REVIEW ARTICLE

Knudsen flow 75 years on: the current state of the art for flow of rarefied gases in tubes and systems

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Knudsen flow 75 years on: the current state of the art for flow of rarefied gases in tubes and systems

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Abstract

Following a brief historical introduction an overview is given relating the most recent studies of rarefied gas flow to the early work of Knudsen. The first paper submitted in October 1908 (published in 1909) initiated a period of intense activity by Knudsen, Smoluchowski and, a little later, by Gaede and Langmuir. This also covered the transition to the already well established hydrodynamic flow expressed in terms of the ratio of mean free path to critical apparatus dimension—which we now refer to as the Knudsen number. The desorption, evaporation and scattering of molecules from surfaces was described in terms of the Knudsen cosine law of scattering. The Knudsen effusion method for determining vapour pressure, also introduced in 1909, has become the main tool for studies of the related problem of the dissociation, chemical bonding and the vaporisation process itself. Clausing developed, as an alternative to conductances, the concept of transmission probability, still referred to as the Clausing factor, and provided a procedure for their more accurate evaluation in long and short tubes. A number of misconceptions of these early efforts have found their way into the literature and current books on vacuum science and technology. However, detailed studies have clarified the problem of gas-surface interactions; the gas flow in tubes has been tackled with Clausing-type integral equations and by statistical computation techniques based on Monte Carlo analysis procedures adaptable to more complex systems. Results have been confirmed experimentally using molecular-impact pressure probe measuring techniques.

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Contents

1. Introduction

Martin Knudsen published three papers in 1909 in which he explained theoretically and demonstrated experimentally for the first time our understanding of the flow of gases at low pressure in terms of molecular flow. These, together with contributions in the immediately following years, provided the stimulation for responses from Smoluchowski, Dunoyer, Gaede, Langmuir and Dushman. Between them they set the scene for vacuum science and technology as well as laying the base for what are now well developed disciplines and quite separate subjects. Their common origin is Knudsen's early work, to be remembered in this very brief historical review.

Before discussing the particular problems of Knudsen flow we should, therefore, remind ourselves of the broader impact of these early publications on related work as indicated in table 1.

2. Molecular flow

In his first paper, Knudsen (1909a) introduced the important criterion for molecular flow in terms of mean free path in relation to tube dimensions. He also discussed the transition to the already well established hydrodynamical or Poiseuille flow (see § 6 below) expressed in terms of the ratio of mean free path to critical apparatus dimension—which we now refer to as the Knudsen number (Kn).

The concept of mean free path had already been introduced by Clausius (1858). It stimulated a critical and more rigorous treatment by Maxwell (1860), taking the Maxwell distribution of velocities into account, and served as a basis for his theory of transport processes. Other properties of gases, such as conduction of heat, frictional (viscous) forces and interdiffusion are also usefully expressed in terms of mean free paths. This was followed by further important theoretical contributions by Maxwell, Boltzmann, Chapman and Enskog. A useful discussion of the historical developments, including a reproduction of many of the original papers, was given by Brush (1965/6), and much of it is now a standard ingredient of books on the kinetic theory of gases, such as Jeans (1925), Loeb (1934), Present (1958) and Kennard (1938). The Knudsen number $\mathrm{Kn} = \lambda / d$ is, of course, basic to the more recent studies of rarefied gas flow in aerodynamics as shown, for example, in the useful books by Patterson (1956) and Schaaf and Chambre (1961).

Direct collision-free streams of molecules under conditions of large mean free paths were the subject of experiments devised by Dunoyer (1911) who was stimulated by Knudsen's first paper (as noted in his lecture (Dunoyer 1912; see Dunoyer 1926 p 32), see also Dunoyer 1926). This, in turn, eventually led to the development of molecular beam techniques (as outlined, for example, by Fraser (1937) and Smith (1955) and many more detailed reviews). There was a comment a few years later in a paper by Langmuir (1916a) drawing attention to the historical development and pointing out that the rectilinear propagation of molecules *in vacuo* had been long familiar to those concerned with the manufacture of incandescent filament lamps and referring to experiments by Anthony (1894). However, as pointed out by Holland (1962) in his

Table 1. Publications from 1909 of Knudsen and contemporary workers in related fields showing developments arising from them.

Knudsen (1909a)		
	Molecular flow condition in terms of mean free path λ.	Mean free path from Clausius (1858) and Maxwell (1860). Refers to Kundt and Warburg (1875) experiments. Knudsen number $Kn = \lambda / d$.
	Linear propagation of molecules when $Kn \gg 1$.	Anticipated by Fleming (1883) in shadow effect due to evaporation in filament electric lamps. Stimulated Dunoyer (1911), first molecular beam techniques—confirmed by Stern (1920).
	Scattering at surfaces by cosine law of diffuse reflection.	Analogy between gas transport and thermal radiation. Cosine law in relation to kinetic theory. Further experiments by Wood (1915), Knudsen (1916) and Langmuir (1916a).
	Molecular flow in long cylindrical tubes of circular and arbitrary cross section.	Corrections by Smoluchowski (1910) for other than circular cross section—gives solution for rectangular section.
	Flow resistance concept.	Gas flow analogy with electrical circuits.
	Transition from molecular to viscous Poiseuille flow.	Knudsen 'minimum' in pressure against conductance plot. Stimulated Gaede (1913a) and much recent work.
Knudsen (1909b)		
	Orifice flow and effusion molecular impact rate $\frac{1}{4} n\bar{v}$ per unit area.	Flow resistance of orifice suggest use for temperature and/or molecular weight determination of vapour.
Knudsen (1909c)		
	Effusion method of vapour pressure measurement.	Determines vapour pressure of Hg. Relates to Hertz (1882). Further studies by Knudsen (1915b) on maximum evaporation rate of Hg.
Knudsen (1910a, b, d)		
	Thermal molecular flow.	
Smoluchowski (1910)		
	Transpiration and diffusion.	Critical comment on Knudsen (1909a, b) correction for long tube flow, for arbitrary cross section—important formula used by Clausing (1929, 1930a, b, c, 1932).
Knudsen (1910c)		
	An absolute manometer.	Knudsen gauge—related to radiometers. Smoluchowski (1911a) comments on theory; see Steckelmacher (1951) for history and bibliography.

