A Comparative Study on the Molecular- and Continuum-Based Simulation Methods for Gas Flow and Mixing in Micro/Nanochannels

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Abstract - A comparative study is conducted between the molecular and continuum approaches to treat the gas flow as well as the mixing problem in micro/nanoscale channels. The molecular-based simulations are performed using the direct simulation Monte Carlo DSMC method; however, the continuum-based simulations are accomplished using the finite-volume FV method incorporating suitable slip/jump boundary conditions for the gas mixture flows. Employing these two methods, we simulate the mixing process of two gases in mixers with different micro and nano scale sizes working in both slip and transitional flow regimes. The comparisons are provided for the corresponding results of these two methods including their flow velocities, mass flow rates, species mass fractions, and diffusion fluxes. Using the outcome of the performed comparisons, we eventually describe the effects of rarefaction on the achieved accuracy of the continuum-based method.

Keywords: Gas mixing, Rarefaction, Micro/nanochannels, Molecular simulation, Continuum simulation, DSMC, CFD, Finite-volume

1. Introduction

The simulations of micro/nanoscale flows can be considered as an active research filed because of the new developments and applications of the micro/nanoelectromechanical systems (MEMS/NEMS). The special attention is paid to these flows because of difficulties associated with modeling their physics. The simulation methods based on the classical continuum-based governing equations and the associated boundary conditions can accurately describe the heat and mass transfer phenomena in macroscale flow problems. However, it is well known that their accuracy would be degraded as the characteristic length scale of the flow, which reduces to the levels comparable with the molecular mean free path (λ) of the gas. So, the Knudsen number (Kn), defined as the ratio of λ to the characteristics length of flow, is usually regarded as a measure of inaccuracy of classical continuum-based simulation approaches.

A common, but not generally, applicable classification of the flow regimes based on the Knudsen number (Kn) is considered as follows [1]. The flows with Kn<0.001 are considered as continuum flows, and the classical continuum-based models can describe them accurately. The flows with the Kn in the range of 0.001 to 0.1 are referred to as slip flows. The continuum-based models can still be applied for these flows providing that appropriate slip/jump models are incorporated at the solid boundaries. The flows with Kn>10 are known as free-molecular flows and can be described using the free-molecular models. The flows in the Kn range of 0.1 to 10 are referred to as the transition flows. The continuum-based models have poor accuracies in describing this flow regime and the Boltzmann equation must be used to describe their physics. The direct simulation Monte Carlo (DSMC) [2] is a molecular based method that is vastly used in simulation of these flows instead of the solution of the Boltzmann equation.

The DSMC method can be applied to the whole Kn range provided that the gas is dilute. However, its high computational cost restricts its usage in high Kn flow regimes. Moreover, the continuum-based simulations may have

enough accuracy for low Kn flows if attention is paid to accurate modeling of the near wall discontinuities. As a result, it is desirable to perform studies on the extents of the validity of slip/jump corrected continuum-based solutions.

There are many research papers, which present the comparisons between the results of continuum-based methods incorporating slip/jump boundary conditions with the molecular-based methods for gas flow problems. However, such comparative studies are less performed on the mass transfer in gas mixture flows. Qazi Zade et al. [3] presented a comparison between the continuum-based and molecular-based methods for the flow of H_2/N_2 and $H_2/N_2/CO_2$ mixtures in planar microchannels to verify the accuracy of slip/jump boundary conditions developed for multicomponent flows [4]. In the present work, we are going to perform a comparison between the DSMC results and the results of the continuum-based methods incorporating suitable slip/jump boundary conditions for the gas mixing flows through micro/nanochannels.

