across a W substrate, which is of the order of 1 eV.\textsuperscript{25,26,39} This indicates that for the resupply, more energy is required than merely the diffusion of O from the pores to the surface. The activation energy for the resupply thus represents the activation energy for the formation of free O in the pores. Although the value of the activation energy is in agreement with previous reported studies, the assignment of the activation energy to the formation of O represents a new interpretation which can only be made on the basis of the present surface coverage measurements.

D. Coverage and emission at dynamic equilibrium

The surface coverage at dynamic equilibrium is determined by the removal and resupply rates, and thus depends on the incident ion flux and the cathode temperature. The surface coverage at equilibrium is shown in Fig. 9 as a function of the ion flux for different cathode temperatures (filled symbols). In addition, Fig. 10 shows the equilibrium coverage as a function of the ion flux for a fixed ion flux of $3 \times 10^{13} \text{ ions cm}^{-2} \text{s}^{-1}$. Here, we did not differentiate between the Ba and O coverages because it is observed that the Ba/O ratio is constant during the IB. As discussed in the previous section, the Ba atoms removed by the IB are resupplied very fast, but can only be present on the surface bound
to O atoms. Therefore, the Ba/O ratio is approximately equal to unity during the IB, independent of the ion flux or cathode temperature.

The electron emission during the IB, as determined in the diode configuration, is also shown in Figs. 9 and 10 (open symbols). It is observed that the decrease in electron emission is identical to the decrease in coverage. This "one-to-one" relation between emission and coverage indicates that each Ba–O site can be regarded as an individual emission site, and that the total emission equals the sum of the local emission sites. Moreover, it confirms that the fraction of the surface covered during the IB has the correct Ba–O arrangement required for optimum emission. Quantitative analysis of different top-layer dispenser cathodes has shown that the Ba–O dipole density for the cathodes is of the order of \(3 \times 10^{14}\) atoms cm\(^{-2}\). At these densities, the Ba–O dipoles are so far apart that the interaction between the dipoles is negligible. This is in agreement with this one-to-one relation where each dipole is an individual emission site, and does not interact with the neighboring sites. Note that the one-to-one relation between emission and coverage has been assumed in previous investigation of the emission degradation of dispenser cathodes under IB, but without actual experimental verification.

A model that describes the surface coverage and electron emission during the IB can be derived using the insight gained from the separate investigations of the removal and resupply processes. Combining Eqs. (1) and (4) for the removal and the recovery rate, respectively, the dependence of the equilibrium coverage \(n_{eq}\) on the time constants for removal \(\tau_{IB}\) and resupply \(\tau_{res}\) is given by

\[
\frac{n_{eq}}{n_0} = \frac{1}{1 + \frac{\tau_{res}}{\tau_{IB}}}.
\]

(6)

Since the one-to-one relation between coverage and emission has just been proven, this Eq. (6) can also be used to describe the emission during the dynamic equilibrium. The O coverage determines also the Ba coverage, and thus the time constants for O removal and resupply must be used in Eq. 6. In order to verify that the O resupply rates should also be applied for the modeling of the emission, we have determined the time constants for the emission recovery after the ion bombardment has been stopped. These time constants are compared to the O resupply rates derived from the reactivation studies in Fig. 8 (filled circles). The good agreement between the time constants confirms the O resupply determines the emission recovery if sufficient Ba is generated.

The time constants for the O removal in the model cannot be deduced straightforward using the derived sputter yield for O in Sec. B. In the removal process, the Ba and O atoms are removed in a step-by-step manner, which means that O can only be sputtered if the Ba atoms are not present on top. The Ba atoms thus form a protective layer over the O atoms. In order to remove an O atom, collision chains induced by the incident ions have to hit the surface at the same O position twice within the time it takes for the Ba removed in the initial event to be resupplied on top of the O atom. The probability for the same position to be hit twice is proportional to the incident ion flux, and the probability for the Ba to be resupplied between those two events depends on the temperature. The probability to remove O from the surface thus depends on both the ion flux and temperature. In Fig. 11, the effective sputter yield for the O removal is shown versus the ion flux for two different cathode temperatures. The effective sputter yield has been determined from the slope of the initial decrease of the electron emission immediately after the start of the IB, when the resupply of O is negligible since \(n_{eq} = n_0\) (see Fig. 2). Here, we made use of the one-to-one relation between emission and coverage. In Fig. 2, it is observed that at low ion flux the effective sputter yield for O is much smaller than the sputter yield determined at room temperature \((Y = 0.62)\) due to the protective function of the Ba atoms. However, as the flux increases the finite Ba resupply rate is not sufficiently high to protect the O layer, and the effective sputter yield increases to the value of approximately \(Y = 0.6–0.7\). When the cathode temperature increases, the protective function of the Ba layer can be maintained up to a higher ion flux due to the faster Ba resupply.
These effective sputter yields for O derived from Fig. 11 are used in combination with the O resupply rates in Eq. (6) to model the behavior of the dispenser cathode under IB. The results of the model are shown in Figs. 9 and 10 by the solid curves. An excellent agreement between the model and the experimentally determined coverage and emission during IB is observed. We emphasize that no additional fitting has been performed. Thus, we conclude that the behavior of the cathode surface during IB can be accurately described by the derived model including the O removal and resupply rates.

