Effusion of ions through small holes

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The effusion of ions through small holes with cylindrical geometry, mady in conductive materials, has been theoretically studied, assuming that each ion striking the wall is converted into a neutral particle. The results of the calculations are compared with some preliminary experimental results.

1. Introduction

Sampling ions from plasmas, for mass spectrometric investigations has become a widely applied technique, such as in gas discharge studies[1,2] and experiments of aeronomic interest.[3] In most of these experiments the ions are withdrawn through an orifice, the dimensions of which are small compared with the mean free path of the particles. In this case it is generally agreed that the formulas of molecular flow may be applied. Hence the number of ions effusing through the sampling orifice per unit time is given by Meyer's relation:

$$N = \tfrac{1}{4} n_0 \langle u \rangle S \tag{1}$$

where n_0 is the ion density, $\langle u \rangle$ the arithmetical average velocity of the charged particles and S the area of the leak hole. By applying this formula, the derivation of which can be found in any textbook on kinetic theory of gases,[4] one tacitly assumes that the sampling holes are made in a wall with negligible thickness. In reality however these holes must be considered as small tubes, which are usually made by perforating a thin metal plate. Thus a loss of ions will occur due to the discharging effects on the inner walls of these tubes.

In the present work a theoretical study is made of the ion effusion through small holes, taking into account this loss of charged particles.

2. Outline of the treatment

The calculation of the ion effusive flow through an orifice with diameter $2R$ in a wall with thickness h will be made under the assumption that each ion striking the wall is lost. This means that it will be neutralised on the surface and reappear in the gas as a neutral particle. This assumption is reasonable if the ion energy is low and no secondary effects take place, such as ion reflection or emission of secondary electrons, which give rise to secondary ionisation or ion–electron recombination. It is obvious that, provided the mean free path is larger than the distance between inlet and outlet of the orifice, the calculations will also be valid for the effusion of neutral particles through two identical orifices (with infinitely small thickness) placed one after the other, if the intermediate space is evacuated with infinitely high pumping speed.

Under the assumptions mentioned above and referring to Figure 1, the number of ions effusing through the infinitesimal surface area $\mathrm{d}S$ is given by the integral:

$$\mathrm{d}v = \int_{u=0}^{\infty}\int_{\theta=0}^{\theta_m}\int_{\phi=0}^{2\pi} f(u) u \cos\theta \,\mathrm{d}\omega \tag{2}$$

$$f(u) = n_0 \left[\frac{m}{2\pi kT}\right]^{3/2} \exp\left(-\frac{mu^2}{2kT}\right) \tag{3}$$

is the distribution function for ionic velocities, and

$$f(u)\mathrm{d}\omega = u^2 \mathrm{d}u \sin\theta \,\mathrm{d}\theta \,\mathrm{d}\phi\, f(u) \tag{4}$$

represents the fraction of ions that are moving with speeds between u and $u + \mathrm{d}u$ and in a direction, which makes an angle between θ and $\theta + \mathrm{d}\theta$ with the polar axis and in a plane through the axis making an angle between ϕ and $\phi + \mathrm{d}\phi$ with the reference plane for ϕ.

The total effusing number through the tube can now be obtained after integration over r and β

$$N = \int_{r=0}^{R}\int_{\beta=0}^{2\pi} \mathrm{d}v \, r\mathrm{d}r \,\mathrm{d}\beta \tag{5}$$

In the foregoing derivation it has been assumed that the ion gas is at rest as a whole. If however mass motion is occurring the distribution function must be replaced by:

$$f(u) = n_0 \left[\frac{m}{2\pi kT}\right]^{3/2} \exp -\left[\frac{m}{2kT}(\mathbf{u} - \mathbf{w})^2\right] \tag{6}$$

