

The theory of cathodic bombardment in a glow discharge by fast neutrals

To cite this article: R S Mason and R M Allott 1994 *J. Phys. D: Appl. Phys.* **27** 2372

View the [article online](#) for updates and enhancements.

Related content

- [The contribution of fast neutrals to cathode erosion in glow discharges](#)
I Abril, A Gras-Marti and J A Valles-Abarca
- [Sputtering in a glow discharge ion source-pressure dependence: theory and experiment](#)
R S Mason and M Pichilingi
- [Energy distribution of heavy particles generated in the cathode fall of a gas mixture glow discharge](#)
Z Wronski, J L Sullivan and C G Pearce

Recent citations

- [Influence of Pressure on the Morphology and Structure of Surfaces Sintered in Pulsed DC Annular Hollow Cathode Discharge](#)
S.F. Brunatto *et al*
- [Emergence and consequences of lateral sample heterogeneity in glow discharge spectrometry](#)
Andrew P. Storey *et al*
- [Silvio Francisco Brunatto *et al*](#)

The theory of cathodic bombardment in a glow discharge by fast neutrals

Rod S Mason and Richard M Allott

Department of Chemistry, University College of Swansea, Swansea SA2 8PP, UK

Received 6 April 1994, in final form 20 July 1994

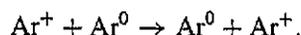
Abstract. A mathematical model is developed to calculate the flux and average energy of fast neutrals in a glow discharge based on the charge transfer model used to describe the energy distribution of ions. According to this model the average energy and relative distribution functions of both fast atoms and ions are found to be independent of pressure. Using experimental values found in the literature it is found that the fast atom flux is at least twice as large as the ion flux, but with an average energy similar to the ions. The sputter erosion of the cathode is therefore probably caused mainly by fast atom bombardment.

1. Introduction

The glow discharge has been of both fundamental and practical interest for well over a century. The birth of the semiconductor industry and the requirement for precise and accurate analytical measurements has, however, led to renewed interest. Industrial applications include surface cleaning, plasma etching and thin film production [1]. In addition, the glow discharge lends itself as an excellent atomization source for analytical investigation—in particular, glow discharge mass spectrometry [2] (GDMS) (the motivation behind this work), and optical analytical techniques including absorption [3], emission [4] and opto-galvanic [5] spectroscopies.

In GDMS the sample for analysis forms the cathode of the glow discharge system, and is subsequently sputter eroded by ions and also fast neutrals, resulting in atomization of the sample. In order to understand these processes fully, a knowledge of the energy distributions of the particles responsible for cathodic sputtering is vital.

Ions originating in the negative glow region of the discharge are accelerated towards the cathode by the cathode fall potential which spans the relatively narrow cathode dark space (CDS). The final kinetic energy of the ions is moderated by symmetrical charge transfer (SCT), i.e.



When the accelerating ion undergoes SCT, a new ion is formed some distance into the CDS, with effectively zero energy. This ion then begins to accelerate through the potential drop until it either strikes the cathode or itself undergoes charge exchange. It is clear then that the ions striking the cathode have a distribution of energies and that the average energy will be much less than the full

cathode fall potential. This simple model was developed in 1963 by Davis and Vanderslice [6] to describe their experimental results pertaining to directly measured ion energy distributions at the cathode. The model is now generally accepted and has been used as the starting point for virtually all the theoretical work performed over the past decade.

In addition to moderating the ion energies each SCT collision leads to the production of a fast neutral atom. Since symmetrical charge transfer is a long range process, there is very little momentum exchange, so that the neutral atom will continue on towards the cathode with essentially the same energy as the parent ion. However, collisions with the discharge gas particles may slow or even 'stop' the neutrals before they strike the cathode. The combination of this process with charge exchange collisions determines the neutral atom energy distribution.

The theoretical prediction of this neutral energy distribution has been the subject of a great deal of work based on relatively complicated mathematical and computational procedures over the past ten years. A brief resume of the main contributions is given in section 3. In this work we derive an expression for the neutral energy distribution which includes the contribution due to scattering. Although this expression requires numerical evaluation, we are able to present analytical solutions for the total neutral flux and for the average neutral sputtering energy. The model is based on a more simplistic procedure than has been seen previously, and this allows results to be calculated for 'real' practical situations such as those found in the GDMS laboratory.

