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Experimental data and theoretical modeling of gas flows through metal capillary leaks

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Metal capillary tubes are commonly used as leak elements to admit known flows of gases into vacuum systems for calibration of vacuum gaging equipment. In many instances it is desired to generate flow rates over a range of three or more decades, preferably with a single leak element. The generation of flow rates over wide ranges is possible with metal capillary leaks, but in most cases the conductance of the leak element will need to be measured as a function of the relevant pressures due to the changing of the flow regimes. Many fits to experimental data and theoretical models exist for predicting the flow rate through tubes, but their validity is not well established. In this study, measured conductances of stainless steel tubes for flow rates of 10^{-8} to 10^{-14} mol s^{-1} with several inert gases are compared with various experimental and theoretical models of gas flow in the molecular, viscous and transition flow regimes. Characteristics of crimped metal capillaries are also examined over this range of flows.

1. Introduction

Generation of known leak rates is important for a variety of industries that use leak detection as a method of nondestructive testing to assure product reliability and performance. One of the primary uses of calibrated leaks is for calibration of helium leak detectors and mass spectrometers used for leak detection purposes. It is common practice to calibrate these instruments with a fixed gas density reservoir leak, either a capillary or permeation type, and to assume that the instrument has a constant sensitivity. For many applications it would be advantageous to calibrate the instruments over a wide range of leak rates, and in many situations for a variety of gas species. Capillary leaks are best suited to this task as their leak rate can be changed many orders of magnitude by changing their reservoir pressure and they are compatible with most gases.

In many instances it is desirable to know the leak rate of a capillary leak for various gases, temperatures, and pressures. Characterization of capillary leaks for such varied conditions can be very costly due to the need to characterize the conductance of a capillary tube, most often of undeterminable diameter, over a very wide range of flow rates which correspond to changing flow regimes. Various theories and experimental fits have been developed to predict the conductance of tubes as a function of the mean free path of a gas species for small pressure gradients. These theories have limited applicability to the case where the pressure gradient across the capillary is very large, such as for commercially available capillary leaks. In most cases the capillary diameter is required to be on the order of micrometers to generate the desired leak rates. The diameter may also vary along the length of the tube and in some instances the tube may be crimped by the manufacturer to reduce the conductance; in such cases it is not feasible to measure the diameter.

The conductance of two types of commercially available metal capillary leaks has been determined for a variety of gases over a wide range of pressures. The functional form of the dependence of the capillary conductance on the mean free path of the gas is compared with various experimental and theoretical models for flow in the transition regime.

2. Description of apparatus

The flows from the capillary leaks were measured using the primary leak standard that has been developed at NIST[1,2]. The primary leak standard is divided into three major components: the vacuum chamber, the leak manifold, and the flowmeter. The operation of the flowmeter is described in the literature in detail[1,2]. The vacuum chamber is composed of two chambers, upper and lower which are separated by a 1 cm diameter orifice, and a 2500 l s^{-1} turbo molecular pump connected to the lower chamber is utilized to achieve a high vacuum. Three mass spectrometers are connected to the upper chamber for partial pressure measurements as well as three ionization gages for total pressure measurements. The flow from the flowmeter or leakstand can be directed into either the upper or lower chambers.

Determination of the leak rate is accomplished by the following procedure. The leak is placed on the leak manifold and the entire system is evacuated. The reservoir of the leak is evacuated and filled with the appropriate gas to a specific pressure. Gas emitted from the leak is valved into the vacuum chamber, flows through the orifice and is evacuated by the pump. After the gas flow and upper and lower chamber presure reach equilibrium, the upper chamber gas partial pressure indication, as measured using a mass spectrometer(s) tuned to the appropriate mass, is recorded. The leak is then isolated from the system, the flowmeter is pressurized with the appropriate gas and the flow is adjusted such that, upon stabilization, the upper chamber gas partial pressure is the same as the partial pressure recorded while the leak was connected to the vacuum chamber. The flow rate is then measured using the constant pressure flowmeter. The constant pressure flowmeter is capable of measuring flow rate between 10^{-6} and 10^{-12} mol s^{-1}. To measure flow rates lower than 1×10^{-12} mol s^{-1}, a flow division technique is employed and allows extension of flow rate measurements to 2×10^{-14} mol s^{-1}

The uncertainties of the leak rate measurements vary from 1% at 10^{-6} mol s^{-1} to 8% at 10^{-14} mol s^{-1}.

