GAS SEPARATION IN RAREFIED BINARY GAS MIXTURE FLOWS THROUGH LONG TAPERED MICROCHANNELS

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Abstract. *The rarefied gas flow of binary gas mixtures through diverging and converging microchannels is investigated based on linear kinetic theory. The analysis is valid in the whole range of the Knudsen number. Indicative results for the molar flow rates and the axial pressure and molar fraction distributions are provided for the gas mixtures of He-Xe and Ne-Ar flowing through tapered channels of specific geometry due to a given pressure difference. The dependency of the computed fluxes and of the induced separation effect on the species of the gas mixture, its molar fraction as well as on the gas-surface interaction is examined and discussed in detail.*

1 INTRODUCTION

Gas mixture flows through long microchannels, due to their theoretical and practical interest in several technological fields including microfluidics and vacuum technology, have been extensively investigated with considerable success. It is well known that in the case of gas flows far from local equilibrium, as in gas microflows, the classical Navier-Stokes-Fourier hydrodynamic equations are not valid and a kinetic approach as described by the Boltzmann equation must be implemented. In this framework, the McCormack kinetic model has been introduced to obtain reliable numerical results for gas mixture flows between plates and through long channels of circular and rectangular cross sections in the whole range of the Knudsen number [1,2,3]. Comparisons with corresponding experimental data have been performed to demonstrate a very good agreement between measurements and computations [4]. In all these binary gas mixture flows, gas separation, which is due to the different molecular velocities of the species of the mixture, is observed [4,5].

In all these cases the channel cross section remains constant. In several application however, such as in vacuum systems or porous media and microfiltering the channel may have a variable cross section. Recently, a methodology has been developed to simulate pressure driven single gas flows far from local equilibrium through circular tubes of variable diameter [6]. Based on the assumption that the tube is sufficiently long compared to its largest diameter the linear BGK model has been applied to deduce accurate results over a wide range of the Knudsen number. Here, this approach is extended to investigate singe gas and binary gas mixtures flowing between diverging and converging plates. This type of flow configuration is common in gaseous micro electromechanical systems and in vacuum gas distribution systems, where the width is much larger of the height of the channel.

2 FLOW CONFIGURATION

Two large reservoirs A and B, maintained at different pressures P_A and P_B, are connected by a long duct of length L and variable rectangular cross section, defined by $A(z^*)=WxH(z^*)$, where W is the width and $H(z^*)$ is the local height. The height is varying in the flow direction z^*, with H_A and H_B denoting the height at the two ends of the channel. The reservoirs are filled by a binary gas mixture and due to the pressure difference there is a gas flow from the high towards the low pressure reservoir. The temperature of the gas in both reservoirs is maintained at T_0. The configuration of the problem is shown in Fig. 1. Flows through both diverging and converging channels are considered. In the first case $P_A>P_B$ and $H(z^*)$ is increased in the flow direction, while in the latter case $P_B>P_A$ and $H(z^*)$ is decreased in the flow direction. To simplify the formulation and the involved computational effort it is assumed that the width W extends to infinity and the flow does not depend on this direction. Furthermore, it is assumed that L>>H and therefore the flow may be simulated by linear kinetic theory ignoring end effects.

The binary gas mixture is defined by the molecular masses m_1, m_2 and number densities n_1, n_2 of each species. Then the molecular mass of the mixture is given by

$$m(C) = Cm_1 + (1-C)m_2 \tag{1}$$

where

$$C(z) = n_1 / n = n_1 / (n_1 + n_2) \tag{2}$$

is the molar fraction specifying the composition of the binary mixture and z=z*/L. The index 1 denotes always the light species. The main flow parameter is the gas rarefaction parameter defined as

$$\delta(z) = \frac{H(z)\, P(z)}{\mu(z)\, \upsilon_0(z)}, \tag{3}$$

where P(z) is the pressure along the channel varying as $P_A \le P(z) \le P_B$, $\mu(z)$ is the gas viscosity at reference temperature T_0 and $\upsilon_0(z)$ is a molecular velocity given by

$$\upsilon_0 = \sqrt{2kT_0 / m} \tag{4}$$

with k denoting the Boltzmann constant. Both μ and υ_0 are varying in the flow direction, since the molar fraction of the mixture is changing. The local pressure is connected to the local number density according to $P(z)=n(z)kT_0$. The rarefaction parameter δ is inversely proportional to the Knudsen number. It is noted that due to the separation effect the molar fraction is varying along the channel, i.e., C=C(z).