2. The ion energy distribution

Since the theoretical models presented in the literature

are all based on the SCT model of Davis and Vanderslice [6], it is informative to present the final form of the ion energy distribution function f_E^I which we have derived, also based on this model. This is essentially the same as that found in the literature [6] (for a more general expression see also Rickards [7]).

$$f_E^I = I_0 \frac{d}{2\lambda} (1-E)^{-1/2} \exp\left(\frac{-d}{\lambda} [1 - (1-E)^{1/2}]\right) \partial E \quad (1)$$

where I_0 is the total ion flux, d is the extent of the cathode dark space and $\lambda = 1/nq$ is the mean free path for SCT, where n is the gas density and q the cross section for charge transfer. Here E is the relative energy of the ions with respect to the cathode fall potential.

Addition of the term $I_0 \exp(-d/\lambda)$ to equation (1), representing the fraction of ions that traverse the dark space without undergoing collision, allows the self-consistent result $F^I = I_0$ upon integration of the above expression over the full range of energies from eV_c to 0 electron volts. The average ionic sputtering energy can be simply defined as:

$$\bar{E}_{\text{ions}} = \frac{\int f^I E}{\int f^I} \quad (2)$$

yielding an expression of the form

$$\bar{E}_{\text{ions}} = \left[\left(\frac{2\lambda}{d} - \frac{2\lambda^2}{d^2} \right) - \left(1 - \frac{2\lambda^2}{d^2} \right) \exp\left(\frac{-d}{\lambda}\right) \right]. \quad (3)$$

3. Other work

A measure of the interest in the particle energy distributions in glow discharges is reflected in the wealth of theoretical and computational models found in the literature. In particular, the work of Abril *et al* [8, 9] should be emphasized. Their model for the ion and neutral energy distribution functions is again based on the assumptions made by the model of Davis and Vanderslice. Namely, that the electric field variation is linear and the cross section for charge transfer is independent of energy. This, of course, is not strictly true and in fact the cross section is seen to decrease with increasing energy [10]. Also in describing the energy distribution function (EDF) it is assumed that the neutrals formed by symmetrical charge transfer proceed towards the cathode without further collision. In an attempt to remove the uncertainty associated with the electric field variation, a rather complicated model was set up in a later publication [11] in which the electric field was calculated self-consistently. However, the excellent measurements reported by Den-Hartog *et al* [12] using the opto-galvanic effect to measure the electric field variation suggest that it is indeed linear. We therefore base our model on the linear field approximation.

Wronskii [13] modified the Davis and Vanderslice model by including ionization by electrons and ions

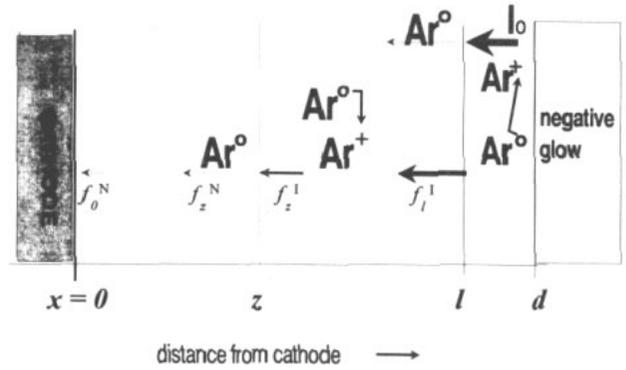


Figure 1. Charge transfer model of cathodic fast atom bombardment.

in the cathode fall region. However, the contribution to the overall energy distributions was found to be small, due to the cross sections for these processes being small as compared to the charge transfer cross section. In calculating the neutral energy distribution, Wronskii ignores effects due to elastic scattering and also bases his model on a nonlinear field approximation. Other groups worthy of mention include Mase and Tanabe [14], Rickards [7], Kuchinskii and Sheikin [15] and Donko and Janossy [16]. With the exception of Mase and Tanabe (who only deal with ions and not neutrals), the above works all rely on rather complicated mathematical functions and/or Monte Carlo simulation techniques to elucidate the distribution functions. Despite the theoretically reasonable expressions derived, the results tend to be somewhat obscured by complex mathematical functions. It is the purpose of this work to provide equations which are not only analytically tractable but also may be related directly to practical situations.