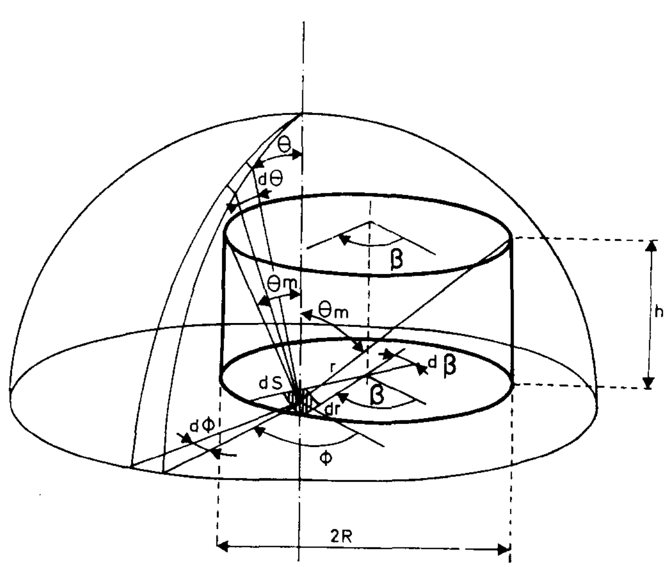

Figure 1. Co-ordinate system explaining the different symbols used throughout the calculations.

where **w** represents the 'ensemble' velocity of the ion gas. Two distinct cases will now be treated:

(1) The ion velocity distribution is isotropic, in other words no mass motion of the gas is taking place.

(2) Mass motion of ions is occurring towards the orifice, which means that the direction of **w** is perpendicular to the base plane of the tube. This case is included because of its possible experimental applications.

3. Isotropic ion velocity distribution

In this case **w** equals zero and the total number of effusing ions is given by:

$$N = n_0\left[\frac{m}{2\pi kT}\right]^{3/2}\int_0^\infty \exp\left(-\frac{mu^2}{2kT}\right)u^3\mathrm{d}u \times$$
$$\int_0^R\int_0^{2\pi}\int_0^{2\pi}\int_0^{\theta_m} \sin\theta\cos\theta\, r\mathrm{d}r\,\mathrm{d}\phi\,\mathrm{d}\theta\,\mathrm{d}\beta \qquad (7)$$

where θ_m is defined by:

$$\cos^2\theta_m = \frac{h^2}{h^2+R^2+r^2(\cos^2\phi-\sin^2\phi)-2r\cos\phi\,(R^2-r^2\sin^2\phi)^{1/2}}. \qquad (8)$$

As can be seen the foregoing integral reduces to Meyer's equation for $\theta_m = \pi/2$. Taking into account that the average velocity is given by

$$\langle u\rangle = \left[\frac{8kT}{\pi m}\right]^{1/2} \qquad (9)$$

equation (7) reduces to

$$N = \frac{1}{4\pi}n_0\langle u\rangle\int_0^R\int_0^{2\pi}\int_0^{2\pi}\int_0^{\theta_m} \sin\theta\cos\theta\, r\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}\beta \quad (10)$$

or after integration over θ

$$N = \frac{n_0\langle u\rangle}{4}\,[\pi R^2 - I_1 h^2] \qquad (11)$$

with

$$I_1 = \int_0^R\int_0^{2\pi}\frac{r\mathrm{d}r\,\mathrm{d}\phi}{h^2+R^2+r^2(\cos^2\phi-\sin^2\phi)-2r\cos\phi(R^2-r^2\sin^2\phi)^{1/2}}. \qquad (12)$$

Again this can be integrated over r thus giving rise to

$$I_1 = \int_0^{2\pi}\frac{\cos\phi}{v}\times \frac{1+2v^2\sin^2\phi}{(1+v^2\sin^2\phi)^{1/2}}\operatorname{arctg}\frac{v\sin^2\phi}{\cos\phi(1+v^2\sin^2\phi)^{1/2}}\,\mathrm{d}\phi$$
$$+\int_0^{2\pi}\frac{\cos\phi}{v}\times \frac{1+2v^2\sin^2\phi}{(1+v^2\sin^2\phi)^{1/2}}\operatorname{arctg}\frac{\cos\phi}{v(1+v^2\sin^2\phi)^{1/2}}\,\mathrm{d}\phi \qquad (13)$$

with $v = h/R$. (14)

Both new integrals can be evaluated by partial integration if we take into account that:

$$\frac{\cos\phi}{v}\,\frac{1+2v^2\sin^2\phi}{(1+v^2\sin^2\phi)^{1/2}} = D\{\tfrac{1}{4}\sin\phi(1+v^2\sin^2\phi)^{1/2}\} \qquad (15)$$

Hence it follows that

$$I_1 = 2\pi\left\{\left(\frac{1+v^2}{v^2}\right)^{1/2} - 1\right\} \qquad (16)$$

inserting this in equation (11) leads to

$$N = N_0\pi R^2\,\frac{(1+v^2)^{1/2}-v}{(1+v^2)^{1/2}+v} \qquad (18)$$

where

$$N_0 = \frac{n_0\langle u\rangle}{4}$$

which expresses the ion flow rate through a metal tube with diameter $2R$ and length h.