E. Ion bombardment induced substrate damage

In the previous section, we discussed the protective function of the Ba atoms against the removal of O atoms by the IB. In a similar manner, the Ba–O complex protects the coating of the top-layer cathodes against the removal of substrate atoms by the collision chains. However, unlike the O atoms, loss of substrate atoms cannot be resupplied and thus leads to unrecoverable long-term emission degradation for top-layer cathodes. The fraction of the surface not covered by O is also not covered by Ba, and if the collision chains reach the surface at such positions, substrate atoms can be sputtered from the surface. The protection efficiency of the adlayer thus depends on the O coverage, and is therefore indirectly related to the incident ion flux and the cathode temperature.

Here, we investigate the damage to the substrate during IB for Os/Ru cathodes with a specially prepared thin Os/Ru layer of approximately 100 nm. To determine the protection efficiency of the adlayer, the cathodes are exposed to a certain fixed ion dose, which is delivered to the cathode at a varying ion flux during operation at 1030 °C. After the applied ion dose, a depth profile of the substrate is made using 3 keV Ar⁺ ions with the cathode at room temperature. In order to study the damage to the substrate inflicted by the ions, and determine the protection efficiency of the Ba–O layer, the depth profiles of the cathodes exposed to the IB are then compared to depth profiles of reference cathodes not exposed to any IB. Note that the time over which the IB dose is applied depends on the ion flux. To correct for any possible change in the substrate due to the (varying) extended time at operating temperature, the reference cathodes were operated simultaneously with the bombardment experiments.

The approach is demonstrated in Fig. 12, where the depth profiles of the reference substrate Fig. 12(a) and the damaged substrate Fig. 12(b) are presented for a dose of $d = 900 \times 10^{15}$ ions cm$^{-2}$ at a flux of $F = 3 \times 10^{12}$ ions cm$^{-2}$ s$^{-1}$. Both profiles show the surface composition versus the applied ion dose, where the composition is determined by the ion scattering analysis. It is evident from the reference depth profile in Fig. 12(a) that as the Os/Ru top layer is removed, the underlying W matrix is exposed. The fixed ion dose applied in these experiments at a varying flux is indicated in Fig. 12(a) by the dashed vertical line at $d_A = 900 \times 10^{15}$ ions cm$^{-2}$. The initial increase in the Os/Ru composition at the start of the depth profiles in [Figs. 12(a) and 12(b)] is due to the removal of WO$_3$ present on the substrate. The WO$_3$ segregates to the surface during cathode operation and covers approximately 10–15% of the substrate, as is discussed elsewhere. Figure 12(b) shows the depth profile of the damaged substrate, where the bottom axis shows the ion dose used for the depth profile of the remaining damaged substrate, while the top axis shows the total ion dose including the IB at operating temperature. The dashed curves represent the reference profiles from Fig. 12(a), shifted along the dose axis to fit the damaged depth profile. The vertical dashed line indicates the required shift of $d_P = 710 \times 10^{15}$ ions cm$^{-2}$, which represents the ion dose that did not affect the substrate due to the protective function of the adlayer. In case of perfect protection, the substrate is unaffected by the IB during operation and the depth profile would equal the reference profile shifted over the applied dose ($d_P = d_A$). If no protection takes place, the remaining depth profile after the applied dose $d_A$ would be equal to the profile to the right of the dashed line in Fig. 12(a) ($d_P = 0$). The extend to which the substrate is protected from the IB can thus be derived from the shift $d_P$ required to fit the reference profile to the damaged profile. Therefore, we introduce the dynamic protection factor $P$ as