Table 1. (continued)

Dunoyer (1911)		
	Molecular beam.	Molecular beam experiments, orifice (see also Dunoyer (1960)) method.
Knudsen (1911)		
	Molecular heat conduction. Accommodation coefficient (thermal).	Smoluchowski (1911b), correspondence with Knudsen (1911a, c, d), several papers. Related to Pirani (1906) vacuum gauge. Thermal accommodation reviewed by Wachman (1962), Thomas (1980) and Goodman (1980).
Langmuir (1912)		
	Thermal conductivity.	
Dunoyer (1912)		
	Appreciation of Knudsen's work in modern ideas on the constitution of matter.	For details see Dunoyer (1926).
Gaede (1913a)		
	Viscous and transition flow.	Critical analysis of Knudsen (1909a) on transition flow, Knudsen minimum, hydrogen tests (from electrolysis), liquid nitrogen trap used.
	Low pressure McLeod gauge. Cold trap error effect due to Hg streaming.	Analysed in more detail by Gaede (1915). Cold trap error effect, rediscovered only in 1961 by Ishii and Nakayama (1962).
	Molecular roughness of surfaces.	To explain gas skin effect at higher pressures, but Knudsen flow at low pressures *not* affected. Misleading statement in Davies *et al* (1964) suggests use of 'Gaede effect'; corrected by Lund and Berman (1966).
Gaede (1913b)		
	Molecular pump.	Later improved to turbomolecular pumps.
Langmuir (1913c)		
	Vacuum gauge, viscosity type.	Uses rotary disc.
Knudsen (1914)		
	Determination of molecular weight of vapours. Viscosity gauge.	
Haber and Kerschbaum (1914)		
	Theory of viscosity gauge.	Quartz fibre type—important appendix by Born and Einstein—interpreted theory. Steckelmacher (1973) gives review and bibliography.
Knudsen (1915a)		
	Molecular gas resistance to moving plate.	Viscosity gauge theory.

Table 1. (continued)

Langmuir (1913a, 1915)	Chemical reactions at low pressures.	
Gaede (1915)	Diffusion of gases through Hg. Diffusion pump.	Patent application 25 September 1913.
Wood (1915)	Law of reflection of gas molecules, experiments with spherical bulb.	Experiments to prove cosine law. Stimulated contact with Knudsen (1916) suggesting improved experiment using wall of sphere (Wood 1916).
Knudsen (1916)	Cosine law in kinetic theory.	Relates to earlier Knudsen (1909) and Wood (1915) experiments and Langmuir (1916a).
Wood (1916)	Condensation and reflection of gas molecules.	Further experiments on cosine law.
Langmuir (1916a)	The evaporation, condensation and reflection of molecules and mechanism of adsorption.	Comments on cosine law and experiments by Wood (1915) also Dunoyer 1911. Instigates investigations on surfaces: surface physics and chemistry.
Langmuir (1916b)	High vacuum mercury pump.	Improved diffusion pump (Gaede 1915).
Langmuir (1916c)	The condensation pump.	Further diffusion pump development.
Smoluchowski (1916b)	Brownian motion and diffusion.	
Knudsen (1917a)	Condensation of metal vapours.	Stimulated by Langmuir (1916a).
Langmuir (1916a, 1917)	Condensation and evaporation, adsorption.	

survey of vacuum deposition processes, the phenomenon of casting a molecular shadow had already been clearly demonstrated by Fleming (1883, 1885) (figure 1), also reproduced in the book by Fleming (1919). Details of Dunoyer's molecular beam experiments (figure 2) were outlined by him in a review at the first Vacuum Congress (Dunoyer 1960) and discussed by Fraser (1937) and Smith (1955). With the availability of improved vacuum techniques from about 1920, Stern and his collaborators started a whole sequence of developments based on the application of molecular beams but this is outside the scope here (reviewed, for example, by Estermann (1946, 1959) and many more recently). The special case of beaming effect from tubes will be discussed below (§ 5). Detailed bibliographies are in Ramsey (1956) and Kusch and Hughes

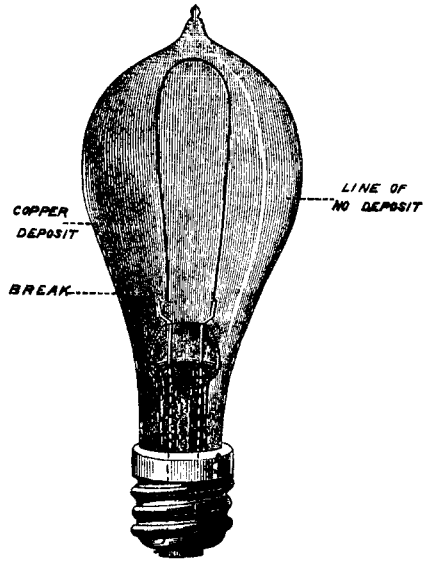

Figure 1. Molecular 'radiation' from Edison incandescent lamp as shown by Fleming. The ends of the horseshoe-shaped carbon filament are held by small copper clamps on the platinum wire leads through the glass. Failure of this joint evaporates a thin copper film over the inside of the bulb; the narrow line free of deposit is a 'shadow' from the filament loop.

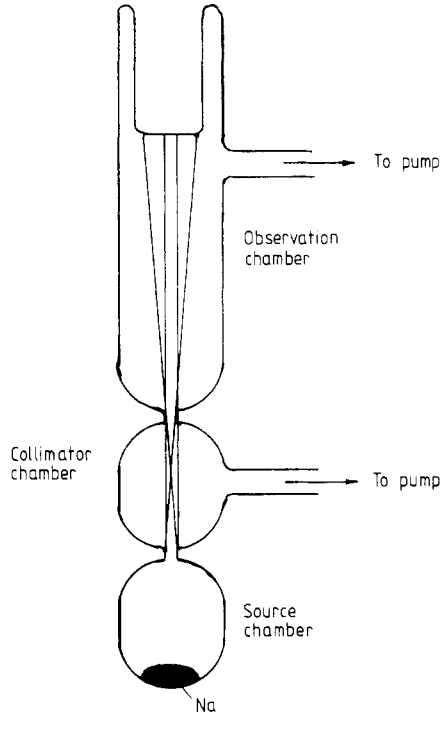

Figure 2. Arrangement used by Dunoyer in his experiment to demonstrate the rectilinear motion of sodium molecules from a source chamber through the evacuated collimator forming a clear deposit under good vacuum conditions.

(1959), supersonic beams in Knuth (1964), general reviews by Leonas (1964, 1979) and Pauli and Toennies (1965), with chemical applications emphasised by Fluendy and Lawley (1973).

3. Gas scattering from surfaces

The mode of scattering of gas molecules from the walls was recognised by Knudsen (1909a) to be a fundamental aspect of his theoretical analysis. He argued that, under free molecular flow conditions, a cosine law of diffuse emission or reflection from the wall surfaces was the most reasonable assumption. We note that in contrast, under conditions of specular reflection, the angle of incidence relative to the surface normal would equal the angle of emission and lie in a plane normal to the surface, whereas the reflection is diffuse if the intensity of emitted molecules as a function of the angle of re-emission obeys the cosine law. This is equivalent to Lambert's law for optical or thermal radiation—already well established by this time. For example, the calculations by Christiansen (1893) for radiation from a circular disc onto a parallel disc, or an infinite parallel plane, proved useful in later applications to the molecular flow equivalent. In a later example, the probability functions set up by Clausing (1932) for short cylindrical tubes were able to be related to papers by Walsh (1920, 1929) treating a light radiation problem which, by virtue of the cosine law applying to it, coincided in its mathematical description with his analysis.