3. Experimental procedure

The data presented here were taken over a two yr period with flow rates ranging from 10^{-8} to 10^{-14} mol s^{-1} for two types of capillary leaks: a nominally constant diameter stainless steel tube and two crimped stainless steel tubes. To measure the flow, the capillary leaks were first evacuated with a roughening pump and then gas was introduced on one side of the capillary element and the pressure was measured with either a quartz Bourdon tube gage or a capacitance diaphragm gage. The downstream pressure was maintained below 10^{-2} Pa. For all of the capillaries examined the pressure ratio across the capillaries was greater than 1000, the upstream pressure was greater than 100 Pa, and the downstream pressure was assumed to be zero. The leak rate was then measured using the procedure outlined in Section 2. Measurements were performed over a pressure range of 100 Pa–0.4 MPa for the constant diameter capillary for helium and over a smaller range for nitrogen, argon, and sulfur hexafluoride. The two crimped metal capillary leaks are designated NBSL48 and NBSL44. The conductance of NBSL48 was determined over a pressure range of 20 kPa–2 MPa with helium, nitrogen and argon. The conductance of NBSL44 was measured over a pressure range of 0.2–2 MPa with helium and argon.

4. Models for transition gas flow through tubes

Due to the lack of theoretical or experimental models for the flow of gases through tubes with high pressure differentials ($P \approx \Delta P$) in the transition flow regime, quite often, models developed for low pressure differentials ($P \gg \Delta P$) are utilized. Some of the models most frequently discussed in the literature are reviewed here.

The viscous flow theory based upon the Navier–Stokes equations[3] does not adequately describe the flow of gases through tubes in cases where the mean free path of the gas approximates the diameter of the tube. An equation that accounts for a non-zero velocity of the gas at the tube walls, known as the slip flow equation[4], is often employed to extend the validity of the viscous equations into the transition regime, namely

$$Q = -\left(\frac{\mu a^4}{8\mu}\right) P_a \frac{dp}{dx}\left(1+4\left(\frac{2}{f_s}-1\right)\frac{\lambda}{a}\right). \qquad (1)$$

The flow rate Q is expressed as a function of the mean free path of the gas λ, the radius of the tube a, the fraction of molecules assumed to be absorbed and re-emitted diffusely f_s, the gas viscosity μ, the average pressure in the tube P_a, and the pressure gradient along the tube dp/dx. When extended into molecular flow, comparisons of the slip flow equation with calculations based upon the kinetic theory of gases show large deviations.

Numerous investigators have modeled the flow of gases through tubes with small pressure gradients in the transition regime both experimentally and theoretically[5–12]. A model for the conductances of tubes as a function of the mean free path of the gas has been developed by Knudsen[5], equation (2), based upon experimental data where P_a is the average pressure in the tube, λ_a is the mean free path of the gas at the average pressure in the tube, k is Boltzmann's constant, m is the molecular weight of the gas, and L is the length of the tube. Knudsen's equation combines an expression for the conductance ($C = Q/P_a$) of long tubes for molecular flow from kinetic theory, the Poiseuille flow equation for viscous flow, and an interpolating function determined by a fit to experimental data to describe the flow in the transition regime.

$$C = \frac{\pi}{8}\frac{a^4}{\mu}\frac{P_a}{L} + \frac{4a^3}{3L}\sqrt{\left(\frac{2\pi kT}{m}\right)}\left(\frac{1+2.507(a/\lambda_a)}{1+3.095(a/\lambda_a)}\right). \qquad (2)$$

Loyalka and Hamoodi[6] have numerically solved the linearized Boltzmann equation and their results are compared in Figure 1 with Knudsen's fit, as well as that predicted from the slip flow model. Over the range indicated in the graph, the values obtained from Knudsen and from Loyalka and Hamoodi have maximum deviations of a few percent while the slip flow model tends to underpredict the conductance by approximately 20% in the molecular flow limit. Loyalka and Hamoodi's results also predict the existence of a minimum in the conductance of tubes with small pressure gradients, first described by Knudsen[5] and since confirmed by others[7,8]. While the numerical solution of the linearized Boltzmann[6] equation gives good results, application of these techniques for gases at high densities is computationally limited.

