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Authors
Heard, Harry G.
Lauer, Eugene J.

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ABSTRACT

Cu^{64} was used as a radioactive tracer to confirm the transfer of anode metal to the cathode in a "non-sparking" d-c discharge in high vacuum. In one run with a 14 mm gap and 100 kv (gradient $\cong 7 \times 10^4$ V/cm) the ratio of transferred copper atoms to electrons was 1:600 for a total drain of 5×10^{-7} amperes. In a different run at 3.5 mm and 50 kv (gradient $\cong 1.4 \times 10^5$ V/cm) the ratio became 1:2 for a total drain of 5×10^{-8} amperes. The relatively large amount of copper transferred suggests that most of the metal crosses the gap uncharged.

The current between electrodes was found to be composed of a steady and a randomly fluctuating component. The fluctuating component was found to be independent of pressures but dependent on the total gap voltage and electrode spacing. The fluctuations would, for a set of fixed conditions, usually decrease with time. Tests showed that the fluctuating component of current could not be used to foretell breakdown.

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INTRODUCTION

One of the mechanisms proposed[1,2] to explain high-voltage breakdown between electrodes in high vacuum* requires the transfer of ions from anode to cathode. The work of Trump and Van de Graaff[1] and later of Filosofo and Rostagni[3] has shown that a definite positive ion current flows from the anode to cathode in the presence of a high voltage. There exists the possibility that their experiments measured the ionization of the surface film of gas on the anode rather than the actual transfer of anode metal. The work described below, while not eliminating the above mechanism as a part of the total breakdown process, definitely establishes that the anode metal appears at the cathode in the non-sparking discharge.

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* "High vacuum" will be used to signify conditions wherein the electrode separation is much much less than the mean free path.

1. J. G. Trump and R. J. Van de Graaff, Journ. Appl. Phys., v. 18, p. 377, (1947).

2. H. W. Anderson, Trans. A.I.E.E., v. 54, p. 1315, (1935).

3. I. Filosofo and A. Rostagni, Phys. Rev., v. 75, p. 1269, (1949)

A radioactive anode was placed opposite a normal cathode in a high-vacuum cavity. When voltages of the order of 50 to 100 kilovolts were applied between the electrodes, active anode metal was found at the cathode, even though no spark passed between the electrodes. From the counting rates of the cathode and anode one can compute the weight of metal transferred. The amount of metal transferred indicates that most of the anode metal crosses the gap either uncharged or perhaps as small aggregates of atoms, a few of which are ionized.

Both the gap current and the gap voltage were monitored during a discharge to determine if their fluctuations could be associated with the onset of breakdown.

DESCRIPTION OF APPARATUS

Vacuum System

The all-metal vacuum system used in these experiments was continuously evacuated with a special two-stage mercury pump.* Interposed between the test cavity and the metal mercury diffusion pump was a multiple bounce-type liquid nitrogen trap having an effective pumping speed for condensables of approximately 1000 l/sec. The intake of the mercury pump was covered by a simple water-cooled baffle designed to reduce the load on the liquid nitrogen trap. An additional cold trap was placed between the mercury pump and the forevac pump, to prevent oil vapors from the mechanical pump from reaching the test cavity.

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* The all-metal 85 l/sec mercury pump was designed by Warren Chupp.

Cleaning of Vacuum System and Electrodes

Previous to this run the entire vacuum casing and traps were sand blasted (inside and outside) and washed in flowing C.P. acetone and C.P. ethyl alcohol (95 percent). All gaskets in the high-vacuum section of the unit were made of commercial 40-60 solder the ends of which had been fused together. Insulators were coupled to the system with lead-encased gum rubber gaskets.

Electrodes and holders were carefully washed with Dreft and water, distilled water, chromic acid made from 37 N C.P. sulfuric acid, and finally rinsed in distilled water, C.P. acetone and C.P. ethyl alcohol. Parts were assembled immediately with grease-free tools and paper towels. None of the parts were touched with bare hands during assembly.

