PREFLASHOVER SPECTROMETRY OF A VACUUM DIODE

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ABSTRACT
A bakeable, ultrahigh-vacuum, time-of-flight (TOF) mass spectrometer has been constructed in order to directly investigate the nature of the currents which flow prior to the flashover of a vacuum diode. The identity of the preflashover anode species is obtained by the time correlation between their arrival at a detector and the high voltage pulse applied to the cathode. By varying the ratio of the pulse and a dc bias applied to the anode, the extent of the ion production region in front of the anode can be determined. Observations at 100 K indicate the presence of multiply charged metal anode ions, as well as ions of adsorbed residual gas. The type of observed species indicates that electron-induced desorption is a primary preflashover production mechanism, although field desorption processes may also be of significance. No detected species were formed in the gap, in contrast to what was expected from the previous results of Davies and Blom. It is suggested that the absence of such ions may be related to the value of the macroscopic field employed since preliminary observations at flashover indicate a noticeable change in type and abundance of observed species.

I. INTRODUCTION
A detailed understanding of the nature of preflashover currents in a vacuum diode is vital to the understanding of the breakdown process itself. Although much information has been obtained from measurements of current and voltage, little detailed information is available on the nature of the species actually present in the gap prior to flashover. Recent observations have indicated the presence and distribution of neutral anode vapor, but the presence of anode ion species prior to flashover has not been reported. The positive identification of these species is particularly important in determining which of the most probable mechanisms responsible for their formation (electron-induced desorption or field-induced desorption at the anode surface) is dominant. Granting the existence of cathode microprotrusions which act as electron field emitters, one might also expect anode protrusions with local fields sufficiently large to cause field desorption of residual gas or thermally enhanced field evaporation of the emitter, resulting in the formation of characteristic, highly charged, metal anode ions.

II. THEORY
During operation, a bias potential $V_B$ is applied to the anode of the vacuum diode. When a high voltage pulse - $V_p$ is applied to the cathode, ions are formed at the anode surface or in the gap of the diode, are accelerated, and pass into a drift region at ground potential through a 0.5-mm hole in the cathode. An oscilloscope at the entrance to the drift region focuses the ions on an electron multiplier one meter away. The time interval between the resulting signal, amplified and displayed on an oscilloscope, and the beginning of its sweep triggered by the pulse applied to the cathode, is a direct measure of the ion’s travel time. If the duration of the cathode pulse is long compared to the ion’s travel time in the acceleration region and if the ion’s initial kinetic energy is zero, its final kinetic energy will be just:

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\[ \frac{1}{2} m v_f^2 = n e V_B \left[ 1 - \frac{d}{D} \left( 1 + \frac{V_f}{V_B} \right) \right] \quad 0 \leq \frac{d}{D} \left( 1 + \frac{V_f}{V_B} \right) \leq 1.0 \quad (1) \]

where \( m \) is the ion's mass, \( v_f \) its velocity at the entrance to the drift region, \( n \) its charge, \( d \) the distance from its formation position to the anode, \( D \) the gap spacing, and \( V_f \) the magnitude of the applied negative pulse voltage. Since the ion's travel time in the gap and lens is short compared to its travel time in the drift region, its total travel time \( T \) is, to a very good approximation, just:

\[ T = L/v_f \quad , \quad (2) \]

where \( L \) is the length of the drift region.

The ion is identified by its mass-to-charge ratio expressed in terms of the applied volts, the measured travel time, the position of formation, and the known drift distance. Combining Eqs. (1) and (2) gives the desired result:

\[ \frac{m}{n} = \frac{2 e^{1/2} v_f}{L} V_B \left[ 1 - \frac{d}{D} \left( 1 + \frac{V_f}{V_B} \right) \right]^{1/2} \quad , \quad (3) \]

where distances are measured in meters, volts in kilovolts, time in microseconds, and \( m/n \) in atomic mass units.