Since the first measurements by Davis and Vanderslice, various attempts have been made to measure the ion EDF experimentally. One of the most recent is the work of Peter *et al* [17], where ion energies are measured in gas mixtures. On the whole, the various theoretical models fit well to the experimental data, though it was found that the measured ion energies were higher than those predicted theoretically. To date there seem to have been no convincing experiments performed to extract neutral atom energy distributions.

4. Theory

4.1. The bombarding fast neutral flux

Fast neutrals that are formed by symmetrical charge transfer at a point $x = z$ (see figure 1) continue towards the cathode with an energy equal to that of the parent ion, unless they are 'stopped' by collision with the gas. The parent ions originate at a point $x = l$. Under steady state conditions, the flux of ions of all energies passing through the plane $x = l$ must be I_0 , and the fraction undergoing charge transfer creates a flux f_l^I .

As these ions accelerate to the point $x = z$, a proportion will undergo charge transfer, so that the fraction of ions surviving to the point $x = z$ is

$$f_z^I = f_l^I \exp\left\{\frac{-(l-z)}{\lambda}\right\}. \quad (4)$$

Now a fraction of these ions will undergo charge transfer at the point $x = z$, creating a flux of fast neutrals f_z^N , where

$$f_z^N = f_z^I \frac{\partial z}{\lambda}. \quad (5)$$

The energy of this fast neutral flux will be equivalent to the energy gained by the parent ions in accelerating through a field defined by

$$\varepsilon_x = \frac{2V_c}{d} \left(1 - \frac{x}{d}\right) \quad (6)$$

between the points $x = l$ and $x = z$, so that

$$E_{l-z}^N = \frac{2eV_c}{d} \left(l - z - \frac{l^2}{2d} + \frac{z^2}{2d}\right). \quad (7)$$

In order to simplify the expressions further, we define the neutral flux as being relative to the total ion flux I_0 , and the energy term above will be expressed simply as E , the relative energy with respect to the cathode fall potential V_c .

Considering the case where there is no attenuation due to collision with the gas, the total neutral flux is given by

$$F^N = \frac{1}{\lambda^2} \int_{l=0}^d \int_{z=0}^l \exp(-l/\lambda) \exp(z/\lambda) \partial l \partial z. \quad (8)$$

Integration of equation (8) yields the expression

$$F^N = \frac{d}{\lambda} + \exp(-d/\lambda) - 1. \quad (9)$$

This expression does not include the contribution to the flux from ions formed at the point $x = d$. Adding the term

$$\frac{1}{\lambda} \int_{z=0}^d \exp(-d/\lambda) \exp(z/\lambda) \partial z = 1 - \exp(-d/\lambda) \quad (10)$$

to equation (9) above leads to the result $F^N = d/\lambda$ (which is consistent with that of Abril *et al* [8, 9]).

However, not all of the neutral flux will survive to the cathode and a fraction will be 'stopped' by collisions with the gas. We define a 'stopping' cross section q_s , to account for this process. The magnitude of q_s will be related to the elastic scattering cross section and is discussed later. Again, q_s is assumed to be independent of energy as with q , the charge transfer cross section. The mean free path for 'stopping' is then given by

$$\lambda_s = \frac{1}{nq_s}. \quad (11)$$

From equation (8) the flux of neutrals reaching the cathode is

$$f_0^N = \frac{1}{\lambda^2} \exp(\alpha l) \exp(\beta z) \partial l \partial z \quad (12)$$

where $\alpha = 1/\lambda$ and $\beta = (1/\lambda - 1/\lambda_s)$. The total neutral flux is then given by integrating (12) first with respect to z for $0 \leq z \leq l$ and second with respect to l for $0 \leq l \leq d$, so that

$$F^N = \frac{\lambda_s}{(\lambda_s - \lambda)\lambda} \left[\lambda \exp\left(-\frac{d}{\lambda}\right) - \lambda_s \exp\left(-\frac{d}{\lambda_s}\right) + (\lambda_s - \lambda) \right]. \quad (13)$$

Again, there is a small contribution to the flux from those ions created at the point $x = d$. This is given by

$$\frac{1}{\lambda} \int_{z=0}^d \exp(-d/\lambda) \exp(\beta z) \partial z \quad (14)$$

yielding, upon integration, the term

$$\frac{1}{\lambda\beta} \{ \exp[d(\beta - 1/\lambda)] - \exp(-d/\lambda) \} \quad (15)$$

which should be added to equation (13) to give the total neutral flux arriving at the cathode ($x = 0$).