A typical base pressure for the system was of the order of $1\text{-}3 \times 10^{-7}$ mm Hg on an untrapped Westinghouse 5966 ionization gauge. The lowest pressure recorded on a trapped gauge was 8×10^{-8} mm Hg.

Spark Monitoring Device

Because of the relatively large amount of metal transferred to the cathode during breakdown (see Run 3, Table I), the success of this experimental technique rests largely on the ability to maintain the electrodes in a non-sparking discharge. In an earlier experiment at this laboratory, visual monitoring of the gap was tried. The results, though similar in nature, were discounted because of the uncertainty involved. In this experiment the flow of charge was monitored

electronically. With the electronic equipment, a 1 volt pulse developed across a metering capacitor of 1 microfarad triggers the spark alarm. The alarm is thus sensitive to a 1 microcoulomb charge transfer. That the electronic alarm gives a rather conservative definition of sparking is borne out by one test (see Run 2, Table I) wherein no detectable active material was found on the cathode after a spark of short duration and sufficient amplitude to trigger the alarm.

X-Ray Gap Current Monitoring

The time dependence of x-ray emission produced during the non-sparking discharge was monitored with a stilbene crystal-photomultiplier combination (1P21) in conjunction with an amplifier, cascade scalers and an oscilloscope. The instrumentation described above is shown in Fig. 1.

Electrodes and Bombardment

Both electrodes were hemispherically-capped cylinders of electrolytic copper of 1 inch radius. The electrode gap was varied external to the high-vacuum system through a sylphon bellows. Because of thermal expansion of the anode, the gap spacing was monitored continuously with a cathotometer. The electrode spacing is known to $\pm$ 0.05 mm.

The anode was bombarded with 8 Mev deuterons in the 60-inch cyclotron. Tantalum absorbers were used to reduce the beam energy 19.5 to 8 $\pm$ 1 Mev. Most of the resulting activity is due to the reaction Cu^{63} (d,p) Cu^{64}. Cu^{64} was chosen for the radioactive run because of the relatively high yield, freedom (under optimum bombardment

conditions) from disturbing long lived secondary products, and availability. The above reaction[4] has a maximum cross section $\sigma = 0.25 \times 10^{-24}$ cm^2 at approximately 8 Mev and largely yields the 12.8 hour Cu^{64}.

The total bombardment of the anode was limited to a 1 cm^2 area on the nose. Reaction yields predict a 4.5 millicurie source of Cu^{64} for the 1.5 microampere-hour bombardment obtained.

Current and Charge Metering Circuits

A General Radio 715-A d-c amplifier was used in conjunction with an Esterline-Angus recorder to monitor the electrode drain. The amplifier was operated from a voltage stabilizer (Sola) and allowed several hours to reach thermal equilibrium prior to measurements. The relatively slow response of the recorder approximately 5 cps gave a continuous integrated record of the electrode drain.

A coulombmeter consisting of a d-c amplifier and watt-hour meter combination was used to register the total charge transfer. Drift was minimized by using a long warm-up period and stabilizing the 60-cycle supply voltage with a Sorensen S-1000 regulator. Previous experience with this coulombmeter had shown that it did not meter accurately the rapid fluctuations in anode current. To overcome this difficulty the input to the 715-A amplifier (whose output the coulombmeter records) was paralleled with a high quality (Plasticon) 1 microfarad condenser. This combination integrated the current so as to make the coulombmeter error negligible but did not impare the response of the drain recorder. A calibration run on d-c and square waves showed the error to be less than 5 percent.

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4. E. T. Clark and J. W. Irvine, Phys. Rev., v. 69, p. 680, (1946)

High Voltage Supply and Metering

The continuously variable high voltage supply used in these experiments was composed of two aiding 60 kv, 10 ma. supplies. A rather severe cascade RC- filter system reduced the power supply ripple to 0.3 percent at 110 kv.