The necessity of the cosine law as arising from the second law of thermodynamics was put forward by Gaede (1913a) and Knudsen (1916) by considering the distribution of energies and directions of molecules emerging from a stationary surface. It was re-iterated by Millikan (1923) and discussed again by Clausing (1930b, c). The earlier discussion stimulated a series of experiments initiated by Wood (1915, 1916) (figure 3), Knudsen (1916, 1918) (figure 4) and Langmuir (1916a, 1917) demonstrating the

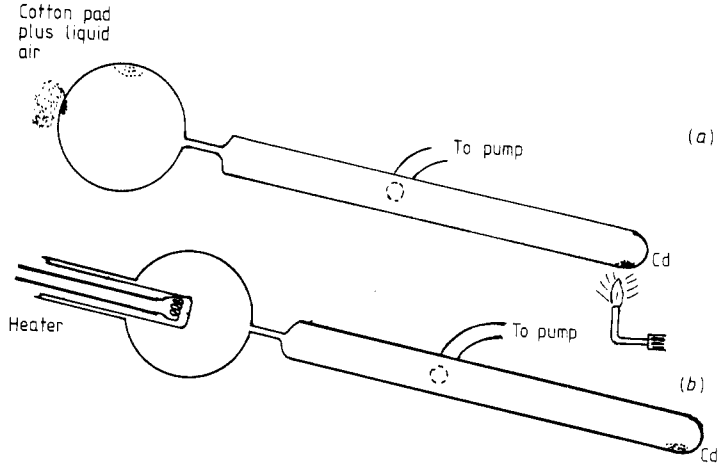

Figure 3. Experiments by Wood with cadmium vapour to show the linear propagation and the cosine distribution of re-evaporated material on the evacuated spherical bulbs.

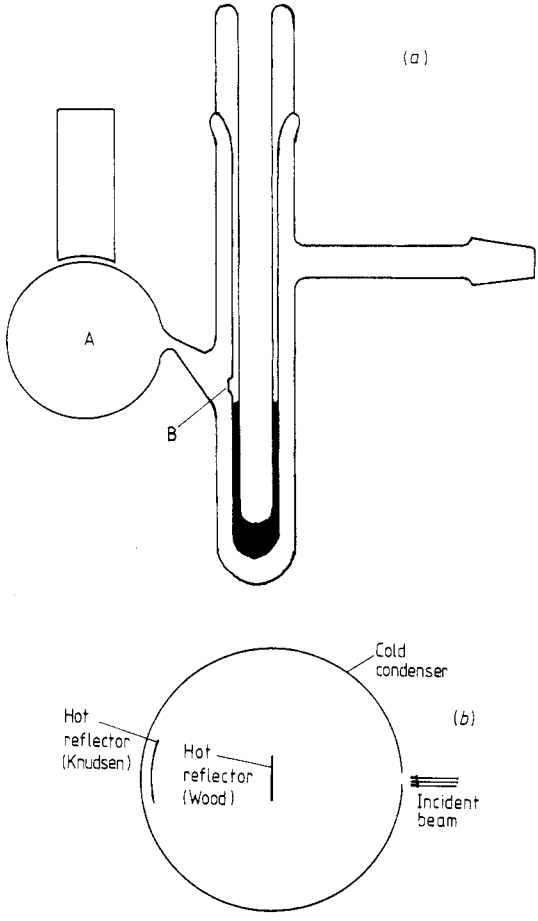

Figure 4. (*a*) Experiments by Knudsen demonstrating the cosine law in terms of uniform deposition on the inner surface of the sphere (using mercury). (*b*) Diagram comparing Knudsen's experiment with Wood's.

cosine law and showing the uniform deposition from a surface source at the circumference of a spherical glass chamber onto its inner surface†. These concepts were, of course, fundamental to more recent applications of thin-film deposition. Examples of the calculation of the thickness distribution of thin films deposited from various types of sources were given by Holland and Steckelmacher (1954) and evaluated experimentally. A technique using radioactive tracers was developed by Preuss (1953, 1956) with which he obtained polar plots as in the example shown in figure 5. It is not possible here to mention all the numerous subsequent studies concerned with detailed investigations of the interactions (usually energetic rather than thermal) of molecular beams with a variety of surfaces (as in the early review, for example, by Hurlbut (1959)) especially at glancing angles of incidence, or sputtering effects as observed with highly energetic beams. After a slow start in the 1930s, following the classical diffraction experiments of Estermann and Stern, these studies have become an important technique not only for surface crystallography, but also to yield information on interaction potentials, low-energy excitations, reactive scattering, the reactive and

† Discussed also in Knudsen (1934) and shown in Hirth and Pound (1963).

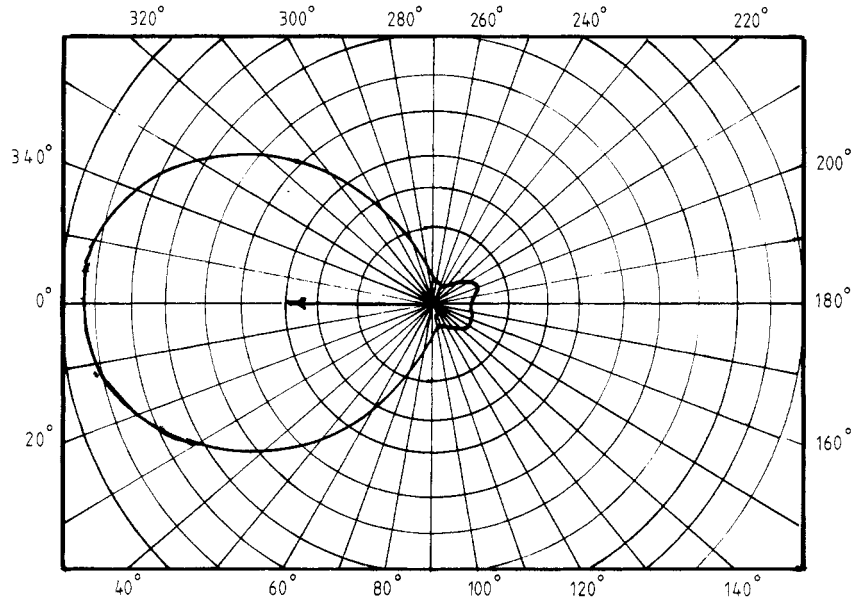

Figure 5. Polar plot of dispersion patterns when evaporating ^{51}Cr from a 45° conical source obtained by Preuss (1956).

catalytic mechanism and a whole range of processes depending on energy and momentum exchange (Stickney 1967, Frankl 1952, Goodman 1977, 1980). We should only note that the study of the angular- and velocity-resolved kinetic properties of the desorbing or scattered molecules would hardly have been rewarding were the angular distribution of the flux of these molecules always to be a cosine one. So how about the Knudsen cosine law for applications to problems in rarefied gas flow?

One must concede that there are certain surface conditions for which variations from the expected cosine law of diffuse scattering are known to occur. Apart from the diffraction effects involving specially prepared clean surfaces, usually single crystals, sorption and chemical interaction effects can also play an important role. These were outlined in the review by Steckelmacher (1974) giving references to recent studies of surface effects on flow. Special problems pertinent to the Knudsen effusion technique were outlined in the recent overview by Carter (1979). The following effects have been found to be important.

(*a*) Sorption of vapour or gas on the tube walls.

(*b*) Delayed sorption effects, allowing for a residence time of the sorbent species; this was considered particularly by Clausing (1930d, 1962). The phenomenon is related to the Blears effect in ionisation gauges used for measurements in oil-contaminated systems.