4.2. The neutral energy distribution

At a fixed value of E the value of z can be obtained in terms of l and d by rearranging equation (7) to give a quadratic equation for which the physically reasonable solution is

$$z = d - d \left(1 - \frac{2l}{d} + \frac{l^2}{d^2} + E\right)^{1/2} = d(1 - g_{l,E}) \quad (16)$$

where

$$g_{l,E} = \left(1 - \frac{2l}{d} + \frac{l^2}{d^2} + E\right)^{1/2}. \quad (17)$$

Partial differentiation of E with respect to z at a fixed value of l gives

$$\frac{\partial E}{\partial z} = -\frac{2}{d} g_{l,E}. \quad (18)$$

Substitution of z and ∂z into equation (12) gives the neutral flux at the cathode in terms of l and E

$$f_0^N = \frac{d}{2\lambda^2} \frac{\exp[\alpha l + \beta d(1 - g_{l,E})]}{g_{l,E}} \partial l \partial E. \quad (19)$$

The total flux f_E^N of fast neutrals with a single energy E is then obtained by integrating equation (19) from the minimum possible value of l to $l = d$. This is then the energy distribution of the fast neutrals

$$\frac{f_E^N}{\partial E} = \int_{l_{\min}}^d f_0^N \partial l. \quad (20)$$

Table 1. Simplified expressions. All symbols and reference conditions are as defined in the text.

Description	Function	Calculated value
Total ion flux I_0		$I_0 \approx 2 \times 10^{16} \text{ cm}^{-2}$
Ion energy distribution function	$f_E^I = I_0 \frac{d}{2\lambda} (1-E)^{-1/2} \exp\left\{\frac{-d}{\lambda}[1-(1-E)^{1/2}]\right\} \partial E$	see figure 2
Average ion energy	$\frac{2\lambda}{d} \left(1 - \frac{2\lambda}{d}\right) eV_c$	68 eV
Total neutral flux F^N	$\frac{\lambda_s}{\lambda} \left[1 - \frac{\lambda_s}{(\lambda_s - \lambda)} \exp\left(\frac{-d}{\lambda_s}\right)\right]$	$2 \times I_0$ for $q/q_s = 2$ $9 \times I_0$ for $q/q_s = 10$
Neutral energy distribution function	$\frac{d}{2\lambda^2} \int_{l_{\min}}^d \frac{\exp[\alpha l + \beta d(1 - g_{I,E})]}{g_{I,E}} \partial l$	see figure 2
Average neutral energy	$\frac{(A+B) \exp(-d/\lambda_s) - D}{F^N}$	59 eV for $q/q_s = 2$ 44 eV for $q/q_s = 10$

For a given energy E , the minimum value of l is when charge transfer occurs at or very close to the cathode surface, i.e. when $z = 0$ in equation (16) giving

$$l_{\min} = d - d(1-E)^{1/2}. \quad (21)$$

In analogy with the total neutral flux, a term

$$f_0^N(\text{add}) = \frac{d}{2\lambda} \frac{\exp(\alpha d) \exp \beta d(1-E^{1/2}) \partial E}{E^{1/2}} \quad (22)$$

has to be added to equation (19) to give the correct expression. Regrettably, there does not appear to be an analytical solution for the neutral atom energy distribution but the function may be computed numerically.

4.3. The average neutral sputtering energy

By direct analogy with equation (2) for ions, the weighted energy of the fast neutral atoms is given by

$$\bar{E}_N = \frac{\int_0^{eV_c} f_E^N E \partial E}{\int_0^{eV_c} f_E^N \partial E}. \quad (23)$$

Clearly, the denominator in equation (23) is equal to F^N , the total neutral flux. However, the numerator appears to be intractable. This may be overcome by considering the fact that

$$\int_0^{eV_c} f_E^N E \partial E = \int_{l=0}^d \int_{z=0}^l f_0^N E_{l-z}^N \partial l \partial z. \quad (24)$$