Thus far, only the situation where small pressure gradients exist along a tube have been examined. It was proposed by Guthrie and Walkerling[13] that Knudsen's model could be applied to tubes where high pressure differentials exist by applying equation (2) to successive elements along the tube. The resulting expression for the flow rate, corrected by Ochert and Steckelmacher[14], is given in equation (3) for the case where the pressure gradient is very large and the downstream pressure can be neglected:

$$Q = \frac{1}{L}\left(\frac{g}{2}P_a^2 + \frac{hc}{f}P_a + \frac{h(f-h)}{f^2}\ln(1+fP_a)\right) \qquad (3)$$

where:

$$g = \frac{\pi}{8}\frac{a^4}{\mu}$$

$$h = \frac{4a^3}{3}\sqrt{\left(\frac{2\pi kT}{m}\right)}$$

$$c = \sqrt{\left(\frac{m}{kT}\right)}\left(\frac{2aP_a}{\mu}\right)$$

$$f = 1.24\sqrt{\left(\frac{m}{kT}\right)}\left(\frac{2aP_a}{\mu}\right).$$

Examination of this equation reveals that all of the summed terms are positive and monotonic for any value of pressure, hence mean free path, and that the observed minimum of the flow rate or conductance seen in tubes with small pressure differentials is not present in this expression.

To model the flow through tubes with high pressure differentials, it would be advantageous to know the relationship between the conductance of the tube and some predictor, such as the mean free path of the gas. It is unclear from the literature which functional form best applies to the described problem, although the Knudsen form seems to be the most widely applied.

5. Conductance measurement results

The results of the conductance measurements for crimped capillary NBSL48 are shown in Figure 2 as a function of the inverse

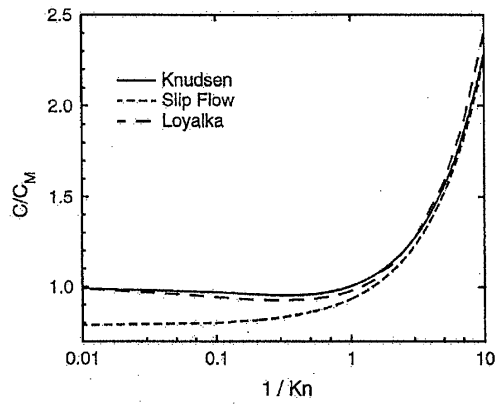

Figure 1. The ratio of the conductance C to the molecular conductance C_M as a function of the reciprocal Knudsen number ($Kn = \lambda/a$). Loyalka and Hamoodi's curve is generated from tabular data[6].

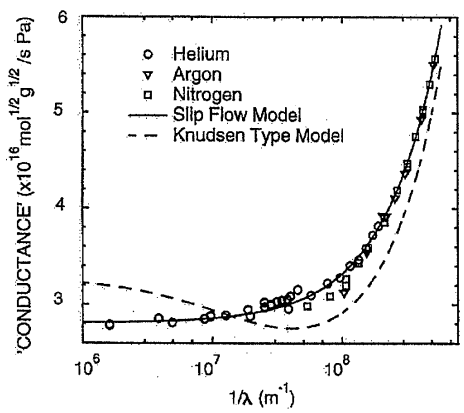

Figure 2. The scaled conductance of crimped tube NBSL48 as a function of the inverse mean free path. The slip flow model assumes total diffuse molecular scattering ($f_s = 1$). The model of Guthrie and Walkerling, not shown, gave results identical to the slip flow model.

mean free path* of the gas at the high pressure end of the tube. The conductances for the individual gas species helium, argon, and nitrogen, have been normalized by multiplying by the square root of the molecular weight of the gas. As can be seen from the figure, the conductances of the individual gas species scale predictably with the mean free path of the gas. The solid line represents a fit to the data using the slip flow model, equation (1), described earlier, with $f_s = 1$ and with the radius of the tube a parameter in the fit. The residuals of the fit are given in Figure 3. The standard deviation of the residuals is 1.8%. A fit of equation (3), Guthrie and Walkerling's model, to the data degenerated to the same form as the slip flow model with identical coefficients. A fit of the Knudsen equation to the data gave poor results, as shown in Figure 2. The mean free paths indicated in the figures correspond to a pressure range of 20 kPa–2 MPa over which the leak rates ranged from 10^{-12} to 10^{-9} mol s^{-1}. The data show no minimum in the conductance over the measured range.