In order to reduce the damage to the electrodes during spark breakdown, a 20 megohm resistor was connected in series with the high-voltage lead to the gaps. Power supply voltage was metered directly with a calibrated 1/2 percent meter and a calibrated 2 percent multiplier.

Pressure Fluctuations

So that the time dependence of electrode drain and system pressure could be compared, an Esterline-Angus recorder was connected to the Westinghouse 5966 ionization gauge by way of another General Radio 715-A d-c amplifier. The writing speed of the chart was synchronized with that of the drain recorder. The emission current of the 5966 was adjusted to 1.2 and 12 ma which, according to a recent calibration run by Warren Chupp, corresponds to 1 microampere of ion current at 10^{-4} and 10^{-5} mm Hg.

EXPERIMENTAL TECHNIQUE

In order to make the great difference in counting rates ($1 \text{ to } 10^9$) of the cathode and anode amenable to quantitative counting in the same geometry, an electroplating technique was developed. Preliminary tests showed that owing to the evolution of hydrogen, the weight of plated

metal was sensitive to voltage and current levels so that reproducible data could not be obtained. The anode material, dissolved from the electrode by acid etching, was plated onto 1 inch diameter platinum discs. By control of plating time and current, an approximation to the weight of plated copper was obtained. The actual weight of copper was determined by weighing the platinum electrode before and after plating. Electrodes were kept in a desiccator and handled with tweezers to prevent the accumulation of moisture from affecting the weight differential. The main source of error accrued from the necessary use of electroplating tape to localize the plating to the upper surface of the electrode. Despite these sources of error, the differential weight of ± 0.2 mg could be detected.

A limitation in this technique is that the amount of plated material must be sufficiently large to make errors in weighing small. Although it was intended that the anode and cathode were to be counted by plating onto identical 5 mil platinum discs, the counting rate of the cathodes turned out to be so low that the cathodes had to be counted directly. An uncertainty exists because of the geometry and scattering difference between the counting of the sample of the active anode and the direct counting of the cathode. This error will be discussed below.

DISCUSSION OF RESULTS

Electroplating and Counting

The amount of anode metal transferred to the cathode during the non-sparking discharge was determined from the simple relation

$$W = W_a D_c / D_a \qquad (1)$$

where the W's refer to the mass of metal and the D's refer to the decay rates of the active material on the cathode and anode. In this relation, it is understood that the electrodes are counted at the same time and in the same geometry. The results of runs with four cathodes for this active anode are given in Table I.

Referring to Eq. (1) the estimated errors in the three factors in the right member can be summarized as,

W_a =	0.62 ± 0.2 mg	±30 percent
D_c:	5 c/m above a background of 23 c/m. Counting for 10 minutes gives an uncertainty of	±15 percent
	Backscattering would be less for material on copper cathodes than for platinum electrodes.	-50 percent
	Variation of activity over the surface of the cathode.	Factor of 2
D_a:	Uniformity of activity within bombarded area	Factor of 2
	Extrapolation of activities to time zero. (Run 4)	-50 percent
	Electroplating metal from anode not in active region.	-Factor of 2
	Uniformity of activity within depth of anode active area used for plating.	negligible
	Extrapolation to same counting geometry	±50 percent

It is concluded that the estimate of copper transferred could be large by a factor of 8 or small by a factor of 4.

To assure that the activities were not caused by material crossing the gap by evaporation in the absence of voltage, an inactive electrode was placed opposite the active anode for approximately 24 hours. Even though the gap spacing was only 1 mm, no active material was found on this cathode.

The number of atoms transferred during the discharge can be written as

$$N_t = \frac{\beta}{\alpha} \frac{QL}{F} \qquad (2)$$

where: N_t = the number of atoms transferred to the cathode

α = ratio of charged to uncharged atoms transferred

β = fraction of the total charge carried by positives $\frac{Q^+}{Q}$

Q = coulombs of charge transferred during the discharge

L = 6.023×10^{23}, Avagadro's number

F = 96,506, Faraday's constant.