Now consider two identical species: one formed at the anode, the other in the gap. For the ion formed at the anode \( d = 0 \) in Eq. (3). Combining the resulting expression for \( (m/n)_{\text{anode}} \) with Eq. (3) and rearranging gives:

\[ (m/n)_{\text{gap}} = (m/n)_{\text{anode}} \left[ 1 - \frac{d}{D} \left( 1 + \frac{V_f}{V_B} \right) \right]^{-1} \quad , \quad (4) \]

Equation (4) indicates that the mass-to-charge ratio of an ion produced in the gap will appear greater than that of the same ion produced at the anode, and will depend on its formation position \( d \). Effectively, an ion produced in the gap will acquire less kinetic energy than its counterpart produced at the anode. It will, therefore, travel a longer time in the drift region, appearing as a slower ion, or one with a greater \( m/n \).

The important point is that the reproducibility of the observed mass-to-charge ratio of a given species is directly proportional to the reproducibility of its formation position in the gap. Furthermore, for a species to be detected, we have the condition from Eq. (3):

\[ 0 \leq \frac{d}{D} \left( 1 + \frac{V_f}{V_B} \right) < 1.0 \quad , \quad (5) \]

By varying the ratio of the pulse-to-bias voltage the extent of the ionization region \( d \) in the gap can be probed. For example, with \( V_f/V_B = 1.0 \), ions can be detected from the anode surface (\( d = 0 \)) to a gap position \( d = D/2 \). For \( V_f/V_B = 2.0 \), ions can be detected from the anode to a gap position \( d = D/3 \). If ions were only produced in significant quantity between \( d = D/3 \) and \( d = D/2 \), the observed ion currents would peak for \( 1.0 < V_f/V_B < 2.0 \).

Using a calibration procedure described previously, one can determine the mass-to-charge ratio of a given species to better than \( \pm 1 \) amu at \( m/n = 50 \) or, in special cases, to better than \( \pm 0.1 \) amu for \( m/n \leq 3 \).
III. APPARATUS

Figure 1 is a schematic diagram of the bakeable, ultrahigh-vacuum, time-of-flight mass spectrometer. Trapped oil diffusion and electron ion pumps allow typical operating pressures of less than $10^{-7}$ Torr after bakeout at 200°C. The spectrometer source is a parallel plate vacuum diode composed of an aluminum alloy anode and type 304 stainless steel cathode of $5 \times 5 \text{cm}^2$ in area and separated by 0.5 mm. The anode, located at the end of a glass cold finger, can be cooled to cryogenic temperatures, encouraging preferential adsorption on its surface when known gases are introduced into the vacuum system.

The high voltage pulse is supplied by a pulse generator of high output impedance, eliminating the need for impedance matching at the cathode. The pulse amplitude is adjustable from 0 to 30 kV with a rise time of 10 to 30 nsec and a width of 100 nsec. A small, in-vacuum antenna detects the arrival of the pulse at the cathode and provides a zero reference for time measurements, as well as a signal to initiate the time-measuring sequence.

The bias voltage which determines the ion kinetic energy is set by a precision high voltage power supply with a ripple of less than 250 mV and a regulation of better than 0.0025% at 20 kV.

Ions are detected with a focused mesh electron multiplier operated at a gain of $10^9$. A grounded grid placed 2 cm in front of its first dynode defines the extent of the drift region and ensures that the ions' travel time in the acceleration region in front of the multiplier is negligible.

IV. OBSERVATIONS AND CONCLUSIONS

In these experiments, the sum of the anode bias voltage and the pulse applied to the cathode was held constant with the resulting field equal to $5 \times 10^3 \text{V/cm}$, approximately 95% below that required for visible flashover. Figure 2 is a photograph of several typical oscilloscope time traces. The identical travel times in successive sweeps indicate a unique ion species. The accuracy in determining the single ion travel time, $\pm 30$ nsec, corresponds to an uncertainty in formation position for a given species of $\Delta R / R = \pm 3\%$. For the maximum ratio of the pulse-to-bias voltage used and the known gap spacing, the minimum extent of the ionization region can be determined from Eq. (4) to be $R = 2L$, corresponding to an uncertainty in ion formation position of $\Delta R = \pm 0.25 L$.

