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Revisiting Maxwell's accommodation coefficient: A study of nitrogen flow in a silica microtube across all flow regimes

Wenwen Lei, David R. McKenzie *

School of Physics, University of Sydney, Sydney, 2006, Australia

HIGHLIGHTS

- First experimental study on flow rate across all flow regimes in a well-defined microtube.
- Extend Cha and McCoy theory for molecular flow regime.
- Demonstrate the Maxwell accommodation coefficient is different in the slip and molecular flow regimes.

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ABSTRACT

Gas flows have been studied quantitatively for more than a hundred years and have relevance in modern fields such as the control of gas inputs to processes, the measurement of leak rates and the separation of gaseous species. Cha and McCoy have derived a convenient formula for the flow of an ideal gas applicable across a wide range of Knudsen numbers (Kn) that approaches the Navier–Stokes equations at small Kn and the Smoluchowski extension of the Knudsen flow equation at large Kn. Smoluchowski's result relies on the Maxwell definition of the tangential momentum accommodation coefficient α, recently challenged by Arya et al. We measure the flow rate of nitrogen gas in a smooth walled silica tube across a wide range of Knudsen numbers from 0.0048 to 12.4583. We find that the nitrogen flow obeys the Cha and McCoy equation with a large value of α, unlike carbon nanotubes which show flows consistent with a small value of α. Silica capillaries are therefore not atomically smooth. The flow at small Kn has $\alpha = 0.91$ and at large Kn has α close to one, consistent with the redefinition of accommodation coefficient by Arya et al., which also resolves a problem in the literature where there are many observations of α of less

* Corresponding author. Tel.: +61 293515986.
E-mail addresses: wlei@physics.usyd.edu.au (W. Lei), d.mckenzie@physics.usyd.edu.au (D.R. McKenzie).

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than one at small Kn and many equal to one at large Kn. Silica capillaries are an excellent choice for an accurate flow control system.

1. Introduction

Calculation and accurate control of the flow of gases in microtubes are important in many fields. In biology and botany, air flows in nanodimensioned tubes are of fundamental importance in respiration and transpiration. Gas flows in microfluidic devices are used for sensing and many process control applications require an accurate knowledge of the flow of a gas. Nitrogen has been selected for the current work as it is representative of an ideal gas, it is the majority constituent of air and the control of its flow is required in many processes.

The use of a capillary for obtaining a calibrated gas flow rate has the advantage that the kinetic properties rather than the thermal properties of the gas are important. Such a method would use a capillary of known dimensions to control the flow by means of the pressure difference across it. The only property of the gas needed to specify the flow rate is then the molecular mass.

The Knudsen number Kn, is defined as the ratio of the mean free path to the tube characteristic dimension:

$$Kn = \frac{\lambda}{D} \tag{1}$$

where λ is mean free path and D is a characteristic dimension, conventionally the diameter of a cylindrical tube. The value of Kn specifies the flow regime. For $Kn < 0.01$, the continuum flow regime applies, where the gas behaves as a viscous fluid with a non-slip boundary condition at the walls. Non-slip flow is described well by the Navier–Stokes equations that give the Poiseuille law for the flow rate [1]. The range $0.01 < Kn < 0.1$ defines the slip flow regime where the boundary conditions change from non-slip to slip. An additional parameter, such as the accommodation coefficient introduced by Maxwell, is needed to specify the boundary conditions. For $0.1 < Kn < 10$ defines the transition flow regime where the existence of molecules needs to be taken into account and the accurate first principles description of flow requires a solution of the Boltzmann equation. $Kn > 10$ defines the molecular flow regime in which the interactions between molecules are weak and relatively simple kinetic theory models apply. Knudsen derived a simple formula for the flow of an ideal gas assuming that the molecules all suffer diffuse reflection at the walls, a formula that has been found to describe the results of experiment for many ideal gases very accurately [2]. Smoluchowski introduced a modification of Knudsen's model soon after that included the effect of the Maxwell tangential momentum accommodation coefficient α in the molecular flow regime [3]. The definition of α is based on the Maxwell hypothesis that a fraction of reflections are fully diffuse while the remainder is fully specular. A value of α less than one gives a flow rate higher than the Knudsen value, with the flow rate diverging to infinity as α tends to zero. Until recently, there have been no well authenticated cases of Smoluchowski's equation being required (that is $\alpha < 1$) instead of Knudsen's equation ($\alpha = 1$) in the molecular flow regime. This is in contrast to the slip flow regime where values of α of less than one are frequently reported. Recent results for air flows in carbon nanotubes have found that the Knudsen equation seriously underestimates the flow in carbon nanotubes in the molecular flow regime, suggesting that the Smoluchowski result may apply with small α. Carbon nanotubes may be exceptional in having atomically smooth walls and diameters less than 2 nm [4]. Meanwhile, the flow of argon and helium in fabricated silicon tubes of similar dimensions has subsequently been found to follow Knudsen's equation even for very high values of Kn up to 10^7 [5].

In order to simplify the calculation of flow across all flow regimes, extended Navier–Stokes equations have been derived which match the Smoluchowski result at large Kn and the Poiseuille result at small Kn and also appear to fit the intermediate regimes well with only one additional