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THE LOSS OF MATERIAL FROM THE CATHODE OF METAL ARCS

R. Seeliger and H. Wulfhekel

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16. Abstract

A study was made of the effect of arc length, cathode thickness, current strength, gas pressure and the chemical nature of the cathode material and filling gases upon the material loss from Cu, Fe, and Ag cathodes in arcs. The results show that the analysis of the phenomenon is complex and the energy balance is difficult to formulate.

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THE LOSS OF MATERIAL FROM THE CATHODE OF METAL ARCS

R. Seeliger and H. Wulfhekel

Material loss from the cathode [1] of a glow discharge, the so-called cathode evaporation, has been investigated in a large number of works. At present we can consider the situation essentially explained, thanks particularly to the later works of Blechschmidt, Guentherschulze and v. Hippel. It is, therefore, quite surprising that there are so few similar studies for the cathode of an arc discharge, except for those more technologically oriented about the so-called electrode burnout of carbon arcs. For metal arcs, aside from one old investigation by Guentherschulze at the cathode of a mercury rectifier, there has been, to our knowledge only one (detailed) work by v. Issendorff, also on the mercury cathode in a mercury arc, and a short orienting study by Seeliger and Schmick on iron cathodes [2]. An important finding of the latter was the demonstration that the material loss from the cathode depends, other things being equal, on the cooling of the cathode, so that it cannot directly be viewed as a specific constant which can be evaluated theoretically. This recognition is of great importance for all theoretical speculations on the processes depending on an energy balance at the arc cathode. But there is another reason for assigning fundamental importance to such investigations. This can be seen from the theories of cathode evaporation, discussed recently by v. Hippel, and the ideas sketched by Schmick and Seeliger, which have led to a comprehensive theory of the processes at arc and glow discharge cathodes. Therefore, it seemed useful to us to expand the experimental material by systematic experiments. In the course of the investigation, it appeared that without a really exhaustive study, many of the experimental parameters that played parts could only be suspected, and that we could work only with an orienting examination, even if it were with quantitative goals and on a considerably broader basis.

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Experimental Arrangement and Methods of Measurement

The problem itself is a very simple one. It deals with quantitative determination of the amount of material which the cathode of an arc discharge loses under specified experimental conditions. But solution of this problem encounters a series of substantial difficulties. Some of them are purely experimental, but part involve fundamental questions. First, it is not only the cathode of an ordinary arc which loses material, but the anode also; and we can hardly avoid the question of whether material is not transformed from the anode to the cathode, masking the actual material loss from the cathode. Now although the gross loss from the cathode may well be of interest in many respects, it would be simpler, clearer, and more useful for theoretical discussions to have measurements of only the true material loss. Further, if the arc burns in a gas which is not chemically inactive, the cathode material can enter into compounds with the filling gas. Part of these can remain on the cathode. Again, measurement of the cathode weight change does not yield a conclusion about the actual material loss. In order to avoid those uncertainties in the results, we established the following precautions: By proper cooling of an appropriately designed anode we easily made sure that it would suffer practically no material loss at all. In order to be able to determine the actual material loss from the cathode, it is necessary to replace weight measurement by weighing by a quantitative chemical analysis, which can be carried out simply and accurately with well-known titration methods. In this process it is more practical and more accurate -- we shall consider another advantage later in the discussion of the experimental results -- not to analyze the cathode before and after the arc burns, but to carry out this analysis on the total material evaporated or sputtered off the cathode. The material carried away from the cathode deposits in the vicinity of the cathode if no volatile gaseous compounds form. It also deposits on the anode, in particular. Therefore it is necessary to make

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the anode of a different material which can easily be distinguished chemically. Therefore, in experiments with iron and copper cathodes we used a silver anode, and in experiments with silver cathodes we used a copper-coated anode at the points under consideration. It is, of course, of decisive importance to collect the entire deposit quantitatively and to conduct the analysis quantitatively.