2. Numerical Method

2.1. Direct Simulation Monte Carlo

Direct simulation Monte Carlo (DSMC) method [2] is a molecular-based method introduced by G. A. Bird for the simulation of rarefied gas flows. Nowadays, it is widely used to simulate the gas flow in micro-geometries. A brief description of the DSMC algorithm is as follows. As the first step to this method, the flow domain is divided into a number of computational cells. A number of simulated molecules is distributed in each cell based on suitable macroscopic initial conditions. Each simulated molecule would represent a specific number of real molecules. At each time step, the simulated molecules are moved according to their velocity characteristics. Those molecules, which collide with the walls, would be reflected into main domain with a velocity determined by using suitable wall reflection models; however, those exiting the open boundaries are removed from the simulation domain. New molecules would be injected from the open boundaries into the domain to maintain the desired stationary flow/state boundary conditions. Afterwards, each molecule is attributed to its new cell. The collision partners are determined stochastically among the molecules within each cell. The post-collision velocities of collision partners are determined using an appropriate collision model. After taking sufficient time steps, in which the effects of initial conditions are eliminated and a steady state mean flow is established, the sampling should be started. The microscopic properties of the molecules are sampled in each cell after each certain number of time steps during the sampling period. The macroscopic properties are then calculated by taking suitable time-averages of the collected sampled data. The time step advancement and the sampling are continued until the statistical errors are damped sufficiently out. In this work, we use an extended version of the dsmcFoam simulation code [5] to perform the current molecular-based simulations.

2.2. Computational Fluid Dynamics

The governing equations for the compressible flow of gas mixtures are as follow:

$$\frac{\partial}{\partial t}(\rho) + \frac{\partial}{\partial x_k}(\rho \boldsymbol{u}_k) = 0 \tag{1}$$

$$\frac{\partial}{\partial t}(\rho \boldsymbol{u}_i) + \frac{\partial}{\partial x_k}(\rho \boldsymbol{u}_k \boldsymbol{u}_i - \boldsymbol{\tau}_{ki} + p\boldsymbol{\delta}_{ki}) = 0 \tag{2}$$

$$\frac{\partial}{\partial t}\left[\rho\left(e + u^2/2\right)\right] + \frac{\partial}{\partial x_k}\left[\rho \boldsymbol{u}_k\left(h + u^2/2\right) + \sum_j \boldsymbol{J}_{cjk} h_j - \boldsymbol{\tau}_{ki}\boldsymbol{u}_i + \boldsymbol{q}_k\right] = 0 \tag{3}$$

$$\frac{\partial}{\partial t}(\rho Y_i) + \frac{\partial}{\partial x_k}(\rho Y_i \boldsymbol{u}_k + \boldsymbol{J}_{ik}) = 0 \tag{4}$$

in which, δ is the Kronecker delta. The calorically perfect gas relations are used to calculate the pressure and enthalpy as follows:

$$p = \rho RT \tag{5}$$

$$h = e + p/\rho = \frac{\gamma RT}{\gamma - 1} \tag{6}$$

In these relations, R and γ are the gas constant and specific heat ratio, respectively. The constitutive relations describing the stress tensor and heat conduction flux vector are as follows:

$$\boldsymbol{\tau}_{ij} = -\mu \left(\frac{\partial \boldsymbol{u}_i}{\partial x_j} + \frac{\partial \boldsymbol{u}_j}{\partial x_i} \right) + \frac{2}{3} \mu \frac{\partial \boldsymbol{u}_k}{\partial x_k} \boldsymbol{\delta}_{ij} \tag{7}$$

$$\boldsymbol{q}_j = -k \frac{\partial T}{\partial x_j} \tag{8}$$

The generalized Fick's Law is used to calculate the diffusive mass flux using; as proposed by Curtiss [6],

$$\boldsymbol{J}_i = -\rho_i \sum_j \tilde{D}_{ij} (\nabla X_j + (X_i - Y_i) \frac{\nabla P}{P}) - D_i^T \nabla (\ln T) \tag{9}$$

where $\tilde{D}$ is the symmetric diffusion coefficient matrix. The relation of $\tilde{D}$ with the binary diffusion coefficient matrix $\mathcal{D}$ is presented in Ref. [6] and is not repeated here. D_i^T is the thermal diffusion coefficient, which satisfies the condition $\sum_i D_i^T = 0$. In the present work, we use the approximate relations presented by Cowling [7] and Fristrom and Monchick [8] to calculate the thermal diffusion coefficients.