Again the term for the case when $l = d$ must be added to this. The whole expression now becomes amenable to integration and a result of the form

$$\bar{E}_N F^N = (A+B) \exp\left(\frac{-d}{\lambda_s}\right) + (C-A) \exp\left(\frac{-d}{\lambda}\right) - D \quad (25)$$

where

$$A = \frac{d^2 \lambda_s}{6(\lambda_s - \lambda)} \quad (26)$$

$$B = \frac{2(2\lambda - \lambda_s)\lambda_s^4}{d^2(\lambda - \lambda_s)^3} \quad (27)$$

$$C = \{[\lambda_s(d^2\lambda^2 - 2\lambda^4 - 2d^2\lambda\lambda_s - 2d\lambda^2\lambda_s + 4\lambda^3\lambda_s + d^2\lambda_s^2 + 2d\lambda\lambda_s^2)]/[d^2(\lambda_s - \lambda)^3]\} \quad (28)$$

and

$$D = \frac{2\lambda_s(\lambda + \lambda_s - d)}{d^2} \quad (29)$$

is obtained for the case $\lambda_s \neq \lambda$.

In reality since $d/\lambda > 10$, expressions including the term $\exp(-d/\lambda)$ may be disregarded so that the above expressions may be simplified further. Table 1 lists the main results of the theory in their simplified form.

5. Results

5.1. Conditions found in a typical glow discharge

The type of DC discharge used in analytical techniques such as GDMS is an 'abnormal' discharge. This regime occurs when the whole of the cathode surface is covered by the discharge and there is a direct relationship between the discharge voltage and current. These discharges tend to operate in the pressure region 10^{-2} to 10^2 Torr. Typical working conditions found in our glow discharge ion source are pressures between 0.5 and 1 Torr, discharge voltage of 800 V and currents between 0.4 and 1 mA. It must be stressed that these parameters all depend on the geometry, dimensions and materials of any particular ion source.

The extent of the CDS is inversely proportional to pressure (Paschens law [18]). For a pressure in the region of 0.084 Torr (for a planar diode system) Westwood and Boynton [19] reported values of d in the region of 15–20 mm. However, Davis and Vanderslice [6] reported a value of $d = 13$ mm for a pressure of only 0.064 Torr. Clearly, the extent of the cathode fall is rather difficult to predict and will be influenced by the

cell geometry, discharge gas and to a certain extent the discharge voltage [1].

From the above data, Paschens law ($pd = \text{constant}$) implies that $d = 1\text{--}2$ mm at 1 Torr. In our system [20], under the conditions outlined, but using a pin-shaped cathode, the current–surface area relationship was consistent with d values in the region of 1–2 mm. Discharge currents in the region of 1 mA lead to considerable sputter heating of the cathode and temperatures at the cathode surface at 1 mA were in the region of 500–600 K, whilst the bulk gas reached temperatures in the range 340–400 K. There is therefore a decreasing temperature gradient towards the negative glow and hence an increasing gas density. However, as shown in the discussion below, events registering at the cathode occur mostly within the last few mean free paths and the calculated parameters turn out to be largely independent of pressure.

Therefore, in the example calculations below the following reference conditions are used: $p = 1$ Torr, T (gas temperature in the dark space region) = 450 K, $d = 0.2$ cm and V (discharge voltage) = 800 V.

5.2. Comparisons of q and q_s

Symmetrical charge transfer cross sections are relatively easy to measure and are therefore well known [10]. However, the process by which fast neutral atoms are collisionally thermalized is much more difficult to study. Obviously, q_s is related to the total scattering cross section. However since, on average, only half of the energy is lost in each collision, there is a persistence of momentum towards the cathode, and presumably it will take several collisions before the velocity of the fast atom is reduced to the random velocity of the bulk gas. There appear to be no data available on these ‘stopping’ cross sections. Cramer and Simons [21] measured total scattering and charge transfer cross sections of singly charged argon ions as a function of energy, assuming the difference is due to elastic scattering. Based on this, charge transfer to ion elastic scattering ratios in the region of ≥ 1.3 were obtained (however, even this is somewhat unreliable as the total scattering cross sections reported are similar to subsequent charge transfer cross sections alone as measured by other workers [1, 10]). Aberth and Lorents [22] measured differential elastic scattering cross sections and recorded values of $< 10^{-16}$ cm²/sterad for 30° scattering at 30 eV. For high-energy neutral argon collisions the average stopping cross section must be less than the ‘gas kinetic’ collision cross section (pertaining to thermal energy collisions). This has a calculated value of 2.6×10^{-15} cm² (the molecular diameter of Ar being taken as 2.9 Å quoted based on gas phase properties of Ar in [23]). We therefore take this as the upper limiting value for q_s .