These considerations, and preliminary experiments, led us to the following design for the discharge vessel. The cathodes were cylindrical rods 2 - 5 mm thick, of the metals to be studied (iron, copper, silver) inserted in a mount which could be moved by means of a vacuum-tight screw lock. The anode was a copper tube 3 cm in diameter. It was sealed on the end by a thin plate of copper or silver, so as to be vacuum-tight. A strong flow of cooling water passed through it. A glass cylinder (see Figure 1) /90 was placed over the copper tube. It was held by two spring clamps, and had a hole opposite the anode to allow the cathode to pass through. There were also some small hoses on the bottom for communication with the outer space. The entire arrangement was mounted by two ground joints in the center of a large glass sphere of about three liters volume. It was connected with the pump and with the manometers (McLeod and mercury side-arm manometers).

The course of the measurement, which always required two observers, was as follows: After the whole discharge vessel had been filled with the gas to be studied, with the usual safety precautions, to the desired pressure, the arc was ignited by touching the cathode and anode together, subsequently separating the electrodes to the desired electrode separation. With some practice this could be done in a few seconds. Then the arc was maintained for a period -- usually 5 minutes -- measured with a stop-watch. During this time one observer monitored the current and voltage and held them constant, while the second observer checked the arc length with a telescope having an ocular micrometer, and kept it constant by moving the cathode if necessary.

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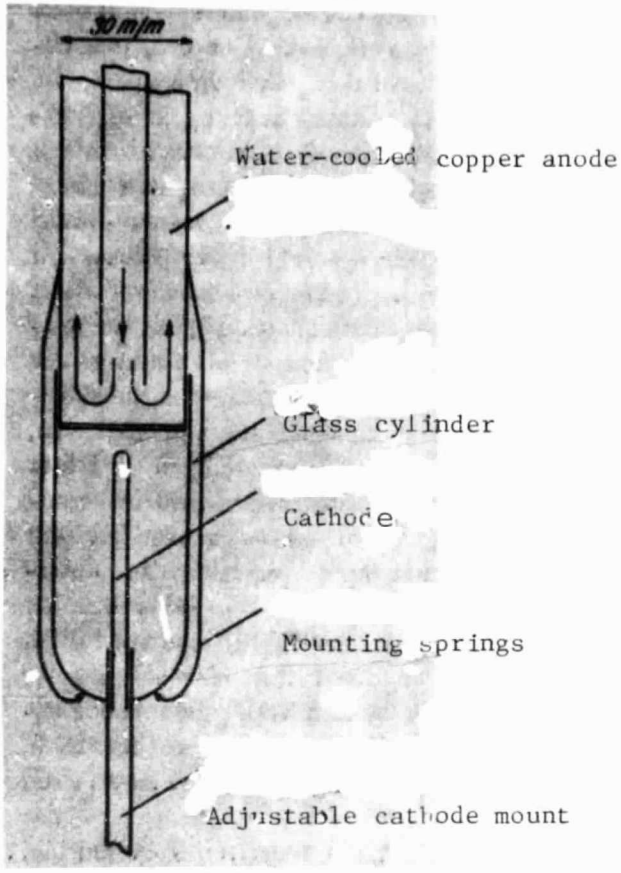

Figure 1. Experimental arrangement (schematic)

Continuous monitoring of the discharge with the telescope was necessary in case any accidental event, such as sputtering or deformation of the cathode by too-fast melting, or separation of part of the deposit from the anode, occurred. Then it could be noted immediately and the experiment could be terminated and rejected. Disturbances due to deformation of the cathode cup occurred particularly with the silver cathodes, particularly if the arc length exceeded a critical value (which decreased with the current). Only somewhat reproducible values for the material loss of the cathode could be obtained. This, unfortunately, severely limited the ranges of the test parameters which could lead to useful results. For instance, in the experiments shown