To solve the above set of governing equation, we use an extended version of the rhoCentralFoam solver of the OpenFOAM simulation package. The rhoCentralFoam is a finite volume solver that exploits semi-discrete, non-staggered central schemes for collocated variables [9]. It is originally developed for the simulation of compressible flow of the simple gases or the homogeneous gas mixtures. We have extended this solver to solve the species transport equations and to account for the diffusion terms in the energy equation. For the sake of more consistency with the DSMC results, we add the calculation of transport properties using the gas kinetic theory relations for the VSS collision model to this solver. The first-order approximation of the coefficient of viscosity for a simple gas obeying the VSS collision model is given as follows [10]:

$$\mu = \frac{5(\alpha + 1)(\alpha + 2)}{4\alpha} \frac{\sqrt{m_s k_B T_{ref}/\pi}}{(5 - 2\omega)(7 - 2\omega)d_{ref}^2} \left(\frac{T}{T_{ref}} \right)^\omega \tag{10}$$

Also, the thermal conductivity of the simple gas can be approximated using the Euken relation [10] as follows:

$$k = \left(\frac{15}{2} + \zeta \right) \frac{\mu k_B}{2m_s} \tag{11}$$

in which, ζ is the number of internal degrees of freedom of the gas molecule.

In this work, we use the following approximations for the coefficient of viscosity [11] and thermal conductivity [12] of gas mixtures:

$$A = \sum_i A_i \left[1 + \sum_{j \neq i} \varphi_{ij} X_j / X_i \right]^{-1} ; \quad A : \mu \text{ or } k \tag{12}$$

In this relation, φ_{ij} is a weight function with the following form

$$\varphi_{ij} = \frac{1}{2\sqrt{2}}\left(1+\frac{w_i}{w_j}\right)^{-1/2}\left[1+\left(\frac{A_i}{A_j}\right)^{1/2}\left(\frac{w_j}{w_i}\right)^{1/4}\right]^2 ; \quad A: \mu \text{ or } k \tag{13}$$

in which, w_i is the molar weight of the ith constituent of the mixture.

We implement a simplified version of the slip/jump boundary condition developed by Qazi Zade et al. [3], [4] for the nonreactive flows to the current solver. For a horizontal wall, the simplified boundary condition can be written in the following form:

$$u_s = \left\{\frac{\sigma-2}{\sigma}\sqrt{\frac{\pi}{2\bar{w}\bar{R}T}}\,\tau_{xy} + \frac{1}{5\rho\bar{R}T}\sum_i\frac{\rho_i K_i}{\sqrt{w_i}}\frac{\partial T}{\partial x} - \sum_i\frac{J_{ix}}{\sqrt{w_i}}\right\}\Bigg/\left\{\sum_i\frac{\rho_i}{\sqrt{w_i}}\right\} \tag{14}$$

$$T_s - T_w = \left[\left\{\frac{2-\sigma}{\sigma}\sqrt{\frac{\pi}{2R_u T_s}}\frac{1}{\rho\bar{R}}\sum_i\frac{\rho_i K_i}{w_i}\right\}\Bigg/\left\{\frac{\bar{\gamma}+1}{4(\bar{\gamma}-1)}\sum_i\frac{\rho_i}{\sqrt{w_i^3}}\right\}\right]\frac{\partial T}{\partial y} + \frac{1}{R_u}\left\{\sum_i\frac{\rho_i}{\sqrt{w_i}}\Bigg/\sum_i\frac{\rho_i}{\sqrt{w_i^3}}\right\}\frac{\bar{\gamma}-1}{\bar{\gamma}+1}u_s^2 \tag{15}$$

in which, σ is the fraction of diffuse molecule-wall collisions that is assumed to be the same for all species in the mixture to derive the simplified boundary conditions. In the present work, we assume that the walls are fully diffuse. In other words, we assume that σ has a unity value. R_u is the universal gas constant and its overbar indicates an average value for the mixture.