(*c*) Surface diffusion.

(*d*) Chemical reactions at the walls, dissociation of polymers, etc.

(*e*) In the Knudsen effusion techniques, Knudsen cell and detector geometrical effects can also obscure the underlying problems.

An important development in the discussion concerning the cosine law was the critical analysis by Comsa (1968) followed by Wenaas (1971). They emphasise the need to distinguish between the 'equilibrium cosine scattering law' and the 'Knudsen law of diffuse scattering' at the gas-surface interface. As outlined in their discussion

of flow through cylindrical tubes, Steckelmacher and Lucas (1983) have come to the conclusion that the Knudsen law can be accepted as a correct basis for the evaluation of rarefied gas flow in both simple and complex vacuum systems—as well as related fields such as the distribution of evaporated thin-film deposits and certain aspects of molecular gas dynamics.

It is interesting to note that the full implication of the cosine distribution under molecular flow conditions is occasionally still ignored as in the recent paper by Perram and Dorko (1983). The necessary correction for the flux distribution and flow pattern through a rectangular slit orifice was given in Steckelmacher and Man (1985).

4. Molecular flow in tubes

4.1. Long tubes

In his first paper Knudsen (1909a) set out to study molecular flow in tubes and determine the dependence on tube dimensions. He derived an expression for the gas flux through a tube (long compared with its cross-sectional dimension) (figure 6) from which he obtained the flow resistance Z given by

$$Z = \frac{3}{8}\left(\frac{\pi}{2}\right)^{1/2} \int_0^L \frac{o}{A^2}\,\mathrm{d}l \tag{4.1}$$

where o is the circumference and A is the cross-sectional area. For a cylindrical tube with circular section, radius R, this leads to

$$Z = \frac{3}{8}\left(\frac{2}{\pi}\right)^{1/2} \frac{L}{R^3} \tag{4.2}$$

still accepted and much used today. We also note that for a rectangular slit section tube of width b, separation a, he derived from the integral the flow resistance when $a \ll b$:

$$Z = \frac{3}{4}\left(\frac{\pi}{2}\right)^{1/2} \frac{L}{a^2 b}. \tag{4.3}$$

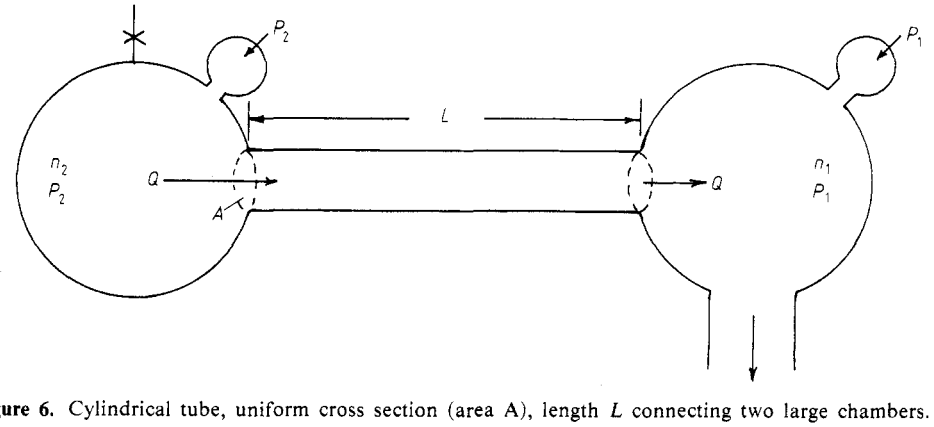

Figure 6. Cylindrical tube, uniform cross section (area A), length L connecting two large chambers. A constant gas throughout Q (pressure-volume units) maintains the pressure difference $P_2 - P_1$, where P_2 and P_1 are measured in regions remote from the entrance and exit of the tube.

The integral expression of equation (4.1), supposedly valid for any cross-sectional shape, and its method of derivation was severely criticised by Smoluchowski (1910) who gave an alternative corrected expression involving the integral

$$I = \int_s \int_{-\pi/2}^{+\pi/2} \frac{1}{2} \rho^2 \cos\theta \, d\theta \, ds \tag{4.4}$$

giving for a parallel tube:

$$Z = 2(2\pi)^{1/2} L/I \tag{4.5}$$

where ρ is an arbitrary cross-sectional chord and θ and s refer to the interception at the tube surface (figures 7 and 8). Further details of the derivation and the effect of the error in terms of some typical cross-sectional shapes other than circular (elliptical, rectangular and triangular) were given in Steckelmacher (1978) including references to the earlier literature of which Clausing (1929) should be specially mentioned, since he had already given details for rectangular and annular cross-sectional tubes.

To show the sort of errors involved, take the example of a square section (length of side $= a$), for which (as in Smoluchowski)

$$I = 4a^3[\ln(1+\sqrt{2}) + (1-\sqrt{2})/3] = 2.97321a^3$$

giving

$$Z_S = (2(2\pi)^{1/2}/2.97321)(L/a^3)$$

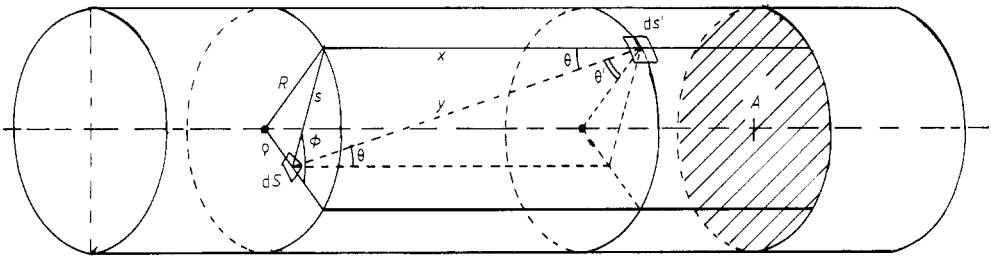

Figure 7. A circular section tube, radius R, showing gas interchange with a small wall element dS' under molecular flow conditions.

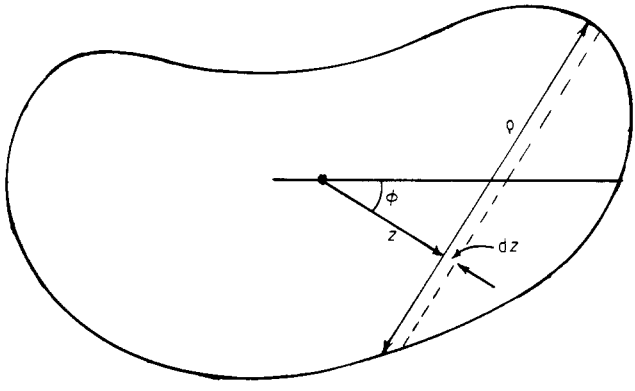

Figure 8. Cross section of general shape. The chord, length ρ, is shown at the angle $(90° - \varphi)$ to a fixed line. This is required in the evaluation of the integral I (4.4) when using the convenient alternative (to the double integral) derived by Barrett and Bosanquet (1944) who showed that $I = \pi$(average value of $\int \rho^2 \, dz$).