$$P = \frac{d_P}{d_A}. \quad (7)$$

(Note that in previous IB studies the term dynamic shielding factor is used, instead of the term dynamic protection factor used here. However, this might suggest that the substrate is shielded from the incident ions by the Ba–O complex, which is not the case.) The thus determined dynamic protection factor is shown versus the ion flux at which the dose $d_A$...
is applied in Fig. 13 (open circles). The coverage of the Ba–O complex during the IB at the corresponding ion flux is also shown (filled circles). In addition, Fig. 13 shows the protection efficiency \( P \) of the remaining adlayer (dashed curve), which is derived by dividing the dynamic protection factor \( P \) by the remaining Ba–O coverage. It is observed that even for complete coverage the protection is not perfect since the substrate is damaged at a very low ion flux (\( P = 0.8 \)). This imperfect protection at low ion dose has previously been reported by Gärtner et al. in investigations of the influence of IB on the emission of Scandate cathodes with a Re/Sc2O3 coating. The protection efficiency of the adlayer is smaller than unity because in a fraction of the collision chains the outermost substrate atom is not able to transfer its energy to the Ba–O complex on top, and thus is sputtered from the surface. Although the Ba/O ratio is equal to unity and every O is protected by a Ba atom, this is not the case for the substrate atoms. Even for complete coverage of the substrate by the Ba–O complex there is not a Ba–O complex on top of every substrate atom. Therefore, the protective function of the remaining Ba–O is not perfect. Furthermore, the protection efficiency decreases with increasing ion flux because a large flux increases the surface roughness. For rough surfaces the sputter yield increases, and so does the probability to sputter second layer atoms. Consequently, the protection efficiency decreases with increasing ion flux.

In first order approximation, the protection efficiency of the adlayer can be taken to be \( P = 0.7 \)–0.8. The dynamic protection factor of the adlayer is then proportional to the surface coverage according to \( P = \epsilon_p \times \theta \). This implies that if one determines the surface coverage or electron emission during the IB, one can estimate the protection factor of the Ba–O layer for any given flux at any given temperature. This provides a fast method to estimate the protection during accelerated lifetime tests of dispenser cathodes, where the lifetime ion dose that the cathodes receive in cathode-ray tubes is given at a higher flux than in the actual tubes in order to have acceptable timeframes for the investigations. Due to the higher ion flux, the coverage and the resulting dynamic protection factor are smaller than in the case in the real tubes at lower flux. When investigating the influence of the IB on the cathode substrate, the difference in the protection factor must be corrected for. This correction can be performed if one knows the surface coverages (or electron emission) for the fluxes used in the tube and in the accelerated lifetime test: the protection factor scales with the surface coverage. Note that in the cathode-ray tubes the ion dose is not uniformly distributed across the surface, but is peaked in the center of the cathode.

**F. Ion bombardment in oxygen background**

It has been shown that the surface coverage during ion bombardment is determined by the removal and resupply rate of O atoms. The limited rate of Ba resupply when it is governed by the O resupply, compared to the potential resupply rate of Ba when O is already present has been demonstrated in Fig. 6. The decrease in Ba–O coverage and damage to the substrate due to the IB can be diminished by increasing the O resupply rate. To substantiate this conclusion, we performed an IB experiment in an O2 background pressure in order to enhance the O resupply rate. Figure 14 shows the Ba coverage during IB in an UHV (closed circles) and in the O2 background (open circles) using a flux of \( F = 2 \times 10^{13} \) ions cm\(^{-2}\)s\(^{-1}\) at a cathode temperature of 920 °C. The partial oxygen pressures were 1 \( \times 10^{-12} \) mbar in an UHV, and 5 \( \times 10^{-9} \) mbar in the O2 background, respectively.

The solid curves for the IB and resupply with the above determined time constants for removal \( \tau_{IB} \) and resupply \( \tau_{res} \) in an UHV. The solid curve for the UHV bombardment shows the coverage as a function of time, deduced by combining Eqs. (1) and (4) with the above determined time constants for removal \( \tau_{IB} \) and resupply \( \tau_{res} \) in an UHV. The solid curve for the bombardment in an O2 background was deduced in a similar manner, but using the time constant for resupply \( \tau_{res}(O_2) \) as a fitting parameter. (The solid curve for the O2 background has been slightly

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**FIG. 13.** The protection factor (open circles) and protection efficiency (dashed curve) as a function of the ion flux, derived from the IB simulations. The Ba–O coverages at the dynamic equilibrium are also shown for comparison (filled circles).

**FIG. 14.** IB in an UHV (filled circles) and an O2 background (open circles) using a flux of \( F = 2 \times 10^{13} \) ions cm\(^{-2}\)s\(^{-1}\) at a cathode temperature of 920 °C. The partial oxygen pressures were 1 \( \times 10^{-12} \) mbar in an UHV and 5 \( \times 10^{-9} \) mbar in the O2 background, respectively. The solid curves represent the results of the derived model, and the dashed line represents the situation for an infinite fast O resupply.
shifted to the right for clarity.) The thus derived time constant for resupply in O₂ background gives \( \tau_{\text{res}}(\text{UHV})/\tau_{\text{res}}(\text{O}_2) = 2.6 \).