in Figure 5 for silver in air at 50 cm Hg filling pressure and a current of 2.5 amp, the arc length was limited to less than about 2.5 mm. After quenching of the discharge, dry air was let in and the entire anode with the glass cylinder was removed. The anode and the inside of the glass cylinder (and, if necessary, the jacket surface of the cylindrical cathode) were freed of the deposit on them. As preliminary tests had shown, this could be done most simply and accurately by wiping the coated surfaces with paper filters which were then ashed and titrated. It should be noted that most of the deposit was always found on the end plate of the anode. Only about 5% at most was found on the glass cylinder, and only a negligible small fraction on the part of the cathode which remained cold. This latter fraction, therefore, did not need to be considered in the final experiments. Most of the deposit on the anode end plate was concentrated on a central region of a few millimeters diameter. Its thickness decreased rapidly moving out on the anode endplate surface from the middle of the plate, directly opposite the center of the cathode. The accuracy of the measurements may be characterized by the statement values for the cathode material loss reproducible to less than 5% could be determined. Problems really occurred only with the iron arc in nitrogen and with longer arcs at low gas pressures. The Fe - N_2 arc, in agreement with previous experiment, could not be brought to burn smoothly and evenly for any extended period. We always had to make use of transient forming in air. The longer arcs at low pressure easily showed the well-known tendency to creep along the cathode rod. It was, therefore, a matter of luck and patience to keep such an arc steady for at least a few minutes. When the beginning of creeping appeared in the telescope the experiment was terminated, and the material loss found was extrapolated, in proportion to the burning time, to a burning time of 5 minutes.

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Results

We shall discuss the measuring methods in the next section. First, we shall summarize the results and present them in order. As the preliminary experiments had shown, and as was mentioned in the introduction, a large number of experimental parameters is involved. Of these, the current, the diameter of the cylindrical cathode, and the chemical nature of the filling gas and of the cathode were, of course, well defined. The arc length cannot be determined as accurately, as the discharge did not always take exactly the shortest path between the cathode end plate and the anode end plate. Rather, it oscillated about the shortest middle position except, of course, for the cases where the strike point of the cathode on the jacket of the cathode rod crept and the measurement was terminated. The uncertainty which this causes is small, though. In the following, the arc length is always given as the shortest cathode-anode distance. The burning time, to which the material loss is referred, and to which it should be proportional, is in our experiments the time between ignition and quenching of the discharge. With an experimental duration /93
of 5 minutes the short period of a few seconds needed to pull the electrodes apart to the desired length had no effect at all. But there was an unpleasant and unavoidable uncertainty of another type. The cathode first begins to heat up at the beginning of the burning time, and a steady thermal state must be reached in the cathode and in its vicinity. As preliminary experiments showed, the material loss is not actually strictly proportional to the burning time. Instead, it increases with the burning time, first slowly and then more rapidly. Only in this second stage is the relation between burning time and material loss linear. The length of time until this linear rise is reached, i. e., the length of the "warmup time", depends on the experimental conditions, and principally on the thickness of the cathode rod and on the current. But in the region of the conditions

which we selected these differences are small. The length of the warmup time, about ½ minute, is small in comparison with the burning time of 5 minutes, which was maintained as much as possible, so that the resulting errors are small. As a result we have not separately determined the slope of the linear rise in the final experiments by variation of the burning time, with calculation of the material loss per unit time. Rather, we took the material loss directly from the experimental result, with a total burning time of five minutes, calculated as indicated above from ignition to quenching of the arc. (As already mentioned, it was necessary only in some exceptional cases to limit the burning time to (at worst) 3 minutes, and then to extrapolate proportionally to 5 minutes.) As a result, the values given in the following for the material loss in the steady state are all somewhat too low, but only by 10% at the most, as an estimate based on preliminary experiments showed. For quite well-known reasons, it is quite inaccurate to select the "gas pressure" as a parameter. Instead, we report the filling pressure measured cold. One need only say that it changes in the same sense as the actually interesting (mean) gas density in the discharge /94
space, i. e., in the inside of the glass cylinder we have described. Therefore, the statements on the relation between material loss and gas pressure give only a qualitatively correct picture of the relation between the material loss and the gas density (which varies in and in the vicinity of the arc discharge, and which is therefore difficult to determine).