compared with the Knudsen value

$$Z_K = \tfrac{3}{2}(\sqrt{\pi}/2)(L/a^3)$$

so that

$$K = Z_K/Z_S = 2.97321 \times 3/8 = 1.11495. \tag{4.6}$$

Furthermore, Smoluchowski pointed out that an expression for the flux Q containing an inverse proportionality with the circumference was most unlikely to be correct since Q could then be arbitrarily reduced merely by the insertion of a number of slots into the circumference. This is a misleading statement. Consider dividing a square section tube by n equally spaced (thin walled) partitions on each side to create a bundle of n^2 square section tubes (as in multiple capillary beamed systems mentioned in § 5 below); then the overall Q will be reduced by the factor $1/n$. The error was conceded in Knudsen's reply, and I have only mentioned it yet again because the incorrect Knudsen formulation, which found its way into most current books on vacuum science and technology (as well as some on the kinetic theory of gases), is still reproduced in the very latest editions. We may also note that the correct alternative was already available in books by Lorentz (1927), Herzfeld and Smallwood (1931), Loeb (1934), giving both derivations, and Present (1958). Many of the other books try to correct things by introducing a factor K (ratio of true conductance to incorrect Knudsen value) tabulated or as a graph for selected values of rectangular or annular sections. Values for K were first listed in Guthrie and Wakerling (1949)† taken from a report by Barrett and Bosanquet (1944). For a square section, they gave $K_B = 1.108$, rather than 1.115 as found in (4.6) above. One can therefore readily recognise the origin of the data in all the books using this value of K_B. For comparison see figure 9; some values of K for a range of rectangular cross sections are compared in table 2. In the regime of molecular flow, an alternative and in some ways preferable concept to the flow resistance (or conductance) as introduced by Knudsen (and taken up by Dushman and many others since then) is that of transmission probability, as first discussed by Clausing (1926, 1929, 1932). This turns out to be most useful in the case of short tubes and more complex components or systems as outlined below (§ 4.2). The effect of cross-sectional shape on the transmission probability of long tubes is best appreciated by comparing the ratio of transmission probabilities for elliptical or rectangular section cylindrical tubes of equal cross-sectional areas, as in figure 10. For a more detailed discussion of these and other data, see Steckelmacher (1978).

4.2. Short tubes and thin orifices

The question now arises: what to do about flow through short tubes where entrance and exit effects must be taken into account. Before discussing this we turn to the second paper of Knudsen (1909b) dealing with molecular flow and effusion through orifices. He obtained the usual expression for the conductance, related of course to the impact rate per unit area $\frac{1}{4}\bar{v}n$, where $\bar{v}$ is the mean molecular velocity and n is the gas density (number of particles per unit volume) on one side (assuming a vacuum on the other). These concepts were used in the third paper (Knudsen 1909c) to determine vapour pressure by what has become known as the effusion method, the

† They also quote the correct formula as obtained from Loeb (1934).

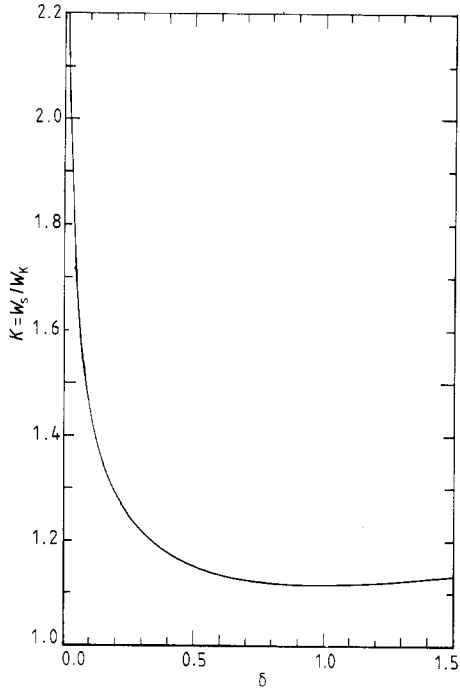

Figure 9. Correction factor K for rectangular section tubes, where K is the ratio of the transmission probability W_S (as obtained by Smoluchowski) to W_K from the Knudsen formula). This is plotted as a function of $\delta(=a/b)$, i.e. ratio of length of sides a, b of rectangle.

developments of which were also reviewed in their historical context by Carter (1979). Here the molar effusion rate is related to the vapour pressure, i.e. from $p = nKT$, or one obtains a relation for the mass effusion rate† (also important in thin-film deposition to determine deposition rates from evaporation rates).

Knudsen (1909b) also indicated the idea of treating flow as an impedance or resistance in the electrical sense so that one could derive a combination of a tube and an orifice in terms of the addition of their flow resistances. This was taken up by Dushman (1922) who postulated that the flow resistance of a short tube ought to be the sum of the resistances of the entrance opening and that of a long tube. However, there is no good reason for employing in this addition a formula derived for $L \gg d$ (i.e. very long tubes) and the problem of short tube conductances was not properly resolved until the detailed studies by Clausing (1932). Reviews of the molecular flow conductance for systems of tubes and components together with related questions were given in Steckelmacher (1966, 1974) and Lyubitov (1967) while Venema (1973) gave a very useful appraisal of Clausing's contribution to these problems. We can only mention here that he first introduced the concept of transmission probability in preference to flow resistance (or conductance). He derived an integral equation for this probability which he then attempted to solve by suitable approximations. These provided for the first time a table of values for a wide range of L/R serving as a basis for flow calculations in tubes for more than twenty years.

† As derived by Hertz (1882).

Table 2. Comparison of transmission probabilities for rectangular tubes of Smoluchowski W_S with Knudsen W_K.

$$\delta = a/b \qquad K = W_S/W_K = \tfrac{3}{8}(1+\delta)F$$

where

$$F = \frac{1}{\delta}\ln\left[(\delta+(1+\delta^2)^{1/2}\right] + \ln\left[\frac{1}{\delta}+\left(1+\frac{1}{\delta^2}\right)^{1/2}\right] - \frac{\delta}{3}\left(1+\frac{1}{\delta^2}\right)^{3/2} + \frac{\delta}{3}\left(\frac{1}{\delta^3}+1\right)$$

$$W_S = (a/L)F$$

for square section, side a_0;

$$W_\square = \frac{2a_0}{L}\left[\ln(1+\sqrt{2}) + \frac{1-\sqrt{2}}{3}\right] = 1.4866\frac{a_0}{L}.$$

In terms of square of *same area* as rectangle $ab = a_0^2 = a^2/\delta$:

$$W_\square = 1.4866a/L\sqrt{\delta}.$$

Hence

$$\frac{W_S}{W_\square} = \frac{\sqrt{\delta}F}{1.4866}$$

δ	$1/\delta$	K	K_B	$W_S/W_\square$
1	1	1.114 95	1.108	1.0
$\frac{2}{3}$	1.5	1.126 69	1.126	0.9901
0.5	2	1.148 98	1.151	0.9716
$\frac{1}{3}$	3	1.199 13	1.198	0.9314
0.2	5	1.290 41	1.297	0.8626
0.125	8	1.397 93	1.400	0.7881
0.1	10	1.455 57	1.444	0.7506
0.05	20	1.647 4	—	0.6293
0.01	100	2.202 6	—	0.3912
0.001	1000	3.303 9	—	0.1872

$K_B = K$ from Barrett and Bosanquet (1944) (as in Guthrie and Wakerling 1949).