The directly measured values of q (charge transfer) in beam experiments at collision energies in the range 50 and 100 eV are between 4 and 5×10^{-15} cm². However, from their glow discharge experiments, Davis and Vanderslice derive a value of 5.5×10^{-15} cm² for

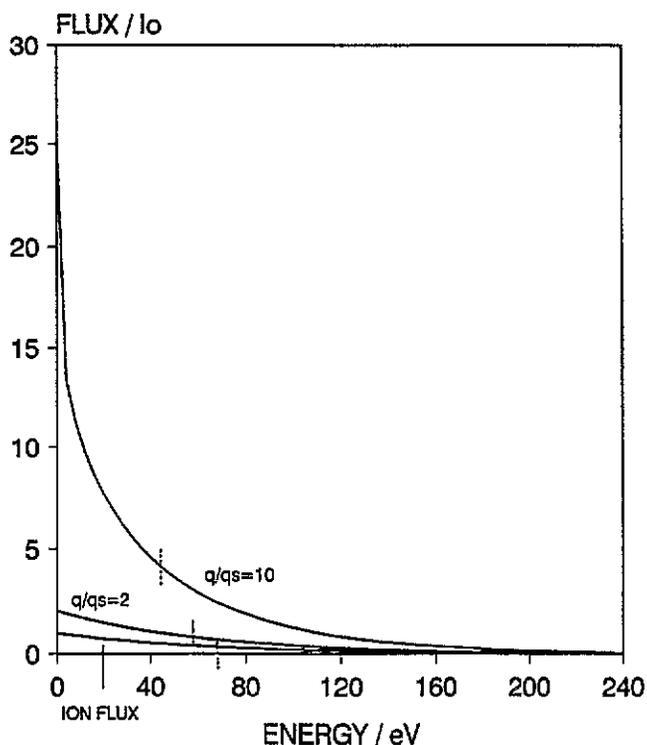


Figure 2. Variation of relative fast particle flux with particle energy at the cathode. Vertical dotted lines indicate values of respective energy averages.

q , the charge transfer cross section, and this will be the value used in the calculations below.

We find therefore that the lower limit of the ratio q/q_s is two. In reality, this ratio is probably much higher so results are obtained for a range of q/q_s ratios.

5.3. Calculations

The ion and neutral energy distribution functions calculated using the reference conditions discussed above are shown in figure 2. Average energies are indicated by the vertical dotted lines crossing the respective curves. Due to the intractable integral in equation (19) the fast neutral curves were obtained by numerical integration. Analysis of the additional terms for the case where $l = d$ shows that they have a negligible effect on the total neutral flux and average energy and in fact all results are calculated using the simplified expressions found in table 1.

6. Discussion and conclusions

The major finding of this work is that the flux of fast neutral atoms is dependent on the ratio between the mean free paths for symmetrical charge transfer and collisional ‘stopping’. In argon then, if λ_s is taken as the gas kinetic cross section then the fast neutral flux is at least twice that of the ions, but could be significantly greater. The variation of flux with the ratio q/q_s is shown in figure 3 to increase almost linearly. In contrast, the average energy of the neutrals is slightly less than that of the ions and indeed decreases with increasing q/q_s ratio (see