We can estimate an upper limit of the improvement due to the increased O resupply from the results in Fig. 6, where it has been observed that the resupply of Ba on the oxidized substrate is enhanced by approximately a factor of 4 compared to the Ba resupply on the clean substrate. Therefore, for an infinitely fast O resupply, the time constant for resupply of the Ba–O complex is determined by the Ba resupply rate and can be decreased by a factor of 4 compared to O limited Ba–O resupply rate on a clean substrate (\( \tau_{\text{res}} = \tau_{\text{res}}(\text{UHV})/4 \)). In this case, the Ba coverage in Fig. 14 would equal \( n_{\text{Ba}} = 77\% \) as indicated by the dashed line, which represents the maximum Ba coverage possible at this flux and temperature, and is limited by the Ba resupply rate. We can thus conclude that enhancement of the O resupply rate improves the performance of the cathode during IB. Moreover, the increased coverage diminishes the damage to the substrate and thus increases cathode lifetime. The protection factor \( P \) for the substrate increases by approximately the same fraction as the surface coverage is increased. An increase in cathode performance has also been observed by Gärtner during O₂ poisoning investigations. However, we emphasize that above a certain O₂ pressure the poisoning by Ba–O layer is very important for coated cathodes, where damage to the substrate leads to unrecoverable emission degradation. The importance of IB resistivity is most evident for the scandate cathode which has a coating containing a Ba–Scandate complex. Although the scandate cathode has an enormous emission potential of up to 400 A cm⁻², its breakthrough for application has not been possible so far due to its strong susceptibility to ion bombardment damage and emission degradation. The scandate cathode does not have a Sc resupply mechanism, so when the Ba–Scandate complex is sputtered from the surface the emission enhancement by this complex is no longer present. Based on the evidence of O resupply presented here, a possible approach to increase the resistivity to IB is to increase the O resupply rate so the Ba–Scandate complex is better protected from the IB. Hasker et al. have indicated that the Sc₂O₃ complex regulated the O concentration on the surface. Definite conclusions about the role of the complex in the O resupply requires similar experiments as performed here for Os/Ru cathodes on Scandate cathodes.

**IV. CONCLUSIONS**

In this article, we have studied and modeled the surface coverage and electron emission of dispenser cathodes during IB. All the results presented here clearly demonstrate the crucial role of O in the surface coverage of the dispenser cathodes. Many cathode studies reported in the literature have mainly focussed on the Ba coverage, and have underestimated the importance of O. However, Ba can only cover the cathode substrates if it is bound to O present on the surface. During cathode operation, with or without IB, the Ba/O ratio equals unity. The surface coverage during the IB is thus determined by the O removal and resupply rates, as long as an ample supply of the Ba is available in the pores.

The Ba atoms in the outermost layer protect the O atoms situated below from the IB. In order to remove an O atom, collision chains initiated by the incident ions must reach the surface at the same position twice within the time required to replace the Ba atom sputtered by the first collision chain. The probability to remove the O atoms is thus determined by the ion flux, which determines the probability that a collision chain reaches the surface twice at the same position, and the cathode temperature, which determines how fast the lost Ba is replaced.

Any O removed from the surface must be resupplied by diffusion from the pores. The O resupply is not in the form of BaO as has been suggested in many studies, but by individual O atoms. Although the surface diffusion of O itself is fast, the O resupply rate is limited by the formation of free O in the pores. The resistivity of the cathodes against IB is thus determined by the O resupply rate. Indeed, cathodes with a fine-grained matrix show improved IB resistivity due to smaller diffusion distances. The decisive proof of the importance of the O resupply was given by performing an IB experiment in an O₂ background pressure. Due to the externally enhanced O resupply, the IB resistivity and the Ba–O coverage during the bombardment increased significantly.

The O resupply is not only important for maintenance of the Ba–O layer, it also indirectly influences the IB-induced damaged to the cathode substrate. The Ba–O layer protects the substrate from the IB, where the protection factor is in a first order approximation proportional to the surface coverage during the ion bombardment. The protection of the substrate by the Ba–O layer is very important for coated cathodes, where damage to the substrate leads to unrecoverable emission degradation. The importance of IB resistivity is most evident for the scandate cathode which has a coating containing a Ba–Scandate complex. Although the scandate cathode has an enormous emission potential of up to 400 A cm⁻², its breakthrough for application has not been possible so far due to its strong susceptibility to ion bombardment damage and emission degradation. The scandate cathode does not have a Sc resupply mechanism, so when the Ba–Scandate complex is sputtered from the surface the emission enhancement by this complex is no longer present. Based on the evidence of O resupply presented here, a possible approach to increase the resistivity to IB is to increase the O resupply rate so the Ba–Scandate complex is better protected from the IB. Hasker et al. have indicated that the Sc₂O₃ complex regulated the O concentration on the surface. Definite conclusions about the role of the complex in the O resupply requires similar experiments as performed here for Os/Ru cathodes on Scandate cathodes.

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