As already mentioned, the thickness of the cathode rod, the current, the gas pressure, and the chemical nature of the cathode material and of the filling gas all play their parts, so that many combinations are theoretically possible and must be studied individually. The results reported in the following do not, of course, exhaust these possibilities. The experimental conditions were always selected so that the most important relations could be detected. The material loss, M, of the cathode in

milligrams per 5 minutes burning time is always plotted as the ordinate. This is always the actual metal loss, that is, the weight of metal contained in the entire deposit in the vicinity of the cathode, as determined by the quantitative titrimetric analysis.

Figures 2 and 3 show the dependence of M on the current and on the thickness of the cathode for iron and silver in air, at constant arc length. Two different things and one general regularit appear noteworthy in both figures. For one thing, under otherwise the same conditions, M diminishes with increasing cathode thickness, as the preliminary experiments which we mentioned initially had already indicated. The effect of the cathode thickness is more apparent the higher the current is; and it is greater with silver than with iron. The figures also show that M is not always proportional to the current. This appears particularly clearly with silver, so that one can distinguish two regions. At low currents M is proportional to the current. At larger currents it increases, linearly to be sure, but considerably more steeply with the current. A continuous transition region connects these two regions. It is apparently more extensive for iron and only for thin cathodes in our experiments is it exceeded so that the second linear region can be detected. /96

Figures 4 and 5 show the dependence of M on the arc length and on the gas pressure at constant current (2 or 2.5 amp., respectively) and constant cathode thickness (3 mm). Here, again, a regularity appears immediately, namely, that M increases with increasing arc length, but that it tends quickly toward a final value, and this applies for all pressures. /97

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Figures 6 and 7 show the dependence of M on the pressure (gas density) at constant current and constant cathode thickness. If we consider first the curves for iron and silver in air, we find a notable result. While M increases steadily for iron with decreasing gas density, it passes through a maximum for copper

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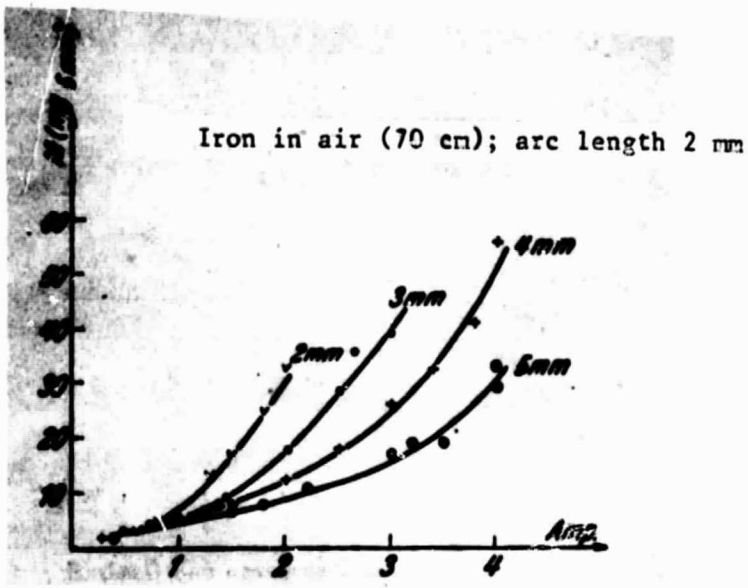