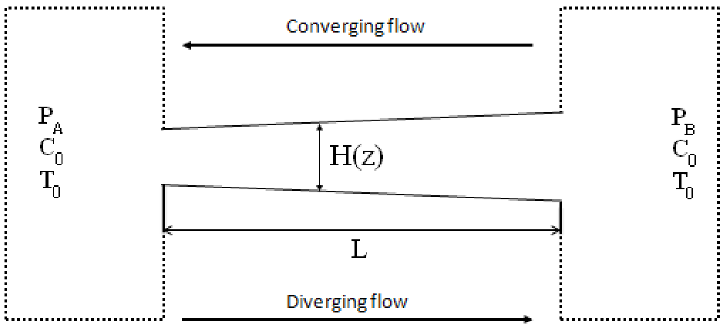

Figure 1. Flow configuration

3 KINETIC FORMULATION

As it is noted in the introduction in general, the solution of gas flows far from local equilibrium requires an implementation of a kinetic approach. Here, the whole approach is divided into two stages. In the first stage the dimensionless flow rates of the each species of the mixture are computed in terms of the local gas rarefaction parameter δ and molar fraction C. An efficiently large kinetic data base is produced. In the second stage based on the mass conservation law and on the kinetic data base, the pressure P(z) and the molar fraction C(z) distributions along the channel as well as the molar flow rates are computed. This methodology has been applied before in [4,5].

In the first stage once the specific binary gas mixture is specified, then the dimensionless flow rates, known in the literature as kinetic coefficients, are obtained in terms of the reference molar fraction C_A and rarefaction parameter δ_A. It is based on the solution of the McCormack kinetic model [7] subject to Maxwell boundary conditions. When the two species of the mixture are assumed to be identical, the McCormack solution is reduced to the corresponding single gas solution based on the Shakhov model. Although this is a pressure driven flow, i.e., the molar fraction at the inlet and outlet reservoirs are equal ($C_A=C_B$), due to the separation effect, a non-uniform molar fraction distribution along the flow is deduced. Therefore, the kinetic data base must include dimensionless flow rates due to pressure and concentration gradients. As a result for each binary gas mixture under consideration the flow rates or kinetic coefficients Λ_{PP}, Λ_{PC}, Λ_{CP} and Λ_{CC} are computed for $\delta_A \le \delta(z) \le \delta_B$ and for various molar fractions close to the reference molar fraction C_A. Since the numerical solution of the McCormack model has been extensively described before no further information is provided here, while all related details may be found in [1- 3].

In the second stage, based on mass conservation and following some routine manipulation it is deduced that [4,5]

$$J_1 = -\frac{1}{2} n(z) H(z)^2 \upsilon_0(z) \left[\left[C(z)\Lambda_{PP} + \left(1 - C(z)\right)\Lambda_{CP} \right] \frac{1}{P(z)} \frac{dP}{dz} + \left[C(z)\Lambda_{PC} + \left(1 - C(z)\right)\Lambda_{CC} \right] \frac{1}{C(z)} \frac{dC}{dz} \right] \tag{5}$$

$$J_2 = -\frac{1}{2} \left(1 - C(z)\right) n(z) H(z)^2 \upsilon_0(z) \left[\left[\Lambda_{PP} - \Lambda_{CP} \right] \frac{1}{P(z)} \frac{dP}{dz} + \left[C(z)\Lambda_{PC} - \Lambda_{CC} \right] \frac{1}{C(z)} \frac{dC}{dz} \right] \tag{6}$$

Here, J_1 and J_2 are the molar flow rates of the light and heavy species respectively, while $J= J_1+J_2$ is the molar flow rate of the mixture. It is noted that these quantities are constant at each cross section. Equations (5) and (6) is a system of two ordinary differential equations to be solved for the unknown distributions P(z) and C(z) subject to the boundary conditions $P=P_A$ and $C=C_A$ at z=0 and $P=P_B$ and $C=C_B$ at z=1. The flow rates J_1 and J_2 will also be part of the solution. In particular, some initial values are imposed on J_1 and J_2 and then the system of ordinary differential equations is integrated starting from the initial conditions at z=0 all the way up to z=1. The computed pressure and concentration at z=1 are compared to P_B and C_B and the whole process is repeated upon convergence. At each integration step, the values of the kinetic coefficients are obtained from the kinetic data base and if needed, some interpolation is introduced. The integration is performed by a typical 4th order Runge-Kutta scheme.