The accurate evaluation of transmission probabilities was taken up again by De Marcus and Hopper (1955) and a series of much quoted reports by De Marcus (1956/9), based on various improved techniques to obtain approximate solutions to the Clausing-type integral equations. More recently, Berman (1965) and Neudachin *et al* (1972) obtained series expansions, also discussed by Essen and Heerens (1976), while results to the highest accuracy likely to be required for some time were calculated by Cole (1977a, b) over a range of L/R from 0.1-1000 with a relative error never greater than 8.6×10^{-2}% as shown in table 3 for some selected values. This was based on maximum and minimum principles for approximations to the Fredholm type of integral equations typical for gas flow and transport problems as devised by Cole and Pack (1975). It will be seen from this, incidentally, that for many practical purposes the early tabulated data of Clausing (1932) would be quite adequate.

Another useful approach, particularly well suited for complex shapes and systems of baffles, traps, etc, is the statistical one using Monte Carlo techniques. This was described by Davis (1960) and Davis *et al* (1964) and many publications since then, but can only be briefly mentioned here. It may just be noted that a very recent contribution by Carette *et al* (1983) found a large discrepancy in their Monte Carlo

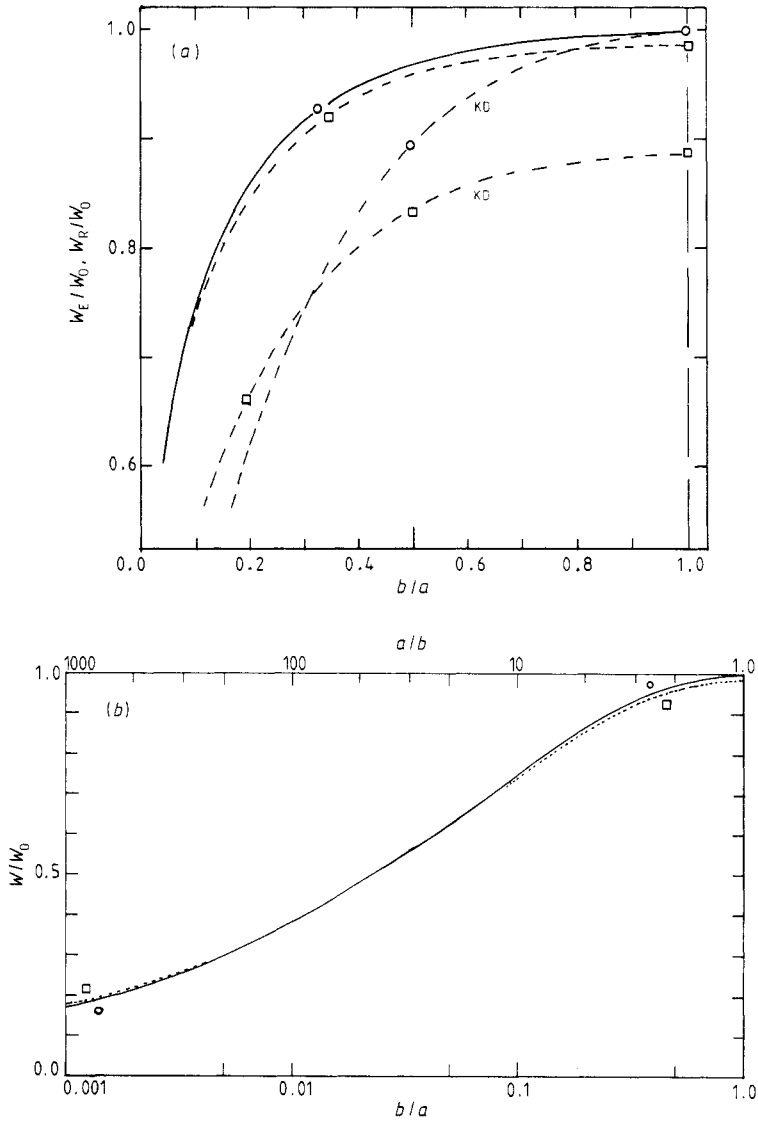

Figure 10. (*a*) Ratio of transmission probabilities (for tubes of equal cross-sectional areas W_E/W_0 and W_R/W_0 as a function of the ratio of axes (or length of sides), respectively, for elliptical and rectangular section tubes. Full lines as derived in Steckelmacher (1978) where ○, elliptical; □, rectangular section; KD, Knudsen and Dushman formula. (*b*) Comparison of transmission probability of equal area elliptical and rectangular tubes over a wide range of b/a.

data for the conductance of cylindrical tubes compared with all published figures (amounting to larger than 56% for long tubes to 25% for as short as $L/R = 5$). However, as indicated in the recent comment (Steckelmacher 1983; see Carette *et al* (1983)), this appeared to be related to an error in their particular application of the Monte Carlo procedure when setting up the cosine law of diffuse reflection at the tube walls, as acknowledged in an erratum. Monte Carlo results calculated by Beijerinck *et al* (1976) showed good agreement with other calculations, as shown in the last column of table 3.

Table 3. Transmission probabilities: comparison of data for cylindrical tubes.

L/R	Clausing (1929, 1932)	De Marcus (1957) and †Lund and Berman (1966)	Cole (1977a, b)	Relative % error Cole (1977a)	Beijerinck *et al* (1976)
0.2	0.9092	0.909 22	0.909 215 028		
0.4	0.8341	0.834 08	0.834 080 661		
0.6	0.7711	0.771 15	0.771 149 758		
0.8	0.7177	0.717 79	0.717 785 082		
1	0.6720	0.671 98	0.671 983 902	5.4×10^{-10}	0.6725
2	0.5136	0.514 23	0.514 230 527	1.7×10^{-6}	0.5148
4	0.3589	0.356 58	0.356 572 25	1.4×10^{-4}	0.3568
5	0.3146	0.310 53	0.310 524 62		
10	0.1973	0.190 99	0.190 941 0	7.7×10^{-4}	
20	0.1135	0.109 32	0.109 304	1.7×10^{-2}	
50	0.0499	0.048 517†	0.048 448		0.0488
100	0.0258	0.025 29†	0.025 258	8.6×10^{-2}	

4.3. More precise evaluation for long tubes

The Clausing-type integral equation procedure discussed above for relatively short tubes clearly also gives results valid for long tubes. It was also applied by Berman (1965) and Pao and Tchao (1970) to obtain an asymptotic solution for very long tubes of circular section†. Thus it is comforting to know that the first term in both expansions for the transmission probability is $8R/3L$, equivalent to that found by Knudsen (1909a). His first six terms would give an error of 1% for $L/R \geqslant 20$. Pack and Yamamoto (1976) developed an asymptotic theory comprising a variational procedure of wider applications. This was recently improved by Yamamoto *et al* (1980) to obtain much higher-order approximations.