The results of the conductance measurements for crimped capillary NBSL44 are shown in Figure 4 as a function of the inverse mean free path of the gas at the high pressure end. The conductances for the argon and helium show good agreement over the range of mean free paths where they overlap. The slip flow fit to the data with $f_s = 1$ gave a standard deviation of the residuals of 1.1%. The mean free paths indicated in Figure 4 correspond to a pressure range of 0.2 –2 MPa over which the leak rates ranged from 10^{-9} to 10^{-7} mol s^{-1}. The data show no minimum and the slip flow fit represents the data well; to within the uncertainties of the measurements.

The measured conductance of the constant diameter metal capillary tube is shown in Figure 5. The conductances for the individual gas species are shown to scale predictably with the mean free path of the gas. The data show a minimum in the conductance for an inverse mean free path value of approximately 2×10^5 m^{-1}. The mean free paths indicated in Figure 5 correspond to a pressure range of 100 Pa–0.4 MPa over which

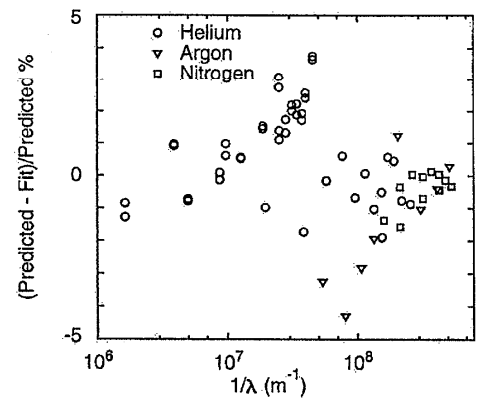

Figure 3. Residuals of the slip flow fit to crimped tube NBSL48 as a function of the inverse mean free path.

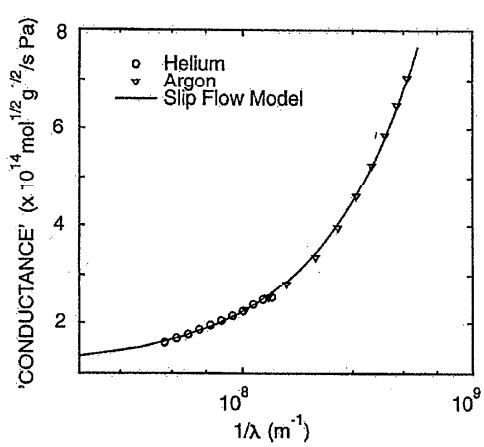

Figure 4. The scaled conductance of crimped tube NBSL44 as a function of the inverse mean free path. The slip flow model assumes total diffuse molecular scattering ($f_s = 1$). The model of Guthrie and Walkerling, not shown, gave results identical to the slip flow model.

*$\lambda = 116.4(\mu/P)(T/m)^{1/2}$ where λ is the mean free path, m; μ is the gas viscosity, Pa s; P is the absolute pressure of the gas, Pa; T is the absolute gas temperature, K; and m is the molecular weight of the gas, g mol^{-1}.