Figure 2. Dependence on current and catnode thickness

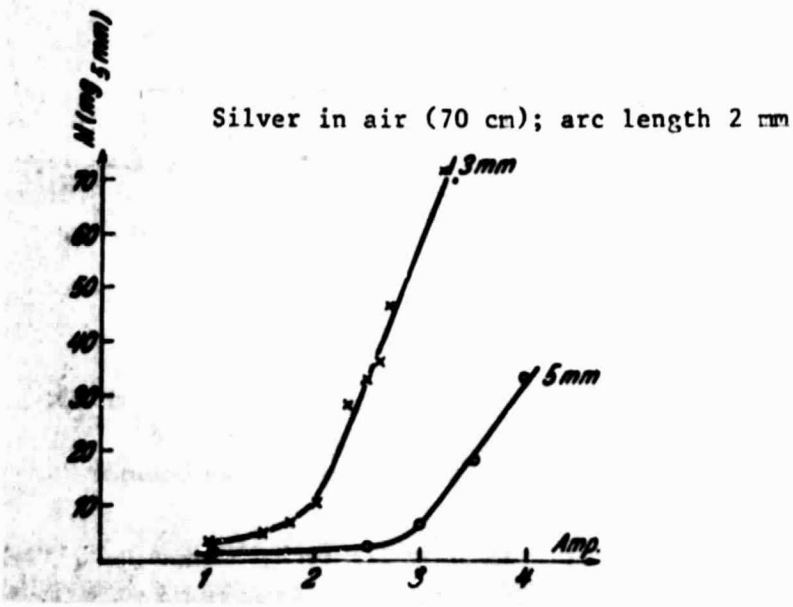

Figure 3. Dependence on current and cathode thickness.

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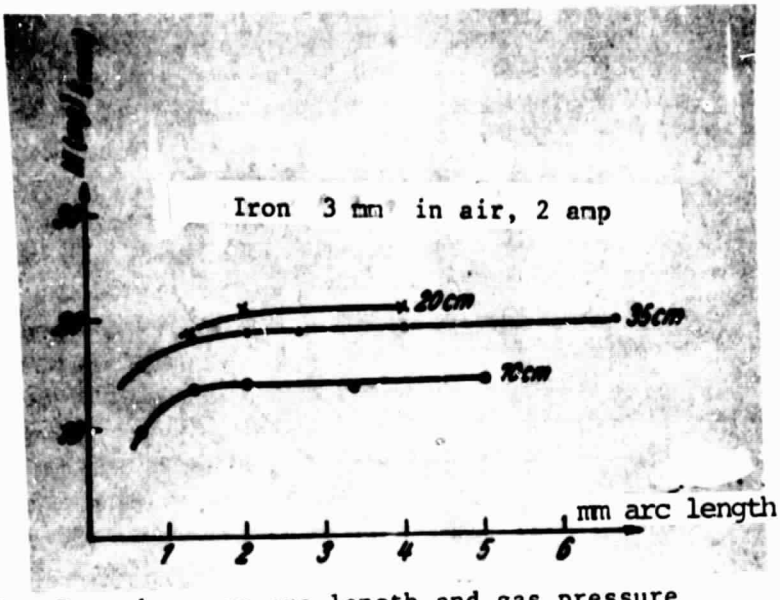