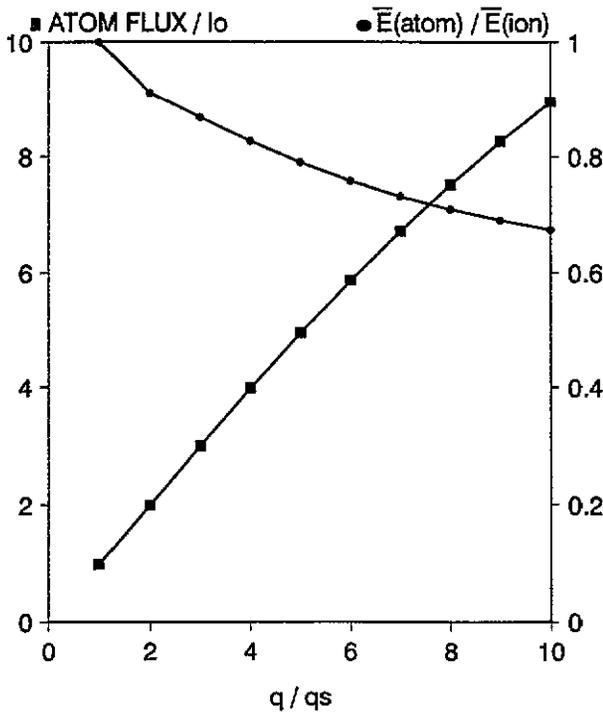


Figure 3. Variation of relative total neutral flux F^N/I_0 with q/q_s , compared with variations in the relative fast atom energy $\bar{E}(\text{atom})$.

figure 3), although the rate of decrease is not as steep as the rate of increase in flux.

We find this result somewhat surprising since if $\lambda_s > \lambda$, one would expect a greater proportion of high-energy neutrals to survive to the cathode, thereby leading to a higher average energy. This would indeed be the case if the accelerating potential in the cathode fall region was linear. In fact it decreases towards the negative glow region (where it is assumed to be zero). The majority of ions which reach the cathode are formed in a region directly adjacent to the cathode where the potential gradient (and hence the energy gained) is greatest. However, the bulk of the atoms must be formed from ions in regions further back from the cathode where the energy gained from the field is less.

Despite this, because the flux of fast neutrals in the glow discharge is likely to be significantly greater than that of the ions, with similar though slightly lower average energy, the greater proportion of cathode erosion in the glow discharge is probably caused by neutrals and not ions.

In addition, we find the average energy of the ions is a simple expression being directly dependent (in argon at least) on the ratio λ/d . Since d is inversely proportional to pressure, the average energy should be largely independent of pressure, as found experimentally. The same is also true of the neutral flux and energy. Using the conditions stated earlier for a typical 'abnormal' glow discharge we find for $q/q_s = 2$: the average ion energy $\bar{E}_{\text{ions}} = 68$ eV, the total neutral flux $F^N = 2 \times I_0$ and the average neutral energy $\bar{E}_N = 59$ eV. Taking the upper limit of $q/q_s = 10$ gives: $\bar{E}_{\text{ions}} = 68$ eV, $F^N = 9 \times I_0$ and $\bar{E}_N = 44$ eV.

Since the approach we have adopted in this work is

somewhat different to that of other authors, it is rather difficult to make any direct quantitative comparisons of results. Nevertheless, extracting data from the work of Kuchinskii and Sheikin [15] for similar values of the CDS their model gave an atom/ion flux ratio of 12 for $q/q_s = 10$ and three for $q/q_s = 2.5$, which compares roughly with the results of our model given above. Here, Green's function is used to solve a somewhat more complicated distribution than we assume in our model. No information on the average energy of the fast atoms was available for comparison. The form of the distribution function presented by Wronskii in *Vacuum* [13a] is similar to that of our model. However, results differ at the high-energy end of the spectrum where there is a crossover, the flux of neutrals being less than that of the ions. However, a different (theoretical) cathode fall function is used whilst in this work we restrict ourselves to the empirically derived function [18].

A number of assumptions and approximations have been made in order to make the problem analytically tractable. First, a planar geometry is assumed which is perfectly reasonable although there will be some effects due to the changing space charge distribution at the cathode edge. Secondly, a linear field variation is assumed and, as discussed earlier, in the light of the measurements by Den-Hartog *et al* [12] this appears to be a valid assumption. The assumption that q and q_s are constants seems to be fair since where experimental values are given, the variation is not more than 30% over the energy range of interest. Incorporating this variation into the model would result in a slight broadening of the energy distributions but would not seriously effect the average energy. Similarly, the assumption that neutrals are either completely 'stopped' or continue on towards the cathode unaffected is not strictly valid. In reality there will be a collisional broadening of the neutral energy distribution towards lower energies and increased flux levels. However, the inclusion of an average 'stopping' cross section may account for this effect. Despite the findings of this work, the rate of erosion of the cathode (consisting of a copper pin) as measured in our laboratory (see, for example Mason *et al* [24]) cannot be fully explained by this model. We would expect to find a fast neutral flux of some $100 \times I_0$ in order to account for the observed erosion rate. The averaging process used to calculate the mean particle energy (equation (23)), tends to shift the average energies to low values. However, the expression used is such that we may regard the full flux as possessing the average energy. By computing the average energy numerically, we have found that there is a 20 eV increase if only those particles with energies above threshold (taken as 15 eV) for sputtering are included in the expression. However, the total flux of these fast atoms is found to be significantly lower so that there appears to be no net effect on the erosion rate. Quantitative comparisons between measured and theoretical sputtering rates are complicated by the fact that in the abnormal glow discharge there is a considerable degree of back diffusion of sputtered material. Hence a model is required for this

