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ACCOMMODATION COEFFICIENT OF GASEOUS IONS AT CATHODES

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The behavior of a cathode in an ionized gas can be studied from three principal points of view: first, electrical relations described by the application of Poisson's equation to the surrounding region; second, thermal relations described by the application of the energy principle to the various processes which develop or absorb heat at the cathode surface; third, pressure reactions which are described by the application of the principle of conservation of momentum at the cathode surface.

The electrical relations were first pointed out by J. J. Thomson[1] and later made more specific and greatly extended by Langmuir and his collaborators.[2] From these considerations, applied to current-carrying and to exploring electrodes, we have gained a nearly complete picture and interpretation of the phenomena occurring between the electrodes. Of equal importance with this, however, is an understanding of the phenomena occurring at the electrode surfaces, especially at the cathode. For this study we need more information than can be gained from Poisson's equation. We need to know, for example, what fraction of the current at the cathode is carried by electrons emitted from it; what is the mechanism responsible for this emission; if this emission is of thermionic origin, what factors maintain the requisite high temperature, etc. Since there are several unknown quantities, we obviously approach the solution by investigating from several independent points of view, so as to get several independent equations. It is particularly for this reason that studies of energy and pressure relations at a cathode have considerable significance. The most complete analysis of these relations, thus far made, is in a recent paper by the author[3] which, while directed particularly at the problem of the mercury arc, is nevertheless generally applicable in principle.

Attempts to investigate cathode phenomena by studying its heat balance were begun about ten years ago[4] but conclusions based on them have largely been vitiated through their failure to recognize the existence of an "accommodation coefficient" for ions neutralized at a cathode surface. This phenomenon was discovered by Compton and Van Voorhis[5] as a result of their observation that the heating of a cathode, bombarded by positive ions of known kinetic energy, was less than expected, even after making due allowance for complicating effects such as secondary electron emission. The interpretation is as follows:

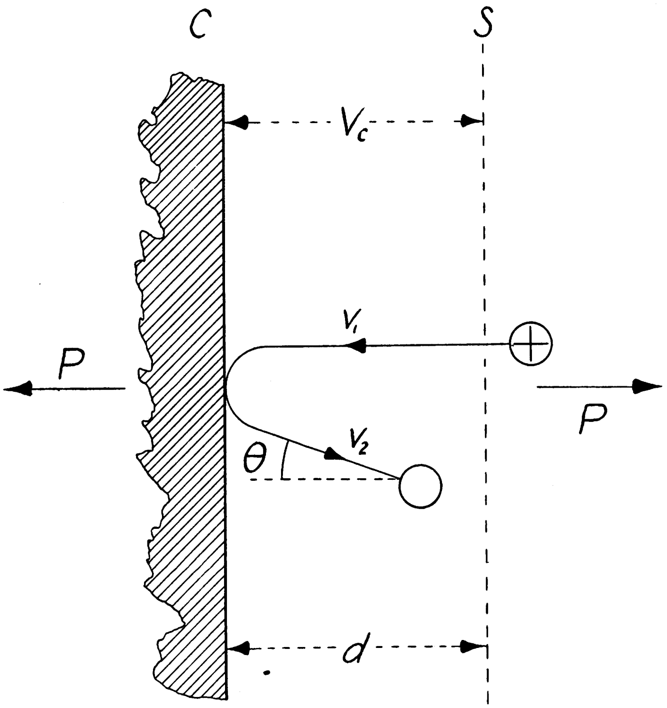

FIGURE 1

In figure 1 let C represent the cathode surface and S the boundary of the positive ion space charge sheath which always surrounds a cathode. The drop in potential V_c between the surrounding gas and the electrode is concentrated within this sheath, whose thickness we will call d. A positive ion, drifting across S, is pulled to the cathode by the field and strikes with kinetic energy $E_1 = eV_c$ and momentum $Mv_1 = (2eMV_c)^{1/2}$. The point which had been previously neglected was that this ion, after neutralization, may leave the cathode with an appreciable energy $E_2 = {}^1/_2\, Mv_2^2$. Thus there is a heating effect on the cathode of $H = E_1 - E_2$ and an impulse delivered to it of $Mv_1 + Mv_2 \cos \theta$. Of this impulse, however, the term Mv_1 is exactly compensated by the mutual pull between cathode and ion during the ion's approach, so that only the term $Mv_2 \cos z$ contributes to pressure on the cathode.

These effects may be described in terms of the "accommodation coefficient" α which has been used to describe the energy transfer of gas molecules at temperature T_1 striking a surface of temperature T_s and leaving with temperature T_2, according to the definition $\alpha = (T_1 - T_2)/(T_1 - T_s)$. If the molecules, during contact, come into thermal equilibrium with the surface, $\alpha = 1$; if the molecules rebound elastically, $\alpha = 0$. Table 1 gives some values of α, thus obtained from thermal measurements.