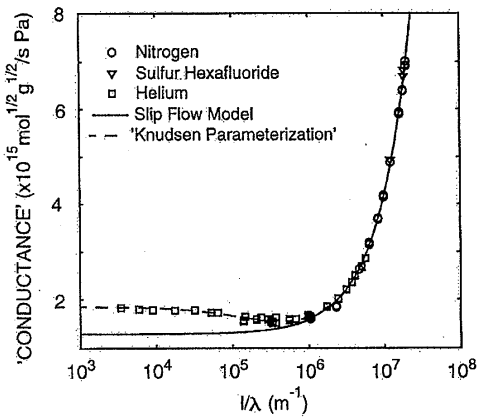

Figure 5. The scaled conductance of a constant diameter tube as a function of the inverse mean free path. The slip flow model assumes total diffuse molecular scattering ($f_s = 1$). The 'Knudsen Parameterization' is calculated from equation (4).

the leak rates range from 10^{-14} to 10^{-10} mol s^{-1}. The slip flow fit represents the data well for inverse mean free path values above 10^6 m^{-1}; however, the slip flow fit underpredicts the conductance of the tube by 40% in the molecular flow limit which is similar to the case for tubes with small pressure differentials, Figure 1. A fit of the data to Guthrie and Walkerling's model degenerated to the same form as the slip flow model with similar coefficients. The data were also fit to a modified form of the Knudsen equation as given in equation (2):

$$C = ba^3\left(0.1472a/\lambda + \frac{1+3.50a/\lambda}{1+5.17a/\lambda}\right) \tag{4}$$

where

a = tube radius

$$b = \frac{4}{3L}\sqrt{\left(\frac{2\pi kT}{m}\right)}.$$

The constants in the second term of the equation were allowed to vary in order to minimize the residuals and to predict the larger minimum in conductance observed in these measurements which is larger than that of the Knudsen equation which was developed for tubes with small pressure differentials. The constants, a and b, are used as fitting parameters to determine the effective tube radius and length. The product ba^3 is the molecular conductance. The effective radius of the tube was determined to be 1.08 μm and the length of the tube 2 mm. These dimensions are of the correct order of magnitude, although the actual dimensions have not been precisely measured. The fit is represented in Figure 5 as the 'Knudsen Parameterization'.

Wheareas the slip flow and the Guthrie and Walkerling models seem to adequately represent the conductance data for the crimped capillary leak over the tested range, large deviations from these models are seen for the constant diameter capillary. It is also observed that while the crimped metal capillary tube shows no minimum in the conductance, the constant diameter capillary shows a pronounced minimum. This seems to agree with the qualitative analysis of Pollard and Present[15], which predicts that there should not be a minimum in tubes for which the diameters vary widely along the tube length or for conductances with irregular geometries such as porous media. From the qualitative arguments given by Pollard and Present as to the origin of the minimum in the conductance for the case of small pressure differentials, one would expect that a minimum would also be present in the high pressure differential case. In any event, integration of an empirical, or semi-empirical equation, as in the case of the Guthrie and Walkerling model, to predict the conductance of tubes with high pressure differentials may lead to significant errors and any agreement is merely fortuitous. For both types of stainless steel capillary leaks, the conductance is observed to vary uniformly with the mean free path of the gas, and deviations between gas species due to specular reflections are not noticeable. The ability of the slip flow fit to predict the conductance of the crimped capillary tubes down to the molecular limit is fortuitous and impossible to model theoretically due to the unknown geometry of the leak. The 'Knudsen Parameterization' given in equation (4) models the conductance of the constant diameter tube well and gives reasonable estimations of the capillary length and diameter. Other fits may represent the data equally well, but the 'Knudsen Parameterization' has the nice properties of reducing to the molecular conductance for very large values of the mean free path, and to the viscous conductance for very small values of the mean free path.

6. Conclusions

The conductances of two types of stainless steel tubes with large pressure differentials have been investigated to model their flow as a function of the mean free path of a gas species. The data show that the crimped metal capillary leaks are adequately described using a slip flow model over the range investigated. The conductance of the constant diameter tube shows a distinct minimum similar in form to previous experimental and theoretical models for capillary tubes with small differential pressures. Neither the slip flow model nor Guthrie and Walkerling's expression for flow through tubes with large pressure differentials adequately represent the form of the observed data for the constant diameter tube. An empirical fit with a form similar to Knudsen's has been found to adequately describe the conductance of this particular capillary tube. Its general ability to preduct the conductance of tubes with high pressure differentials in the transition range, even in form, has not been verified. Based upon these preliminary data, tubes with nonuniform geometry seem to be preferable as leak elements due to their ability to be adequately modeled with a simple form over a wide pressure range.

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