Figure 4. Dependence on arc length and gas pressure

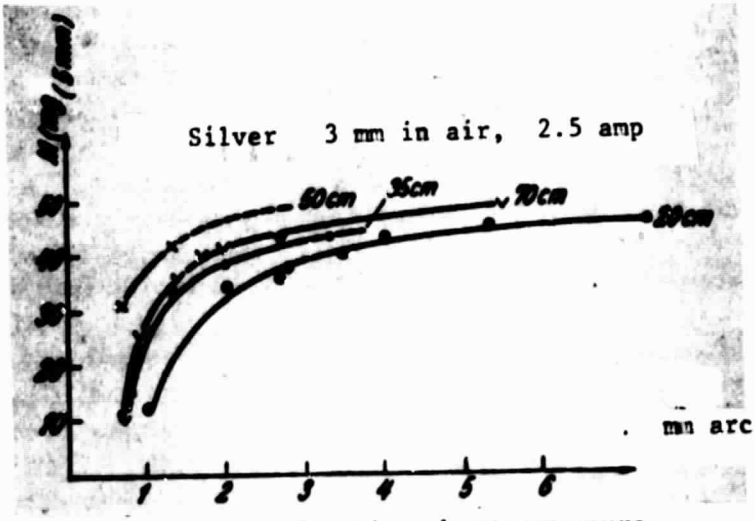

Figure 5. Dependence on arc length and gas pressure

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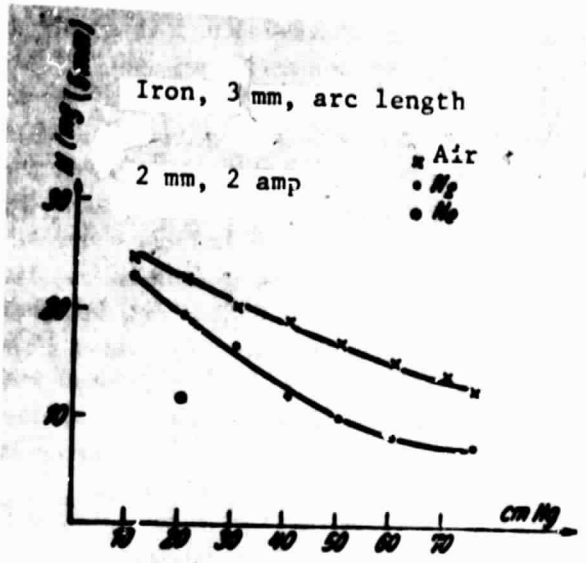

Figure 6. Dependence on the gas pressure.

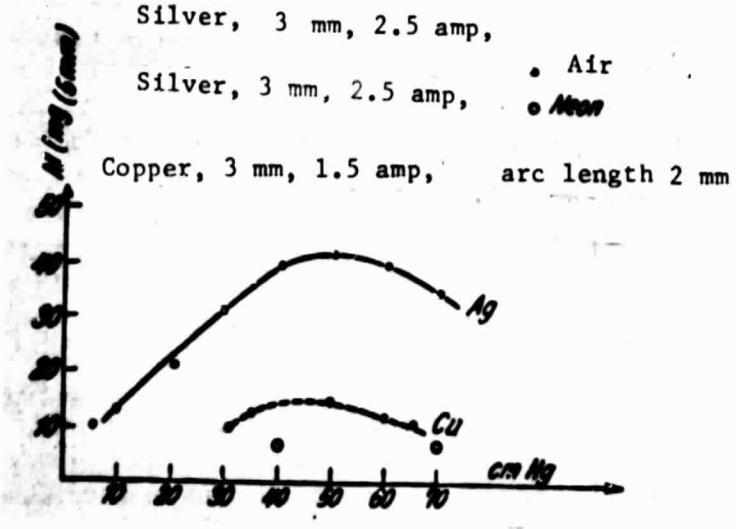

Figure 7. Dependence on the gas pressure.

(and silver). Combination with Figures 4 and 5 shows that this applies for all arc lengths. A second notable result is that M is smaller for iron in nitrogen than in air, and that it is the smallest for iron in neon, where only one point could be measured because of the high gas consumption.

Discussion of the Results

Before evaluating the results shown in Figures 2 - 7 theoretically, we should investigate what physical significance the quantity M actually has. It has been defined heretofore only experimentally as the absolute metal content of the total deposit laid down near the cathode when our experimental arrangement is used. The actual situation is in one respect quite analogous to that which has already been submitted to detailed discussion with respect to measurements of cathode evaporation in the glow discharge, by v. Hippel [3]. Independent of whether one measures cathode material loss directly or from the deposit in the neighborhood of the cathode, the only available measurement is that of how much the cathode finally has lost and has not returned to it. On the other hand, there is a difference between material transport in the glow discharge and in the metal arc, to the extent that in an arc, which burns in the cathode vapor, a reversible material transport is superimposed on the irreversible material loss of the cathode, because the positive ions here consist of charged metal atoms. Unfortunately, the ion current as a fraction of the total cathode current is not yet known accurately. Thus, we can only set an upper (1) limit for transport of metal back to the cathode. Assuming monovalent ions for iron, silver, and copper, because of the high molecular weight, M is always a multiple of the definitively evaporated amounts which we found, namely, 3 M mg/min-amp. Calculation of the detectable amount of substance primarily passing out through the cathode surface from the inside

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(1) Admittedly, it is always well above the actual values.