TABLE 1

ACCOMMODATION COEFFICIENTS OF MOLECULES

GAS	METAL	METAL TEMPERATURE	α	SEE FOOTNOTES
H_2	Pt	20°C.	0.26	6
		−100	0.25	7
		+200	0.15	7
	W	+1500	0.19	8
He	Pt	−100	0.49	7
		+200	0.37	7
		~20	0.34	9
	W	~20	0.06	10 clean
		~50	0.53	11 dirty
		~50	0.17	11 clean
N_2	Pt	~20	0.87	6
	W	+1500	0.60	8
Ne	Pt	~20	0.65	9
A	Pt	~20	0.86	9
	W	~50	1.00	11 dirty
			0.82	11 clean

As might be expected, the values are extremely sensitive to the condition of the surface, such as films or roughness causing multiple impacts. However, so far as the data go, they indicate increasing accommodation coefficients with increasing molecular weights.

In the case of ions, the accommodation coefficient is $\alpha = (E_1 - E_2)/E_1$. From thermal measurements Compton and Van Voorhis[5] found the accommodation coefficients given in table 2. Considering the uncertainties of surfaces, these values are suggestively like those of the corresponding neutral molecules. In fact we should expect the ions to behave as neutral

TABLE 2

ACCOMMODATION COEFFICIENTS OF IONS ON MO, DETERMINED THERMALLY

He	0.35	for $35 < V_c < 51$ volts
	0.55	for $111 < V_c < 141$ volts
Ne	0.65	for $21 < V_c < 141$ volts
A	0.75	for $21 < V_c < 141$ volts

molecules (except as influenced by their higher velocities) since they are presumably neutralized during contact with the cathode.

Three years ago Tanberg[12] reported measurements of pressure against the cathode of a Cu arc in a partial vacuum, and also pressure against a neighboring vane by a stream of neutral particles moving away from the cathode. Since the copper cathode was slowly volatilized, Tanberg assumed that these pressures were due to the impulsive reactions of the evaporating Cu atoms. This assumption led to the conclusion that this copper evaporated at a temperature of about 500,000°C.! The author, however, pointed out that Tanberg's pressures could be explained by momentum transfers by ions neutralized at the cathode surfaces, if reasonable values were assumed for their accommodation coefficients.[13] The decisive test of such an explanation, as contrasted with Tanberg's, would obviously be given if similar pressures were observed with non-volatilizing cathodes. This has actually been done[14] resulting in confirmation of the author's theory of cathode pressures as well as in a new method of measuring accommodation coefficients and, incidentally, the fraction of cathode current carried by electrons. As these results will be published elsewhere in detail, only those parts which are significant for the present subject will be summarized as follows:

The cathode was a molybdenum vane, about 1 cm. square, with its back protected by an insulating cover of glass, serving as the bob of a pendulum. This pendulum bob vane was suspended in a large helium arc tube, carrying several amperes at a gas pressure of the order of 1 mm. The deflection of the pendulum, when positive ions were drawn to it as a cathode, permitted the pressure against it to be computed. Neglecting for the present explanatory purposes two corrections for complicating phenomena (one of which gave us the fraction of current carried by electrons), the pressure is readily shown to be

$$P = I_{+}(MV_c/2e)^{1/2}(1 - \alpha)^{1/2} \qquad (1)$$

if the neutralized ions rebound with equal probability in all directions like light from a matt surface. If the rebound is always normal to the surface, the above expression should be multiplied by 2. The former assumption seems more probable, and gives a slightly more consistent description of the experiments, but this aspect of the problem still needs further examination.

The accommodation coefficient thus calculated from the pressure on a molybdenum cathode in ionized helium is shown in table 3. Two sets of values are given, depending on whether the heat of neutralization of a positive ion of zero velocity at the surface is equal to zero or equal to the difference between the ionizing potential of the gas, V_i, minus the electron

TABLE 3

ACCOMMODATION COEFFICIENTS OF He IONS ON Mo DETERMINED BY PRESSURE

V	$\alpha(\varphi_+ = 0)$	$\alpha(\varphi_+ = V_i - \varphi_-)$
35	0.681	0.463
65	0.570	0.480
95	0.512	0.449
125	0.410	0.365

work function φ_-. The uncertainty here depends on how much of the available energy in neutralization may be radiated away or carried away by the neutral atom in an excited state. The latter phenomenon is known to occur with a high degree of probability in the case of helium ions neutralized by glancing contact with a metal surface. Unfortunately all experimental attempts to measure φ_+ have thus far not been sufficiently devoid of other complications to enable a definite value to be set between these extreme limits.