3. Problem Description

We investigate the mixing of two gas streams coming from two adjacent channels having different nano/microscale widths. These two streams are gradually mixed through a channel called the mixing channel. The schematic of this mixer is shown in Fig. 1. We apply an inlet pressure of 200 kPa, an outlet pressure of 100 kPa, an inlet temperature of 300 K, and a wall temperature of 300 K as the boundary conditions in this problem. We consider the mixing of N_2 and CO gas streams in this study. The N_2 and CO streams enter the mixer from the top and bottom inlets, respectively. To investigate the effects of length scale on the simulation results, we consider different inlet channels widths of H_{in}=0.04, 0.1, 0.2, 0.5, and 1.0 µm. Using the relation for VSS collision model [2], the mean free path would be about 0.056 µm for a mixture of N_2 and CO with a 50%-50% molar composition at a pressure of 100 kPa and a temperature of 300 K. Choosing the inlet height, H_{in}, as the characteristic length, the Knudsen number values would be 0.056, 0.11, 0.28, 0.56, and 1.4 for the above mixer sizes, respectively. Therefore, our study suitably covers the slip and transitional flow regimes.
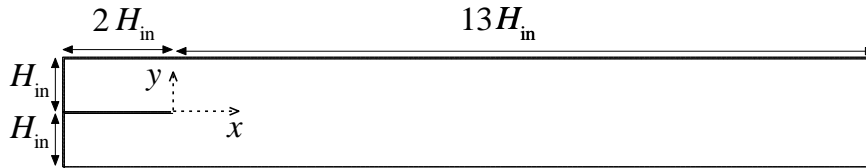
Fig. 1: Schematic of parallel micro/nanomixers.

In the present work, we are going to perform a comparative study between the results of continuum-based governing equations incorporated with the slip/jump boundary conditions and the results of DSMC method for the gas mixing flows through nano/microchannels. For the sake of simplicity, we re-produce the DSMC results from our past published paper [13]. Here, we repeat the simulations using the developed continuum-based solver. Different aspects of predicted flow and mixing behaviors are presented here using the continuum- and molecular-based methods and that their achieved results are suitably compared.

4. Results and Discussion

4.1. Flow velocity and mass flow rate

Figures 2 and 3 respectively present the velocity profiles along the mixer centerline and at the section x/H_{in}=4 considering different mixer sizes. The results indicate that the continuum-based method under-predicts the velocity magnitudes for the mixers working in transitional regime comparing with the values obtained from the molecular-based method. In other words, the rarefaction effects would result in an increase in the velocity magnitudes that cannot be predicted accurately by the continuum-based simulation methods. However, a very good agreement is observed between the predictions of the two different simulation methods for the largest test case which works in the slip flow regime. This assures that the incorporated simplified slip boundary conditions are sufficiently accurate in the slip regime and that they have been implemented correctly.
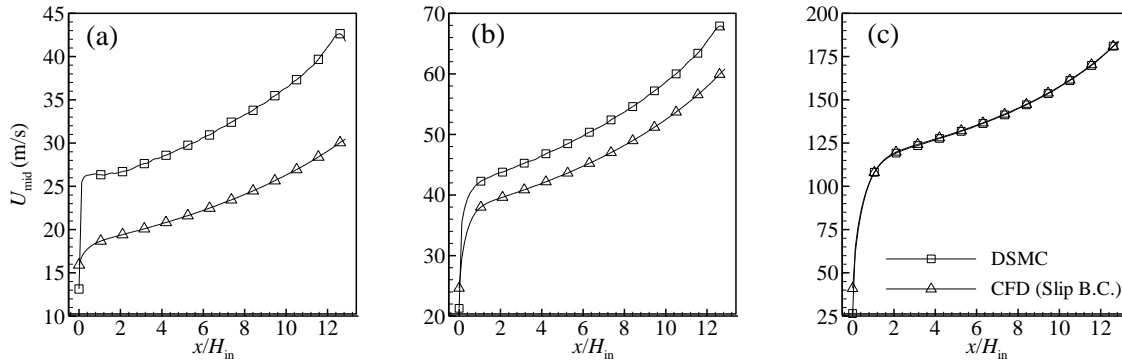

Fig. 2: The velocity variations along the mixing channel centerline; (a) H_{in}=0.04, (b) H_{in} =0.2, and (c) H_{in} =1.0 µm.
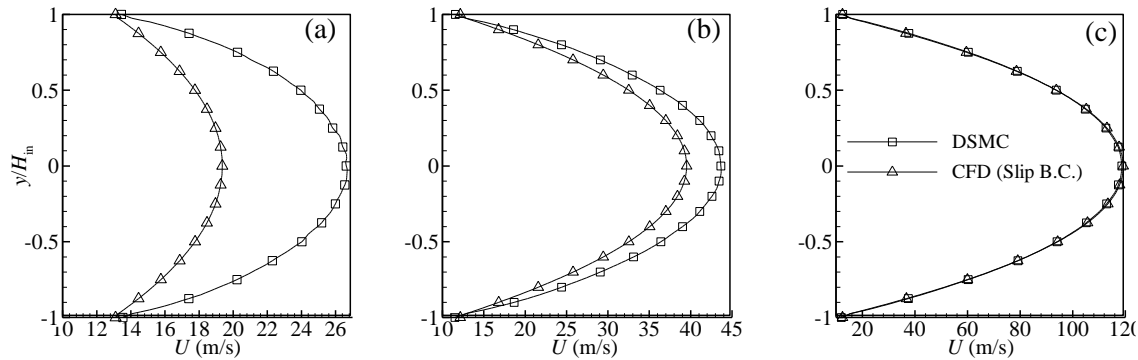