Equation (2) can be solved for β/α which can be determined from experimental data, assuming $\alpha = 1$. The variation of β from large to small gaps suggests that α is less than 1. This implies that anode metal crosses the gap uncharged.

Drain and Pressure Variation

During Run 1 the drain exhibited an unexplained voltage conditioning phenomenon. When the gap voltage (for constant spacing) was raised in increments of approximately 5 kv the drain would stay constant

for approximately 30 seconds and would then drop about 10 percent. The drop in drain, which was immediately followed by a rise of 2 to 5 times the former value, would execute an oscillatory decay to a stable but slightly higher value. The period of this oscillation and the time required to decay to a relatively stable value were found to be a function of the gap voltage. This oscillatory drain phenomenon was only exhibited on the first run although the instrumentation for all runs was identical.

A typical example of the relation of total pressure to electrode drain is that obtained in Run 3. The simultaneous recording of drain and pressure as a function of time showed the following relations as the voltage was raised in increments of approximately 5 kv:

1. Pressure and drain rise simultaneously with the increment of voltage. The decay of pressure to an equilibrium value takes longer than the decay to an equilibrium drain.

2. The maximum drain and pressure attained are a function of the rate of rise of voltage.

3. The absolute level of the equilibrium pressure does not appear to be a function of the applied voltage, however, the pressure does not exhibit fluctuations about the equilibrium value which disappear when the electrode voltage is removed.

4. With the drain at an equilibrium level, the introduction of a leak of air [sufficient to raise the pressure to the same value induced by a positive increment of voltage] causes the drain to drop immediately. If the leak is now shut off, allowing the pressure to return to normal, the drain rises to the original equilibrium value.

5. With the electrodes at 100 kv and spaced at 12 mm, the drain was observed to decrease as the electrodes were moved from 12 mm to approximately 4 mm. The total pressure showed a positive correlation (decreased). These pressure and drain variations were reproducible. When the electrodes were moved closer than about 4 mm, the drain and pressure increased again.

The above observations are in accord with a hypothesis with postulates the existence of a gas film on the surface and in the volume of metal of the electrode. The electrodes used in these tests were not outgassed.

X-Ray Monitored Gap Current

As mentioned above, the gap current was monitored by photocounting techniques. From oscillographic and counting data it was found that the average d-c drain contains small pulses sharply

separated in time and occurring randomly. Comparing pulse shapes with those obtained from a source of known beta energy, the bursts of x-ray pulses were seen to have the usual built-up form.

An attempt was made to associate the time dependence of these current pulses with the onset of gap breakdown. No specific trend was found except that the number of pulses that appeared per unit time was decreased by a gap breakdown.

Comparison of the total gap drain with the number of pulses per unit time showed that these pulses are only a small part of the integrated gap drain. The number of pulses appearing per unit time was found to increase with voltage for a given spacing and to increase with a decrease in spacing for a constant gap voltage.

CONCLUSIONS

1. The large amount of anode metal transferred across the gap in the non-sparking discharge and the non-uniform distribution of transferred activity on the surface of the cathode suggests that the anode material is removed by an evaporation process. The variation of β/d with the discharge conditions suggests that in smaller gaps most of the anode material crosses the gap uncharged.

2. The results of positive ion-electron secondary emission experiments in which only charge transfer

was measured can no longer be relied upon.

3. Comparison of Runs 1 and 4 indicate that the spacing and gradient rather than the total gap voltage are the controlling factor in the amount of material transferred. Because the specific activity of the anode was low and the half-life short, these results could not be checked for reproducibility.

4. The relation of gap current fluctuation with time is not understood. Gap current fluctuation has not been found to be associated with the approach of breakdown.