4 RESULTS AND DISCUSSION

Two binary gas mixtures have been applied, namely Ne-Ar with m_l=20.18 g/mol, m_2= 39.95 g/mol and He-Xe, with m_l=4.0026 g/mol, m_2=131.30 g/mol. These mixtures are representative for binary gas mixtures having species with molecular masses which are close to each other and for mixtures consisting of species with quite different molecular masses. The flow characteristics of the first ones are expected to be close to those of a single gas having equivalent molecular mass, while the flow characteristics of the latter ones are dominated by separation effects and are expected to be different than the corresponding ones of single gases.

Results are presented for these two mixtures flowing through the tapered channels with H_A=1.88 μm and H_B=0.376 μm and the inlet and outlet pressure equal to P_{in} =13,200 Pa and P_{out} =2000 Pa respectively (P_{in} /P_{out} =6.6). In the diverging flow $P_A=P_{in}$ and $P_B=P_{out}$, while in the converging flow $P_B=P_{in}$ and $P_A=P_{out}$ (see Figure 1). Based on the imposed data the flow is in the transition regime. The molar fraction C_A varies from zero to one. As it has been noted as C_A is increased the molar fraction of the light species is increased. Therefore the values of C_A= 0 and C_A= 1 correspond to the single gas flow of the heavy and light species respectively, while the intermediate values correspond to the mixture flow.

Channel Type	C_A	δ_A	δ_B	δ_{mean}	J_1 mol/m/s	J_2 mol/m/s	J mol/m/s	J_1/J
Diverging	0	1.10	0.83	0.97	0.00	9.62 (-8)	9.62 (-8)	0.00
	0.1	1.02	0.78	0.90	3.86 (-8)	8.97 (-8)	1.28 (-7)	0.30
	0.3	0.88	0.67	0.78	1.26 (-7)	7.58 (-8)	2.01 (-7)	0.62
	0.5	0.72	0.55	0.63	2.30 (-7)	5.98 (-8)	2.90 (-7)	0.79
	0.7	0.55	0.42	0.48	3.62 (-7)	4.07 (-8)	4.02 (-7)	0.90
	0.9	0.38	0.28	0.33	5.30 (-7)	1.62 (-8)	5.46 (-7)	0.97
	1	0.22	0.17	0.19	6.43 (-7)	0.00	6.43 (-7)	1.00
Converging	0	0.17	5.50	2.86	0.00	1.03 (-7)	1.03 (-7)	0.00
	0.1	0.16	5.12	2.67	3.42 (-8)	9.66 (-8)	1.31 (-7)	0.26
	0.3	0.13	4.41	2.31	1.13 (-7)	8.31 (-8)	1.96 (-7)	0.58
	0.5	0.11	3.61	1.90	2.11 (-7)	6.71 (-8)	2.78 (-7)	0.76
	0.7	0.08	2.74	1.45	3.40 (-7)	4.72 (-8)	3.87 (-8)	0.88
	0.9	0.06	1.88	0.98	5.25 (-7)	2.00 (-8)	5.45 (-7)	0.96
	1	0.03	1.11	0.56	6.71 (-7)	0.00	6.71 (-7)	1.00

Table 1 : Molar fluxes of He-Xe flow in diverging and converging channels for P_{out}=2000 Pa, P_{in}/ P_{out}=6.6 and various C_A.

In Tables 1 and 2 the molar flow rates of He-Xe and Ne-Ar are provided respectively for both converging and diverging channels and various flow parameters. In particular, in the first column of the tables the tapered channel type is specified. In the second column the value of C_A is given. In the next three columns the gas rarefaction parameters δ_A and δ_B at the two edges of the channel, as well as the mean valued, denoted by δ_{mean} are provided. Since δ is not varying linearly, δ_{mean} is found by integrating the gas rarefaction parameter between δ_A and δ_B. Next, in the sixth, seventh and eighth columns the computed molar flow rates J_1, J_2 of the light and heavy species respectively and the total molar flow rate $J= J_1+ J_2$ are tabulated. Finally, in the last column the ratio J_1/J is given.

It is seen that in both mixtures and channel types as the molar fraction C_A is increased, the molar flux of the light species J_1 is increased and the molar flux of the heavy species J_2 is decreased, while the total molar flux J is increased. In addition, the total molar flux of the mixture varies monotonically between the minimum molar flux of the heavy species at C_A= 0 and the maximum molar flux of the light species at C_A= 1. All these are well justified by the fact that the speed of the light molecules is higher than that of the heavy molecules. This difference is large in the case of He and Xe species and small in the case of Ne and Ar species. Also, in both mixtures the total molar fluxes are slightly higher in the converging channels compared to the corresponding ones in the diverging channels. This is due to the fact that the converging flow is slightly less rarefied than the corresponding diverging flow as it can be seen by the values of the mean gas rarefaction parameter δ_{mean}.