4. Ionic mass motion in the direction of the tube axis

Here formula (7) must be replaced by:

$$N = n_0\left[\frac{m}{2\pi kT}\right]^{3/2}\int_0^\infty \exp\left(-\frac{m}{2kT}(\mathbf{u}-\mathbf{w})^2\right)u^3\mathrm{d}u$$
$$\int_0^R\int_0^{2\pi}\int_0^{2\pi}\int_0^{\theta_m} \sin\theta\cos\theta\, r\mathrm{d}r\,\mathrm{d}\phi\,\mathrm{d}\theta\,\mathrm{d}\beta \qquad (19)$$

where **w** is the mass motion of 'ensemble' velocity, the direction of which is parallel to the axis of the cylindrical holes. Hence

$$(\mathbf{u}-\mathbf{w})^2 = u^2 + w^2 - 2uw\cos\theta. \qquad (20)$$

First we will evaluate the integral

$$I_2 = n_0\left[\frac{m}{2\pi kT}\right]^{3/2}\int_0^\infty \exp\left[-\frac{m}{2kT}(u^2+w^2-2uw\cos\theta)\right]u^3\mathrm{d}u$$
$$= n_0\left[\frac{m}{2\pi kT}\right]^{3/2}\exp\left(-\frac{mw^2}{2kT}\right)I_3$$

with

$$I_3 = \int_0^\infty \exp\left[-\frac{mu^2}{2kT}-\frac{mw}{kT}u\cos\theta\right]u^3\,\mathrm{d}u.$$

By straightforward integration this can be reduced to:

$$I_3 = \frac{2k^2T^2}{m^2} + \frac{kT}{m}w^2\cos^2\theta$$
$$+\left(\frac{2kT}{m}\right)^{1/2}\left[w\cos\theta\left(\frac{3kT}{m}+w^2\cos^2\theta\right)\right]\times$$
$$\exp\left(\frac{mw^2}{2kT}\cos^2\theta\right).$$

Inserting this into equation (19) and integration over β gives

$$N = 2\pi n_0 \left[\frac{m}{2\pi kT}\right]^{3/2} \exp\left(-\frac{mw^2}{2kT}\right) \int_0^R \int_0^{2\pi}$$

$$\left\{\frac{2k^2T^2}{m^2} I_5 + \frac{kT}{m} w^2 I_6 + 3w\frac{kT}{m}\left(\frac{2\pi kT}{m}\right)^{1/2} I_7 + w^3\left(\frac{2\pi kT}{m}\right)^{1/2} I_8\right\} r\mathrm{d}r\, \mathrm{d}\phi \qquad (21)$$

with

$$I_5 = \int_0^{\theta_m} \sin\theta \cos\theta\, \mathrm{d}\theta = \tfrac{1}{2}(1 - \cos^2\theta_m)$$

(note that this integral has already been found in Section 3)

$$I_6 = \int_0^{\theta_m} \cos^3\theta \sin\theta\, \mathrm{d}\theta = \tfrac{1}{4}(1 - \cos^4\theta_m)$$

$$I_7 = \int_0^{\theta_m} \exp\left(\frac{mw^2}{2kT} . \cos^2\theta\right) \cos^2\theta \sin^2\theta\, \mathrm{d}\theta$$

$$I_8 = \int_0^{\theta_m} \exp\left[\frac{mw^2}{2kT}\right] \cos^2\theta \cos^4\theta \sin\theta\, \mathrm{d}\theta.$$