out into the deposit (which is accessible to measurement) does not seem to us to be feasible, because this is not solely a diffusion problem. The arc discharge with the convection flows is sketched in Figure 8, as seen in the telescope. Perhaps the fact that the metal particles thrown off the cathode are negatively charged in the discharge and are drawn by the field from the edges to the center of the discharge tube may also play a part. The transport of the cathode substance away proceeds principally in the "tube" of diluted gases or vapor filled by the arc discharge -- diluted because of the high temperature. One will hardly doubt that /100
the convection currents in the gas also substantially support the transport. But it is impossible to calculate these. Some orienting experiments showed that such convective flows play a quantitatively significant role. If the anode is vertically above the cathode, as in all the experiments described in the preceding, one gets quite a different distribution of the deposits than for the case in which the anode is vertically below the cathode.

Now we can proceed to the actual discussion of the results summarized in Figures 2 - 7. To a large extent they can be understood with one simple assumption. If we speak of the "evaporation" at the cathode only as a brief designation which is not intended to represent anything about the detailed mechanism of the material loss, then we can assume that the evaporation is not only proportional to the current and occurs at the "hot spot" characteristic of the cathode, but that the further vicinity of the hot spot also is involved in the evaporation, depending on the particular thermal conditions. Energy is put into the cathode at the hot spot, and energy is lost by thermal conductivity, radiation, cooling by convective currents in the gas, etc. As a result, a temperature distribution is established on the end surface of the cathode, which is all that is significantly involved in the evaporation, at least in our experiments. The temperature distribution is variable within wide limits, depending on the particular thermal conditions, and is decisive for the participation

of the neighborhood of the actual hot spot in the evaporation.

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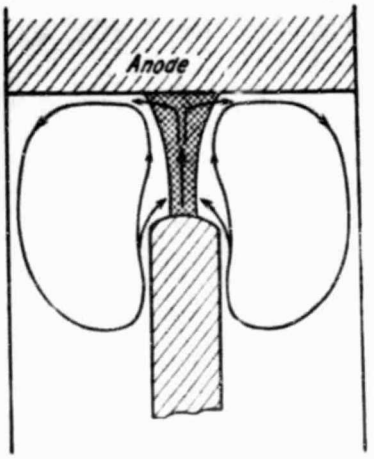

Figure 8. Convective flows in the discharge space.

The finding that M decreases with increasing cathode thickness (see Figures 2 and 3) is immediately understandable. The thicker the cathode is, the more heat is conducted away from the end and the lower the temperature is on the end, or the faster the temperature on the end drops going away form the hot spot to its vicinity. One can also understand the finding that there are two regions of dependence of M on current if one includes the result of the telescope observations. It appears that

at the point where the second region begins the entire end surface has just become a molten cup. The cathode hot spot, even in this state, is still considerably smaller than the cup surface, and wanders rapidly over the surface. This keeps the metal surface in motion. It is not impossible that this causes sputtering of the cathode material along with the regular evaporation, although we could not detect sputtering traces on the collecting surfaces. From the same viewpoint, one can also clarify the increase of M with increasing separation of the anode form the cathode (see Figures 4 and 5). As we have worked with a strongly cooled anode, it is apparent that the cooling of the anode may affect the cathode. Such a thermal coupling between anode and cathode is often found in the literature for the reverse case of a very hot anode [4]. Of course, one must also consider as possible an indirect effect of the convection current in the gas on the transport of cathode vapor away from the cathode, with this effect also depending on the distance between the cathode and the anode plate. The results of Duffield [5], for instance, on burning of the carbon arc electrode, show that the situation can be quite complicated. To be sure, the observations mentioned above on the melting off of the silver cathode above a critical arc length argue that the cathode temperature actually rises with increasing arc length. The fact that M depends on the gas density at all can be explained essentially as an effect of the convection currents depending on the gas density. Another potential explanation for an effect of gas density on evaporation is generally that the composition of the gas around the cathode shifts toward metal vapor as the filling gas pressure decreases, so that this changes the cathode drop. We have no plausible preliminary explanation at all for the fact that this dependence follows quite different rules with respect to gas density for iron and silver, and for copper (Figures 6 and 7).