In comparing the values in tables 1, 2 and 3 it is seen that they agree in order of magnitude and that the agreement between the three methods is about as good as that between different observers using one method, cf. table 1. Precise agreement is scarcely to be expected since the phenomenon is one, like electron emission or optical reflection, which depends on the condition of the metal surface and would therefore be expected to vary with films of gas or other impurity, and with the degree of roughness of the surface. Direct evidence of this is found in the experiments of Michels,[11] quoted in table 1.

Interpretation of Accommodation Coefficient.—As pointed out above, we should expect the phenomena which determine accommodation coefficients to be identical for neutral molecules and for neutralized ions, since the two particles are identical as they leave the solid surface. With ions, however, we have much higher velocities (effective temperatures) than with gas molecules, and we have them striking the surface normally instead of isotropically as regards direction. Both of these peculiarities would be expected somewhat to increase the accommodation coefficient of ions over that of molecules.

The following interpretation of accommodation coefficients appears satisfactorily to account, in a qualitative way, for such observations as have thus far been made.

Zener[15] has recently developed a quantum theory of the accommodation coefficient according to which the loss of energy of a molecule rebounding from a solid surface is computed by summing up the probabilities of transition of energy to the quantum states associated with the various degrees of freedom of the solid. For simplicity he carried through the calculations only for the degree of freedom normal to the surface. In dealing with ions, however, whose effective temperature greatly exceeds that of the solid, it would appear legitimate to apply the simpler classical principles, for exactly the same reasons that these principles may be applied at high temperatures to specific heats.

The simplest classical picture of the phenomenon is that of an elastic impact between the impinging molecule, of mass M and a surface atom of mass M_s. Rebound occurs only if

$$M_s > M \tag{2}$$

and the average energy of such rebounding molecules is

$$E_2 = \frac{M^2 + M_s^2}{(M + M_s)^2}(E_1 - E_s) + E_s \tag{3}$$

whence the accommodation coefficient is*

$$\alpha = \frac{2MM_s}{(M + M_s)^2} \text{ subject to } M_s > M. \tag{4}$$

If we extend these ideas to include the possibility of rebound of molecules after n collisions with surface atoms, we find that the accommodation coefficient α_n appropriate to n collisions is given by

$$\alpha_n = 1 - (1 - \alpha)^n, \tag{5}$$

so that the effect of multiple collisions (as with a roughened surface) is to make the accommodation coefficient approach unity. In general, molecules impinge some once, some twice, some thrice, etc., so that the actual accommodation is a weighted mean of values calculated from Eqs. (4) and (5) with $n = 1, 2, 3\ldots$etc. We have no means of knowing the distribution of n, so that we can only say that Eq. (4) gives a lower limit to α and that α should increase with roughness of surface.

The most striking results of this theory are (1) the conclusion that there is no rebound if $M_s < M$. We should thus expect the accommodation coefficient to equal unity (or *nearly* so, owing to the crudeness of the theory) if the mass of the ion exceeds that of the surface atom and (2) the conclusion that the accommodation coefficient is smallest for light ions.

Conclusion (1) is supported by the fact that there is no evidence of a pressure against the cathode of a carbon arc[17] such as would be caused by the existence of an accommodation coefficient less than unity for the positive ions (which in this case would be of masses equal to or greater than those of surface atoms) and by the fact that the covering of a tungsten wire with naturally occurring impurities, presumably oxide and oxygen layers, raised its accommodation coefficient for argon atoms from 0.82 for clean tungsten to unity.[11] Furthermore, pressure against the cathode of copper and iron arcs in air[17] are also so small that there is no evidence of effect of accommodation coefficient, and this again may be due to the fact that here the cathode surface is covered by layers of oxide so that the outer oxygen layer is composed of atoms with mass equal to or less than those of the bombarding ions.

Mr. Lamar has pointed out to the writer that the absence of pressure in these arcs at atmospheric pressure may also be due to the compensating effect of holding back the atmospheric pressure from the cathode spot—a compensation which could not occur if the cathode were of large surface area, but which can occur as the result of convection currents set up around the cathode if this is of relatively small dimensions. On this assumption Lamar was able to calculate the area of the cathode spot of a copper arc to the right order of magnitude and he is now undertaking an experimental test of this suggestion.

Conclusion (2) is supported by all evidence at present available.

These considerations suggest interesting further experiments on the relation of M/M_s to α, with particular practical interest in the behavior of mercury ions, which is now being investigated.

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* This assumes a *single* collision (as in Zener's theory) but allows for random distribution of direction. Had "head on" collisions only been considered, the factor 2 would have been replaced by 4. These considerations were first advanced by Baule.[16]

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