The computed values of the ratio J_1/J is a clear indicator of the presence of the separation phenomena. It is seen that these values are always larger than the corresponding C_A values. This means that compared to the reference molar fraction a higher number of light particles have been conveyed through the channel. The smaller C_A is, the larger the departure of J_1/J from the reference molar fraction C_A becomes. In both channel types the separation phenomena is about equally dominant.

Channel Type	C_A	δ_A	δ_B	δ_{mean}	J_1 mol/m/s	J_2 mol/m/s	J mol/m/s	J_1/J
Diverging	0	0.62	0.47	0.54	0.00	1.79 (-7)	1.79 (-7)	0.00
	0.1	0.58	0.44	0.51	2.33 (-8)	1.63 (-7)	1.87 (-7)	0.12
	0.3	0.52	0.39	0.45	7.21 (-8)	1.31 (-7)	2.03 (-7)	0.36
	0.5	0.46	0.35	0.40	1.24 (-7)	9.66 (-8)	2.21 (-7)	0.56
	0.7	0.40	0.30	0.35	1.80 (-7)	6.00 (-8)	2.40 (-7)	0.75
	0.9	0.34	0.26	0.30	2.40 (-7)	2.08 (-8)	2.61 (-7)	0.92
	1	0.31	0.24	0.27	2.72 (-7)	0.00	2.72 (-7)	1.00
Converging	0	0.09	3.08	1.58	0.00	1.88 (-7)	1.88 (-7)	0.00
	0.1	0.09	2.91	1.49	2.32 (-8)	1.72 (-7)	1.95 (-7)	0.12
	0.3	0.08	2.59	1.33	7.23 (-8)	1.39 (-7)	2.11 (-7)	0.34
	0.5	0.07	2.28	1.16	1.26 (-7)	1.03 (-7)	2.29 (-7)	0.55
	0.7	0.06	1.98	1.01	1.84 (-7)	6.48 (-8)	2.49 (-7)	0.74
	0.9	0.05	1.69	0.86	2.48 (-7)	2.27 (-8)	2.71 (-7)	0.92
	1	0.05	1.55	0.79	2.84 (-7)	0.00	2.84 (-7)	1.00

Table 2 : Molar fluxes of Ne-Ar flow in diverging and converging channels for P_{out}=2000 Pa, P_{in}/P_{out}=6.6 and various C_A.

Next, the pressure and molar fraction distributions along the diverging and converging channels are shown in Figures 2 and 3 respectively. As it is seen in Figure 2 the pressure distribution between the inlet and the outlet of the channel is not linear as it commonly happens in the case of channels of constant cross section. Here, for diverging channels the pressure drops rapidly in the first one-third of the channel and then in the remaining two-thirds slowly decreases to the outlet value. The situation is opposite in the case of converging channels, where the pressure decreases slowly in the first two-thirds of the channel and then in the last one-third is rapidly dropped to the outlet value. It is also seen that the effect of the molar fraction C_A on the pressure distributions is negligible in the case of diverging channels and very small in the case of converging channels.

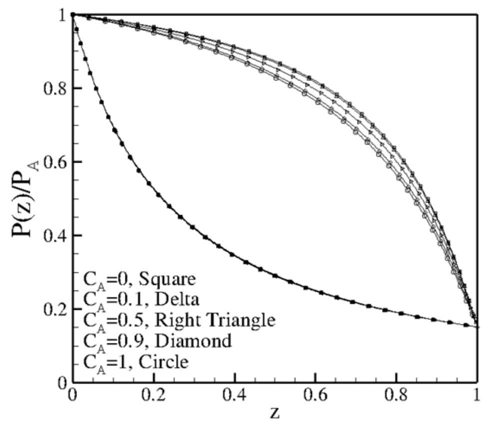

Figure 2. Dimensionless distribution of the pressure for He-Xe flows along the diverging (filled symbols) and converging (empty symbols) channel.

In Figure 3, the normalized molar fraction distribution for diverging and converging flow along the channel is shown for the specific case of C_A= 0.5. In both cases the molar fraction along the channel does not remain constant. Initially it drops from its inlet value up to some certain distance from the inlet and then it is increased to reach the outlet value at the exit. In the diverging channel this variation is almost symmetric around x=0.5. In the converging channel the molar fraction is decreased in the larger part of the channel, it is reaching its minimum value around x=0.85-0.95 and then it is increased to reach the outlet value. The variation on a percentage base is much larger, about one order of magnitude, in the converging flow compared to the one in the diverging flow. Also, in both cases as the molar fraction is increased the deviation from the reference molar fraction is decreased, which is in agreement with the results tabulated in the last columns of Table 1.