Fig. 3: The velocity variations at the cross section x/H_{in}=4; (a) H_{in}=0.04, (b) H_{in} =0.2, and (c) H_{in} =1.0 µm.

As known, the increase of velocity magnitude at rarefied flow regimes would result in the well-known increase of mass flow rate with respect to the predictions obtained from the continuum-based solvers in these flow regimes. To quantify the inaccuracy of the continuum-based method in predicting the mass flow rate, we calculate the ratio of mass flow rates predicted by the continuum-based method with those predicted by the molecular-based method. Figure 4 presents the dependence of this ratio on the Kn value. It is observed that using the continuum-based method would result in 25, 17, 11, 5, and 0.3% reductions in the calculated mass flow rate with respect to the predictions of the molecular-based method for the test cases with Kn= 1.4, 0.56, 0.28, 0.11, and 0.056, respectively. Again, a good agreement is observed for the test cases in the slip flow regime.
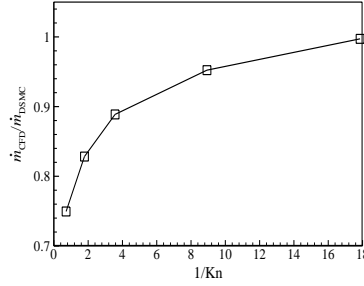
Fig. 4: The ratio of mass flow rate magnitudes predicted by the continuum- and molecular-based simulation methods at different flow rarefaction levels.

4.2. Gas mixing and species distribution

As the two gas streams flow through the mixer, they become gradually mixed and the mixture composition tends to arrive to an equilibrium state. To illustrate this process, we present the variation of N_2 mass fraction along the lower wall of mixers having different inlet heights in Fig. 5. The results indicate that the difference between the results of Continuum- and molecular-based simulation methods are negligible at the slip regime (H_{in}=1.0 µm). However, there are some sensible differences between these results for more rarefied flows. The continuum-based method predicts lower values for Y_{N2} at the inlet (x/H_{in}=-2). This may be attributed to the higher flow velocity magnitudes predicted by this method. It would limit the back diffusion of a species into the inlet channel of the other species.
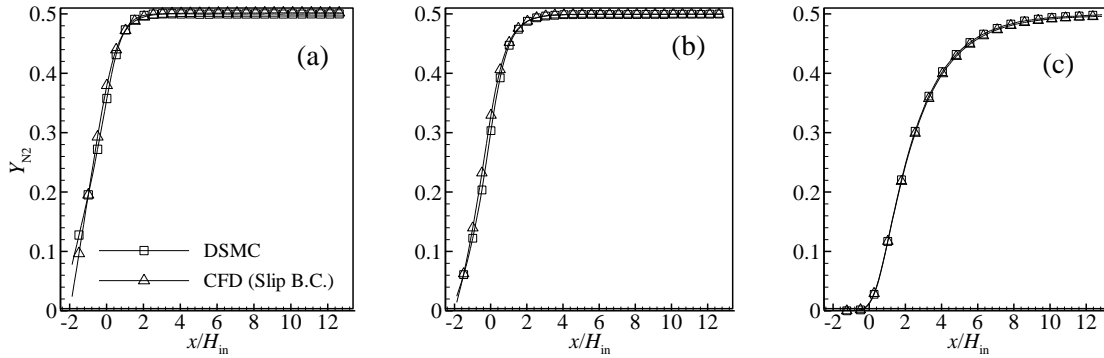

Fig. 5: The variations of N_2 mass fraction along the lower wall of mixer; (a) H_{in}=0.04, (b) H_{in} =0.2, and (c) H_{in} =1.0 µm.