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We cannot yet satisfactorily explain the dependence of M on the chemical nature of the filling gas. The fact that, under

the same conditions M is lower in nitrogen than in air and lower in neon than in nitrogen could, to be sure, indicate that chemical processes between the filling gas and the cathode material are involved in the cathode material loss; but this is still quite unclear. This might, for instance, be a purely electrical effect, i. e., a difference in the cathode drop and current distribution around the cathode in the different gases. As far as we know, there are no measurements from which a decision could be made. Quite generally we may comment on the preceding that we have of course measured the discharge voltage for all the points shown in the figures. We see no indication for an explanation of our observations in the sense of any relation between the M values and the arc voltage or the power consumption. Therefore, we have not particularly noted the arc voltages in the figures.

The result of our experiments can, then, be summarized generally as follows: The material loss from the arc cathode cannot in any case be considered simply as evaporation at the hot spot. Furthermore, the hot spot cannot be considered as a physical concept of constant structure under otherwise identical conditions, with area which grows in proportion only to the current. The entire wide neighborhood of the hot spot participates in the evaporation, and the entire thermal and aerodynamic situation plays a decisive part. v. Issendorf first recognized this, and expressed it clearly, from his measurements in mercury arcs which we cited in the introduction. The situation is much more complex than was believed initially; and formulation of the cathode energy balance is much less simple. A particular problem is that the evaporation term in the energy balance can no longer be taken as proportional to the current (and it also depends on a series of experimental parameters, as is generally the case also for the cathode heat loss by conduction, convection, etc.). The reuslt is that one can no longer set up an energy balance strictly in the original simple form, that is, independent of the current, and that we are apparently led to a problem of fundamental importance. That is, if we consider the energy

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balance as a relation between the current distribution f = electronic portion:ionic portion of the discharge current, and the cathode drop V_e, this relation must still contain the current explicitly. f or V_e or both must depend on the current. Wehrli [6] has already recognized this clearly in his works on the cathode of a gas discharge, and has presented it in formulas.

As no quantitative results at all are available for evaporating metal cathodes other than mercury, we wish to consider this investigation as an initial experiment toward evaluation of the situation in the sense of the general Wehrli energy balance. Finally, with respect to the initially cited speculations of Schmick which had the objective of coordinating the arc and glow discharge cathode processes from the same viewpoint, things appear to be as follows. If a local high and steep temperature jump appears at the impact point of a positive ion on the cathode surface, nd this is to be made responsible for the evaporation, there are essentially two extreme cases which are conceivable. As v. Hippel, for instance, assumed in his theory of cathode evaporation, each such temperature jump can be considered to exist in isolation and to vanish again into the immediate surroundings of the impact point after completion of the evaporation process. But it can also be considered that the individual jumps are not isolated but rather can "run together", whether because of excessive spatial density or temporal density, or both, and heat up the entire cathode to a high temperature. In the initial case, M must be proportional to the number of such spikes. That is, it must be proportional to the current. Even in the second case there is proportionality with the current if the spike density (i. e., the current density) remains constant and the thermal edge effect at the "hot spot" plays only a quantitatively negligible role. As calculation of the applicable heat conduction problem shows, this is the case for current strengths which are not very small, as long as the size of the hot spot is not comparable with that of the entire end area of the cathode. The details

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will be reported in a theoretical investigation. Here it may suffice to indicate that from the results of this work one cannot decide between the two possibilities. Deeper analysis is necessary. We hope, in new measurements of cathode evaporation, to be able to separate the true primary material loss from the cathode from the material regain by back-diffusion. This appears to be possible if the cathode is moved through the gas continuously in a suitable manner so that no diffusion equilibrium can take place.

We thank the Helmholtz Society and the Emergency Association for assured aid. Mr. J. v. Issendorff has contributed to discussion of the results by critical comments for which we thank him.

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