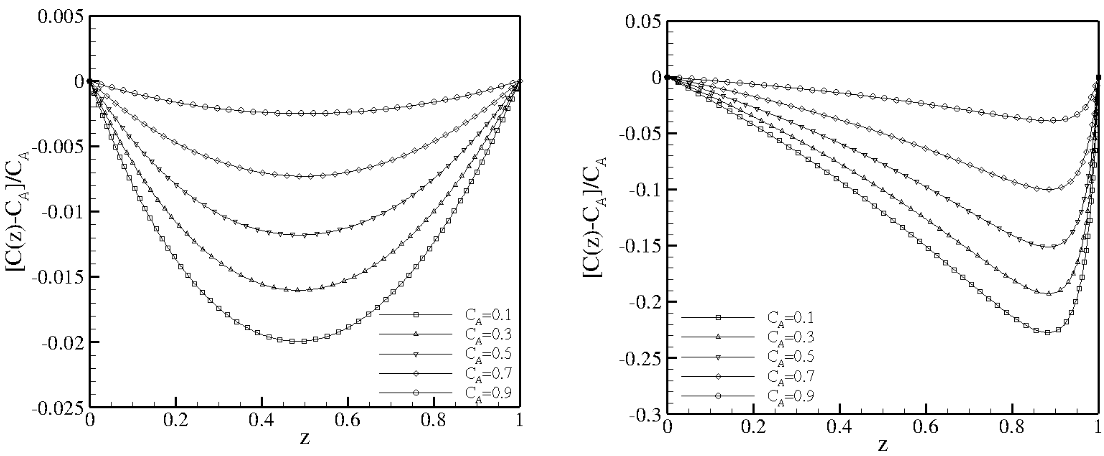

Figure 3. Normalized distribution of the molar fraction for He-Xe flows along the diverging (left) and converging (right) channel.

Finally, in Table 3 the molar fluxes of the species and of the mixture are presented for various values of the accommodation coefficient α, which describes the type of gas-surface interaction. The case α=1 corresponds to purely diffuse scattering, while the cases of α<1 correspond to diffuse and specular scattering. In the latter case only the α portion of the particles reflect diffusely, while the remaining (1- α) reflect specularly. As it is seen in all cases as α is decreased the molar fluxes are increased since the wall drag is reduced. More important, the separation effect is more dominant when the light species has a smaller accommodation coefficient compared to the heavy species. That means that following some specialized surface treatment to increase the specular reflection only of the light species may result to an increase of the separation between the gases and vice versa.

Channel Type	α_{He}	α_{Xe}	J_1 mol/m/s	J_2 mol/m/s	J mol/m/s	J_1/J
Diverging	1	1	2.30 (-7)	5.98 (-8)	2.90 (-7)	0.79
	0.7	0.7	3.22 (-7)	9.66 (-8)	4.19 (-7)	0.77
	1	0.7	2.38 (-7)	8.91 (-8)	3.28 (-7)	0.73
	0.7	1	3.10 (-7)	6.46 (-8)	3.75 (-7)	0.83
Converging	1	1	2.11 (-7)	6.71 (-8)	2.78 (-7)	0.76
	0.7	0.7	2.83 (-7)	1.08 (-7)	3.92 (-7)	0.72
	1	0.7	2.23 (-7)	9.87 (-8)	3.22 (-7)	0.69
	0.7	1	2.66 (-7)	7.32 (-8)	3.40 (-7)	0.78

Table 3 : Molar fluxes of He-Xe for diverging and converging channels for P_{out}=2000 Pa, P_{in}/ P_{out}=6.6, C_A=0.5 and various values of the accommodation coefficients.

5 CONCLUDING REMARKS

The non-equilibrium flow of binary gas mixtures through tapered channels is examined. Both flows through diverging and converging channels are considered. This type of flow are present in gaseous systems with very small dimensions or low densities or both. Typical examples may be found in GasMEMS and vacuum systems. Since the flow may be in the whole range of the Knudsen number a kinetic methodology based on the McCormack kinetic model subject to Masxwell diffuse-specular gas-surface interaction model is introduced. Indicative results for the molar flow rates and the axial pressure and molar fraction distributions are provided for the gas mixtures of He-Xe and Ne-Ar flowing through tapered channels of specific geometry due to a given pressure difference. The gas separation effect is also conidered. It is hoped that the present work may be useful in the design and optimization of such systems.

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