After rearrangement and putting

$$\alpha = \frac{mw^2}{2kT}$$

equation (21) can be written as:

$$N = N_1 + N_2 + N_3 \qquad (22)$$

with

$$N_1 = N_0 \pi R^2 \frac{(1+v^2)^{1/2} - v}{(1+v^2)^{1/2} + v} \exp(-\alpha) \qquad (23)$$

$$N_2 = N_0 \frac{\alpha}{2} \exp(-\alpha)[\pi R^2 - \int_0^{2\pi} \int_0^R \cos^4\theta_m\, r\mathrm{d}r\, \mathrm{d}\phi] \qquad (24)$$

$$N_3 = 2N_0 R^2 (\alpha\pi)^{1/2} (\pi - P) \qquad (25)$$

P being defined as

$$P = \int_0^{2\pi} \int_0^1 \frac{v^3 \exp(-\alpha \sin^2\theta_m)\, t\mathrm{d}t\, \mathrm{d}\phi}{[1 + v^2 + t^2(\cos^2\phi - \sin^2\phi) - 2t\cos\phi(1 - t^2\sin^2\phi)^{1/2}]^{3/2}} \qquad (26)$$

with $v = h/R$ and $t = r/R$.

Again N_2 can be calculated by straightforward integration. Thus it is found:

$$N_2 = \frac{\alpha}{2} N_0 \exp(-\alpha) \frac{\pi R^2}{(1+v^2)^{1/2}}. \qquad (27)$$

The double integral P defined by equation (26) has been numerically calculated for different values of v and as a function of α. The results of this treatment are shown in Figure 2.

Finally the ion flow rate F has been calculated by means of formulas (22), (23), (27) and the numerical calculations of the double integral P. The results of this treatment are summarised in Figure 3. The number of ions N, effusing per unit time through the cylinder is related to the flow rate by:

$$N = N_0 \pi R^2 F$$

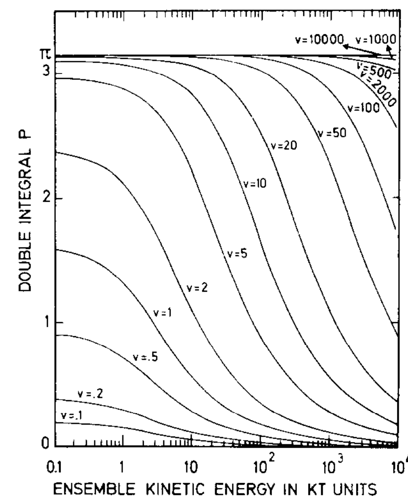

Figure 2. Dependence of the double integral P on the ensemble kinetic energy for different values of the height/radius (v) ratio of the holes.

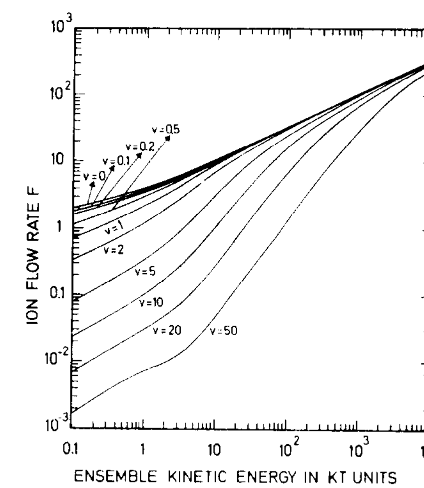

Figure 3. Ion flow rate versus the ensemble kinetic energy. Again the parameter is the height/radius ratio.

5. Experimental verification

At our institute a mass spectrometer for the measurement of the ion composition in the stratosphere has been designed. For this purpose a special molecular leak[5] has been constructed, which consists of a platinum sheet of 12 μm thickness, perforated by means of a ruby laser. In this way 800 holes with an average diameter of 12 μm were obtained in a quadrangular area of 9 mm^2. Behind this membrane an ion lens is mounted, which focusses the charged particles in a quadrupole mass filter. The mass filter section is pumped down by a cryopump to a pressure of about 10^{-5} torr. This experimental set up is shown in Figure 4. In a first attempt to determine the foccusing potentials of

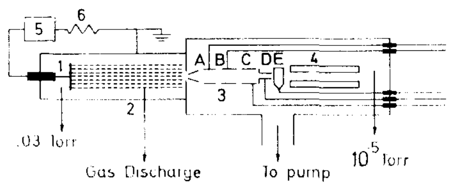

Figure 4. Schematic representation of the experimental set up. 1. Cathode; 2: metal container; 3: ion lens (A, B, C, D, and E: electrodes); 4: quadrupole; 5: high voltage supply; 6: ballast resistor.

the ion lens a gas discharge has been generated at one side of the molecular leak in an airlike mixture at a pressure of approximately 0.03 torr, as an ion source for the mass spectrometer system.