RECOMMENDATIONS

If this technique is used in the future to investigate sparking and drain phenomena, the counting rate of the cathode could be increased by electroplating a layer of high specific activity on the nose of the anode. A theoretical increase in counting rate by a factor of 10^8 with a fixed total source strength is available by this technique. With higher counting rates available, the time required for a given run would be greatly reduced and the individual runs could be checked for reproducibility. It might very well turn out that this approach could single out the determining factor for gap breakdown. Because of the health hazard involved, the proposed experiment would have to be handled with great care.

Experimental measurement of the positive ion-electron multiplying coefficient with a radioactive anode would yield directly the relation between the mass of transferred and number of electrons as well as the state of ionization of the transferred metal.

Acknowledgements

We are indebted to Dr. M. G. White, who was associated with us in the early phases of this work. His advice and encouragement are greatly appreciated.

TABLE I

SUMMARY OF COUNTIN. DATA

RUN	GAP SPACING mm	GAP VOLTAGE KV	GAP GRADIENT kv/cm	TOTAL CHARGE TRANSFER COULOMBS	SPARK (?)	COUNTING D_c c/m	RATES D_a c/m	METAL* TRANSFERRED gms	COULOMB EQUIVALENT OF METAL TRANSFER	$\beta = \frac{Q^+}{Q^-}$
1	14.2	100	70.4	7.56×10^{-3}	No	5	350,000	8.7×10^{-9}	1.3×10^{-5}	1:580
2	12.6	100	79.4	4.98×10^{-4}	Yes	0	-	less than 10^{-10}	-	-
3	1.4	50	357.0	1.60×10^{-5}	Yes	1513	115,000	8.1×10^{-6}	1.2×10^{-6}	1:13
4	3.5	50	143.0	1.69×10^{-4}	No	10	88,000	6.8×10^{-8}	1.0×10^{-4}	1:7

NOTES

* For 0.62 milligram anode sample

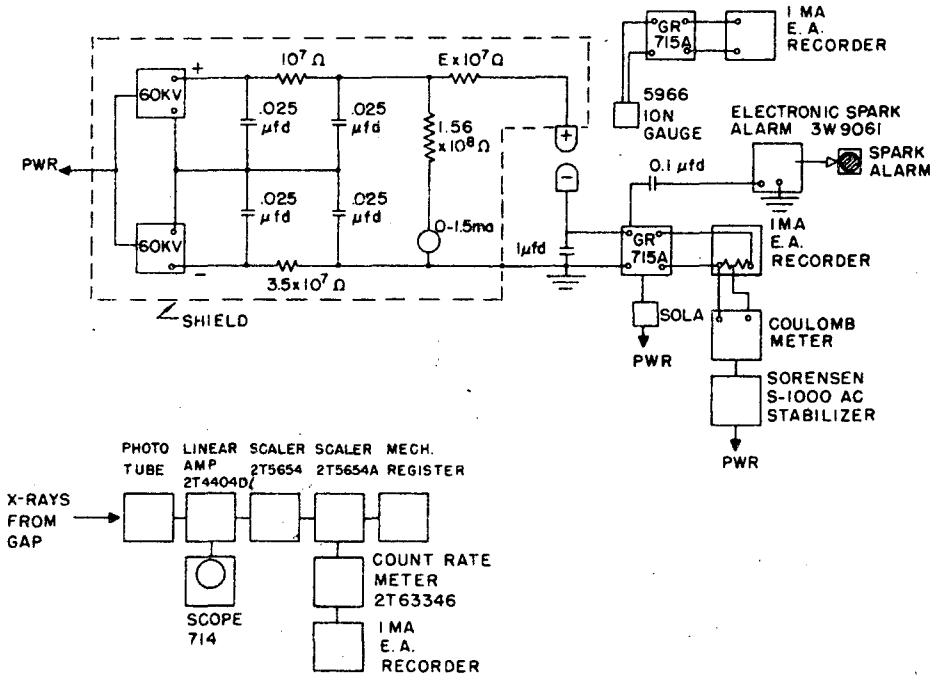

FIG. 1 INSTRUMENT LAYOUT & POWER SUPPLY

END OF DOCUMENT