4.4. Approximate values of transmission probabilities

For many purposes the use of tabulated data can be a nuisance and simple formulae or the use of the first few terms in an expansion would be preferable. The earliest example was the Dushman (1922) method already mentioned above. A better approximation was suggested by Kennard (1938). Applications of a simple short tube transmission probability:

$$W = \frac{1}{1 + L/2R}$$

were considered by Helmer (1967). Henning (1978) made further proposals, while Steckelmacher and Henning (1979) discussed them in relation to other approximations. An approximation for tubes of constant annular cross section was recently proposed by Onusic (1980) and discussed in more detail in Steckelmacher (1981). The construction of graphs and monographs was evaluated by Levina (1980) for the complete range from viscous to molecular flow. New concepts proposed by Santeler (1986a) allowed

† The transmission probability to two terms was

$$W = \frac{8R}{3L} - \frac{2\ln(L/R)}{(L/R)^2}.$$

for the molecular beaming in the combination of tubes. Santeler (1986b) considered the exit loss effect for viscous flow in short tubes.

4.5. Molecular conductance of complex systems

The earliest approach was that of the tube resistance or impedance concept (or its inverse, that of conductance) already suggested by Knudsen (1909b) and developed by Dushman (1922) as mentioned above. The electrical circuit analogy has since been discussed from time to time as outlined more fully than can be attempted here in Steckelmacher (1966). A typical problem is the apparently simple one of how to compound the transmission probability of tubes of varying cross-sectional area considered in a rather *ad hoc* manner by Dushman (1922), and again for tube systems by Harries (1951). The transmission probability (and single component or tube) is evaluated in the first place for the case when this is placed between two large vessels as in figure 6, i.e. gas may be assumed to arrive at the entrance and exit in a random (or chaotic) manner. Hence, the intrinsic conductance is related in the usual way to the product of entrance conductance and transmission probability. However, even for a simple composite system of, say, two tubes, account must be taken of the beaming effect of gas from one to the other. The required correction was discussed in terms of the transmission probability for cylindrical tubes by Oatley (1957), Steckelmacher (1957), Ballance (1965) and more recently by Fustoss (1970), Dayton (1972), Fustoss and Toth (1972) and again by Stubblefield (1983). Related to this is the effect of placing a diaphragm with a thin aperture across, or inside, a tube discussed by Bureau *et al* (1952). Numerical solutions to the integral equations for such an arrangement were obtained by Kato and Fujimoto (1976). Further details of how to deal with the transmission probability for complex systems were also discussed in Holland *et al* (1974) and Saksakanski (1980). Electrical analogues were recently described for solving specific problems by Paulick (1982) and for complex vacuum systems by Ohta *et al* (1983). A useful analysis and review was presented by Haefer (1980), while applications to cryopumping were discussed in his book (Haefer 1981).

5. Molecular beaming effects of tubes

The formation of beams under molecular flow conditions from the end of short tubes was not studied until Clausing (1930a) calculated the emission pattern (figure 11). Further studies of the beaming pattern from moderately short tubes, based on the Clausing procedure, were made by Dayton (1957). The results of his calculations were displayed in a series of diagrams (figure 12) clearly illustrating the narrow and more pointed beam pattern emerging from a tube as its L/R increased. In contrast, a short tube (i.e. thick orifice) showed a relatively small departure from the spherical cosine distribution, which may be compared with related experimental results obtained by Preuss (1956) as in figure 5. He also indicated (and made one aware of) the back-scattering effect, i.e. the lobes at the imput end of a tube due to gas which was back-scattered towards the input by the walls. For a review of this and other subsequent studies of the beaming effect from single and multiple capillary arrays, see Steckelmacher (1966, 1974), Lucas (1973), Steckelmacher *et al* (1978) and Steckelmacher and Lucas (1983).

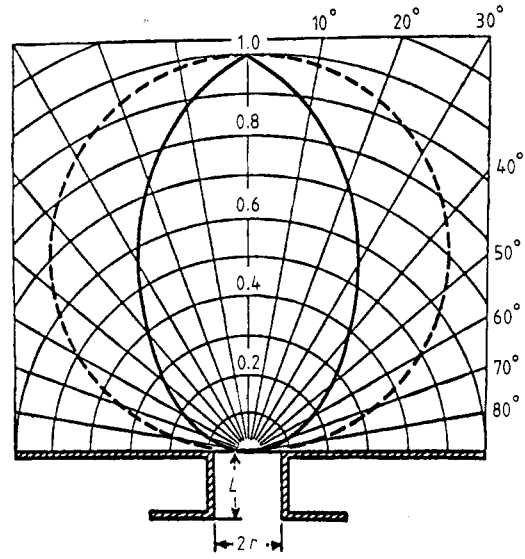

Figure 11. Polar diagrams of gas flow from a cylindrical tube into a vacuum for the special case $L = 2r$ as calculated by Clausing (1930) compared with flow through an orifice (- - -).

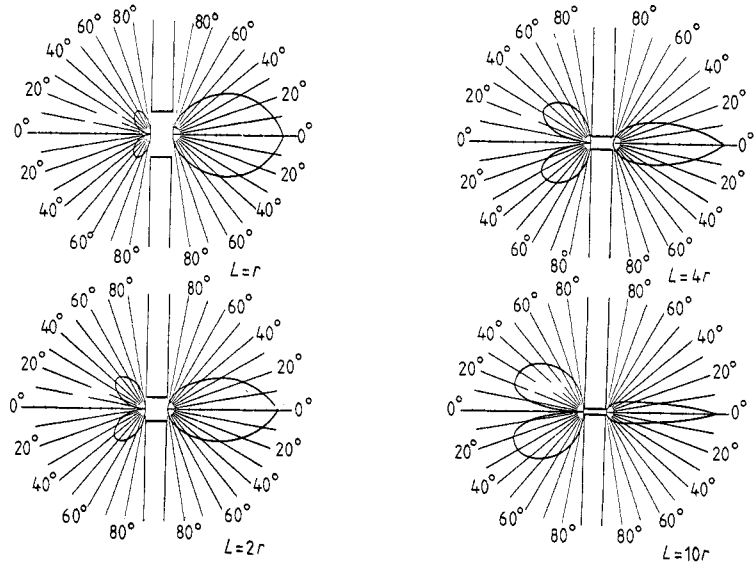

Figure 12. Polar diagrams of gas flow at the entrance and exit of cylindrical tubes (calculated by Dayton (1957)) for tubes with $L = r$, $L = 2r$, $L = 4r$ and $L = 10r$.

A related subject which has also been much investigated is the molecular-impact rate distribution along the inside wall of the tube, which in turn largely determines the density distribution and directional streaming of gas inside, and hence outside, the tube. This again was first studied by Clausing (1930a) as an essential part of his analysis of the beaming effect. It is not possible here, in this brief review, to do justice to the many valuable contributions which have been made since then. An important application of these wall impact studies is in the field of standardisation of measuring

procedures under dynamic rarefied gas flow conditions. Thus wall collision distributions need to be taken into account for adopting the correct procedures in the measurement of the speed of high vacuum pumps, cryogenic pumping, calibration of vacuum gauges and meaningful pressure measurements (see also Steckelmacher 1966, 1974). We will just mention some of the most recent investigations of wall impact for circular cross-section tubes under conditions of purely molecular flow. Experiments by Kurepa and Lucas (1981) in which the impact rate was actually measured by means of a movable slot along the capillary wall obtained results which did not agree with some of the calculated data. This stimulated calculations to a higher accuracy by Davies and Lucas (1983), which however largely confirmed the earlier results of Clausing (1930a), and more recently of Neudachin *et al* (1972), suggesting to them that some of the theoretical assumptions—such as the diffuse molecular scattering—needed to be modified. Critical comments by Steckelmacher and Lucas (1983) supported the theoretical studies and proposed instead to change some of the experimental procedures since the method used to monitor impact rate could have been directionally sensitive. Yamamoto *et al* (1982) have also independently calculated the wall flux to a high accuracy by a different analytical technique with results which are in excellent agreement with those of Davies and Lucas (1983).