process which may be coupled to that for the atom/ion distribution functions. The atom flux is itself impossible to measure and can only be inferred. However, in a recent set of experiments [24] we have been able to measure the average energy of the neutral particles (based on sputtering threshold measurements). In argon, atoms have an energy equivalent to $\sim 6\%$ of the discharge voltage which is not far from the theoretical value presented in this work. Based on this and using a simplified diffusion model leads us to an estimate of the atom/ion flux ratio in the region 10–100. However, more work is required on the diffusion model before a good estimate of the flux ratio is obtained. We are also presently working on a modified version of the theory in which fast neutrals are thought to be produced by some mechanism other than symmetrical charge transfer.

References

- [1] Chapman B 1980 *Glow Discharge Processes* (New York: Wiley)
- [2] King F L and Harrison W W 1990 *Mass Spectrom. Rev.* **9** 285
- [3] Greene J E 1978 *J. Vac. Sci. Technol.* **15** 1718
- [4] Gruen D M, Pellin M J, Young C E and Calway W F 1986 *J. Vac. Sci. Technol. A* **4** 1779
- [5] Doughty D A, Den Hartog E A and Lawler J E 1987 *Phys. Rev. Lett.* **58** 2668
- [6] Davis W D and Vanderslice T A 1963 *Proc. R. Soc. A* **137** 662
- [7] Rickards J 1984 *Vacuum* **34** 559
- [8] Abril I, Gras-Marti A and Valles-Abarca J A 1983 *Phys. Rev. A* **28** 3677
- [9] Abril I, Gras-Marti A and Valles-Abarca J A 1984 *J. Phys. D: Appl. Phys.* **17** 1841
- [10] Massey H S W and Gilbody H B 1974 *Electronic and Ionic Impact Phenomena* vol 4 (Oxford: Oxford University Press) ch 23
- [11] Gras-Marti A, Abril I and Valles-Abarca J A 1985 *Thin Solid Films* **124** 59
- [12] Den-Hartog E A, Doughty D A and Lawler J E 1988 *Phys. Rev. A* **38** 2471
- [13] Wronskii Z, Sullivan J L and Saied S O 1992 *J. Phys. D: Appl. Phys.* **25** 1607
Wronskii Z 1986 *Vacuum* **36** 329
- [14] Mase H and Tanabe T 1983 *Proc. Int. Ion Engineering Congress 1983 ISIAAT '83 & IPAT '83 (Kyoto, Japan) 1983* p 311
- [15] Kuchinskii V V and Sheikin E G 1988 *Sov. Phys. J.* **30** 697
- [16] Donko Z and Janossy M 1992 *J. Phys. D: Appl. Phys.* **25** 1323
- [17] Peter S, Pintaske R, Hecht G and Richter F 1993 *J. Nucl. Mater.* **200** 412
- [18] Llewellyn-Jones F 1966 *The Glow Discharge* (London: Methuen)
- [19] Westwood W D and Boynton R J 1972 *J. Appl. Phys.* **43** 2691
- [20] Milton D 1991 *PhD Thesis* University College of Swansea, UK
- [21] Cramer W H and Simons J H 1957 *J. Chem. Phys.* **43** 1272
- [22] Aberth W and Lorents D C 1966 *Phys. Rev.* **144** 109
- [23] Lide D R (ed) 1992–1993 *CRC Handbook of Chemistry and Physics* (Boca Raton, FL: CRC)
- [24] Mason R S, Milton D M P, Pichilingi M, Anderson P D J and Fernandez M T 1994 *Rap. Commun. Mass Spec.* **8** 187