If the ion lens were replaced by a moving disc electrode, with a diameter $2a$ and which could be placed at a variable distance d from the sampling orifice, it would be possible to measure the ion current to this disc for different values of d. Since this disc would be in a high vacuum region, where the mean free path of the ions is very large it follows that, as can be seen from Figure 5 the number of ions reaching a disc electrode at a distance d from the sheet is the same as the number effusing through a cylinder with diameter $2R$ and length L. This length L is given by:

$$L = \frac{2(d+h)\,.\,R}{R+a}.$$

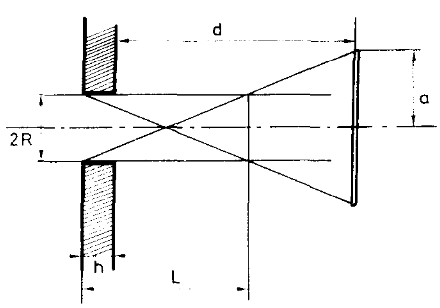

Figure 5. Geometrical arrangement showing the disc electrode replacing the ion lens.

The same result is obtained by measuring the ion current on the different electrodes of the ion lens. Defining the ion transmission rate T by:

$$T(v') = \frac{N(v')}{N(h/R)}$$

where $v' = L/R$ and $N(h/R)$ is the total number of ions effusing through the sampling orifice, T has been calculated with the foregoing theory for $h/R = 2$ and for different values of α. The result of this has been plotted in Figure 6. Some typical values of the ion current obtained on the different ion lens elements are shown in Table 1. The distances defined in Figure 5 are, in our experimental set-up, given by:

$$2R = h = 12\ \mu m$$

and

$$a = 17\ mm.$$

Therefore $v' = L/R = 2(d+h)/(R+a) \approx 2d/a$.

Table 1. Ion transmission rate as measured on the ion lens elements

Lens elements connected to the electrometer	Measured ion current	Corresponding value of d (mm)	v'	$T(v')$
A + B + C + D + E	1.16 nA			1
B + C + D + E	995 pA	41	4.82	0.82
C + D + E	720 pA	65	7.65	0.62
D + E	565 pA	89	10.47	0.49

The values of T obtained by these measurements are shown by the crosses on Figure 6. As can be seen the experimental results can be explained by assuming that the ions, reaching the platinum sheet, have an energy of approximately 85 kT.

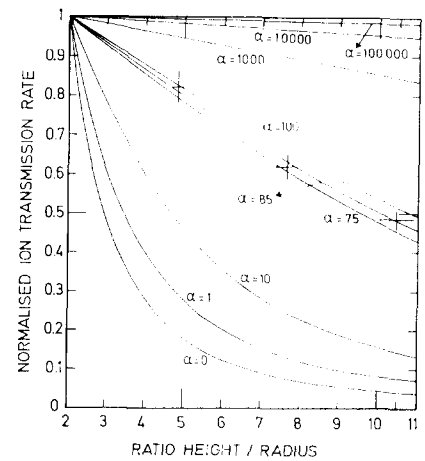

Figure 6. Experimental values of the normalised ion transmission rate shown together with the theoretical curves for various values of the parameter α defined in the text.

It must be emphasized that the experimental results shown here are only preliminary and that they are merely shown to indicate the possibility of verifying the calculations. In principle it would be possible to determine the ion energy from more detailed measurements.

6. Conclusions

A theory has been developed to describe the effusion of ions through cylindrical holes in conductive materials. The theory has been reinforced with some preliminary experimental results. It has been shown that Mayer's relation

$$N = \tfrac{1}{4} n_0 \langle u \rangle S$$

must be replaced by

$$N = \frac{n\langle u\rangle}{4}\,\frac{(1+v^2)^{1/2} - v}{(1+v^2)^{1/2} + v}\,.\,S$$

to take into account the loss of ions by discharging effects on the inner walls of the cylindrical holes, if the ion velocity is isotropic.

If however mass motion of ions is occurring towards the sampling orifice a numerical treatment is necessary to describe the effusion.

The treatment is directly related to all experiments where ions are sampled through cylindrical holes in metal plates, provided the mean free path is larger than the dimensions of the holes. The analysis especially applies to results obtained with drift tubes coupled to a mass spectrometer[3] and to satellite experiments[6] where the relative motion between plasma and space vehicle are important.

Acknowledgements

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