The application of multicapillary arrays has been found to be particularly useful to produce well defined gas beams as gas targets, for example, in studies of electron or ion beam collision phenomena. This has been much simplified in recent years with the availability of thin microchannel plates, produced by a technique originally developed mainly for electron and photomultiplier devices. These are typically only 1 mm thick with capillaries of, e.g., 5 μm radius (i.e. $L/R = 200$) allowing relatively high density localised targets to be obtained by the use of high input preserves, while still operating under molecular flow conditions (as discussed in Steckelmacher *et al* (1978) including references to the earlier literature). To obtain still higher target densities, the input pressure may be further increased, but then the effect on changes in flow conditions through the capillaries must be considered as the mean free path λ becomes comparable to or less than the tube length L and eventually the tube radius R. The intermediate condition, when $\lambda > R$ still holds, but $\lambda \leqslant L$, has been referred to as the 'opaque mode' of flow, as discussed for example by Lew (1967), Beijerinck and Verster (1975) and Brinkmann and Trajmar (1981). Recent applications to obtain absolute cross sections in an ion beam collision system were described in Man *et al* (1986). If the input pressure were increased further, with both $\lambda \ll R$ and $\lambda \ll L$, then the *transition* to the *continuous viscous flow mode* would need to be considered.

6. Transition flow at lower Knudsen numbers

The transition from molecular to viscous flow was one of the main subjects of the early experiments of Knudsen (1909a) and Gaede (1913) as mentioned above. When the mean free path becomes comparable with and smaller than the tube dimensions, intermolecular collisions compete with purely wall collision effects, complicating any analysis accordingly. This is again a subject really outside the scope of this review but it may be useful just to mention a few more recent publications.

Pollard and Present (1948) examined the concept of self-diffusion in this regime and obtained a qualitative explanation of the Knudsen minimum. In a useful review Weber (1954) gives a detailed analysis of the laminar flow of pure gases in tubes and

the self-diffusion coefficient. Scott and Dullien (1962) developed a new equation for the total flow regime in infinite circular tubes in which all flow coefficients were derived from intermolecular and wall collision probabilities. This was applied to vacuum problems as well as flow through porous solids. Lund and Berman (1966) presented a useful model describing transport under a partial pressure gradient (self-diffusion) as well as under a total pressure gradient in capillaries of arbitrary L/R and covering pressures from free molecule to continuum regimes. Comparison with experimental data suggested that surface effects and slip contributions needed to be taken into account. In another approach, a large number of investigations used the more rigorous theoretical analysis based on the Boltzmann equation, but usually under formidable difficulties. However, the numerical calculations by Cercignani and Sernagiotto (1966), using the Bhatnagar, Gross and Krook (BGK) collision model to reduce the integro-differential transport equation to a purely integral one, made it possible to apply finite difference techniques. Thus, the Knudsen minimum, as in figure 13(a) from Knudsen (1909a) could be confirmed on a rigorous theoretical basis close to that obtained experimentally. Typical theoretical and experimental results from Lund and Berman (1966) are shown in figure 13(b). Ferziger (1967), casting the integral equation for the velocity into a convenient form, obtained asymptotic solutions. A variational calculation for the volume flow rate was also obtained. Further elaborations were discussed by Cercignani and Pagani (1967), and in their wider context by Cercignani (1969, 1975). Edwards (1976) presented a brief review covering the transition flow through tubes and nozzles. Recently Datta and Rinker (1981) gave an overall analysis of the various models available to describe rarefied gas flow in long tubes. De Muth and Watson (1986) discussed the transition flow rates through an orifice.

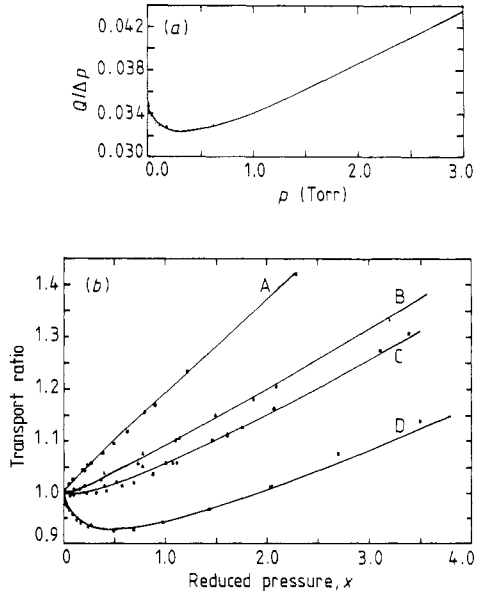

Figure 13. (a) Volume flux $Q/\Delta p$ as a function of mean pressure for CO_2 flow through a long capillary. $R = 0.0033$ cm, $L/R = 613.2$ (from Knudsen 1909), $L = 2.042$ cm. (b) Flow of gases through capillaries of various length to radius ratio. A, He: $L/R = 0.319$; B, Xe: $L/R = 11.84$; C, He: $L/R = 11.84$; D, CO_2: $L/R = 613.2$. Points show experimental results; lines fitted model equation (from Lund and Berman 1966). Note the pronounced minimum only for sufficiently long tubes.

Thermal transpiration, mass and energy flow effects were studied in a sequence of papers by Loyalka and some colleagues of which the fourth (Loyalka *et al* 1982) was concerned with the flow of rarefied polyatomic gases in a cylindrical tube. They found that the flow was adequately described by the Hanson and Morse (1967) model of the linearised Wang Chang-Uhlenbeck equation (De Boer and Uhlenbeck 1964) together with Maxwell's diffuse scattering boundary condition. In a similar procedure Lo *et al* (1983) successfully extended their studies to rarefied gas flows in a cylindrical annulus, obtaining good agreement with experimental results over the complete range of flow.

7. Conclusions

The wide range of material which it was found necessary to review shows how Knudsen's work on the flow of rarefied gases is still of fundamental importance today. It is also interesting to note the effect on what are now quite separate fields and disciplines, which one sometimes feels lack contact with each other. Thus, as we have seen, Knudsen flow is of fundamental importance not only in vacuum and surface science, but also makes its appearance in the quite separate areas of rarefied gas dynamics and flow, and molecular beams (all have their own separate international conference series); one must also mention studies on the kinetic theory of gases as well as apparently unconnected subjects such as thin-film deposition, molecular-beam epitaxy and the characterisation of high-temperature vapours. This will also be evident from the selection of literature references, hopefully useful to the reader.

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