As the pulse amplitude is changed, the observed travel times remain constant, changing only with a change in the applied bias voltage $V_0$. This indicates that the species are being formed at the anode. In Figure 3a, the corresponding mass-to-charge ratios (calculated from Eq. (1) with $d = 0$) are displayed in histogram form. Figure 3b is the corresponding residual gas spectra, taken in the same system and plotted as a histogram, normalized to the $E_0$ peak of Fig. 3a. The excellent agreement in m/e values between the two histograms for known residual gas species indicates that the equation used to determine the species in 3a is valid; that is, the assumption of ion formation at the anode is correct. Furthermore, the comparison shows that residual gas ions from the anode as well as metal anode ions comprise these preflashover currents. From the known multiplier and preamplifier gains, the equivalent microscopic currents can be calculated to be between $10^{-12}$ and $10^{-14}$ amp.
A close examination of the individual data points shows that the species between \( m/n = 13 \) and \( m/n = 14 \) in Fig. 3a actually peak at \( m/n = 13.5 \) corresponding to \( \text{Al}^{2+} \). In this instance, a histogram display, although convenient, can be misleading since details within an individual histogram box are lost.

The peaks corresponding to copper and magnesium species reflect the use of an alloy anode\(^1\) in which these materials are primary constituents. The absence of a peak at \( m/n = 20 \) in Fig. 3a and the large peaks at \( m/n = 12 \) and \( m/n = 16 \) indicate that \( \text{CO} \) is dissociating at the anode surface during the preflashover process. A detailed study of the individual peaks below \( m/n > 1.0 \) shows that dissociation of \( \text{H}^2 \) and \( \text{H}_2^+ \) is also occurring near or on the anode surface. The presence of dissociation processes is not surprising since the macroscopic field in the gap is sufficient, even without electron bombardment of the anode, to cause field dissociation of these species.\(^8\)

Figure 3a plots the data obtained for all ratios of pulse-to-hiss voltage used, between approximately 1.0 and 12.0, for constant field strength in the gap. These correspond to a variation in the extent of the ionization region from \( d = D/12 \) to \( d = D/20 \).

It is interesting to note that with the possible exception of the hydrogen and anode metal ions, the observed species are not seen in the classic shot-probe field ion emitter experiments of Müller and co-workers.\(^1\) This strongly suggests that electron-induced desorption is a major mechanism for the production of preflashover ion species, although the presence of the hydrogen and anode metal ions may indicate that field desorption processes are also occurring.

In view of the previous results of Davies and Biondi, it is perhaps surprising that so measurable a number of ions was observed as originating in the gap itself. Their observation of neutral electrode material in the gap microanemones before flashover would suggest that corresponding ion species should be observed, formed by electron bombardment from the cathode. Were it observed in this experiment a definite increase of several orders of magnitude in the abundance of electrode species as the field in the gap is increased to the value at which visible flashover occurs. Preliminary observations of these species indicate that their initial kinetic energy and energy distribution is appreciable and that fast neutral or metastable as well as ions are present. This, in turn, suggests that field-dependent processes are occurring and that the production of ions in the gap may be a strong function of the applied field.

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REFERENCES


\(^1\)Alloy 2004 consisting T351. Primary constituents: Al (93.7-99.4%), Cu (3.8-4.9%), Mg (1.2-1.9%), Mn (0.5-0.9%), all others less than 1%.
6. Power Designs, Inc., Palo Alto, California, Model 15/6R.
8. Actually, identical travel times are also possible for two different ions formed in different locations in the gap such that:

\[
\frac{m}{n}_1 \left[ 1 - \frac{d_1}{D} \left( 1 + \frac{V_e}{V_B} \right) \right] = \frac{m}{n}_2 \left[ 1 - \frac{d_2}{D} \left( 1 + \frac{V_e}{V_B} \right) \right]
\]

However, repeated identical travel times would imply two unique formation positions in the gap for two unique species, which is highly improbable.