Another comparison between the results of continuum- and molecular-based simulation methods is provided in Fig. 6 for the distribution of species across the mixing channel. This figure presents the variation of N_2 mass fraction across the section x/H_{in}=4. Again, a good agreement between the results of two simulation methods is observed in the slip regime (H_{in}=1.0 µm). All continuum-based simulations and also the molecular-based simulations for the slip flow regime indicate nearly "zero-gradient" variation of species mass fraction on the walls. However, the molecular simulation predicts a non-zero value for the mass fraction gradient on the walls in the rarefied conditions. Moreover, the gradient of Y_{N2} changes sign more effectively moving from one wall to another. This gradient has negative values in regions near the walls. Although not shown here, the y-component of the diffusion mass flux of N_2 gas has non-positive values throughout the mixing channel. This is despite the fact that the ordinary diffusion mass flux of a species tends to be in the opposite direction of its mass fraction gradient as it is predicted by Eq. (9). Having in mind that the two gases have very similar molecular properties (mass, diameter, etc.) and that the temperature and pressure gradients across the channel width are negligible, the baro- and thermal diffusion mechanisms [see Eq. (9)] would not be responsible for these negative mass fraction gradient valuers. This is more justified noting that the continuum-based solver, which takes into account these two mechanisms, does not predict such behaviour. Therefore, it is expected that the classical continuum-based description of the mass diffusion would be completely invalid in the near wall regions of these highly rarefied (transitional) gas flows. Even, using the rarefaction-dependent effective diffusion coefficients presented in Ref. [14] would not be reasonable because it would result in negative values for the effective diffusion coefficient. It seems that using the higher-order constitutive laws is inevitable if one is going to use Eqs. (1)-(4) in simulation of such rarefied gas flows. It is noteworthy to

mention that a similar phenomenon called "cold-to-hot" transfer was observed in studying the heat transfer in rarefied conditions, which could be described by including the higher order terms in the heat flux expression [15].
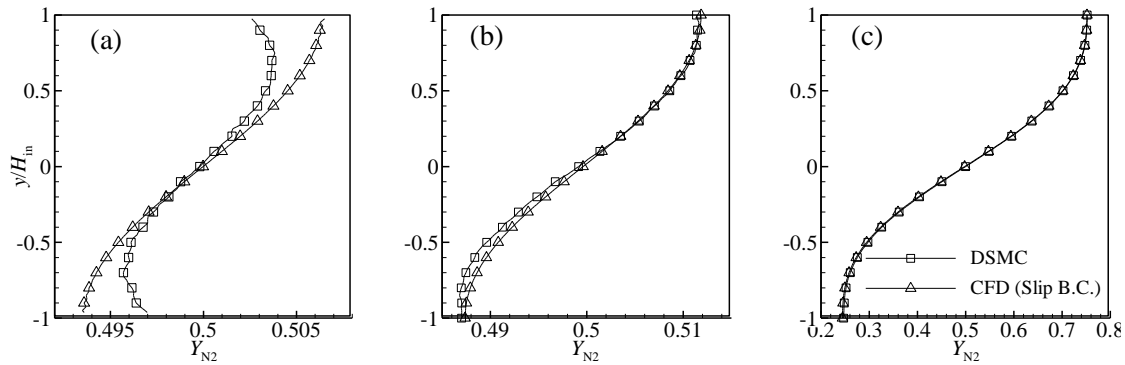

Fig. 6: The variations of N_2 mass fraction at the cross section $x/H_{in}=4$; (a) $H_{in}=0.04$, (b) $H_{in}=0.2$, and (c) $H_{in}=1.0$ µm.

4.3. Diffusion mass fluxes

To compare the results of continuum- and molecular-based simulation methods for the diffusion mechanism, Figure 7 presents the distributions of diffusion mass flux of N_2 gas across the centerline of mixers having different mixer sizes. Although there is fair agreements between the predictions of the two simulation methods over almost the entire simulation domain for the largest mixer size (which is working in the slip flow regime), some major differences are showing up as the mixer size decreases. The differences become more pronounced near the end of splitter plate; i.e. at $x/H_{in}\sim 0$. This region experiences severe mass fraction gradients, which results in severe local rarefaction effects. Detail discussions on these local effects are presented in Ref. [14]. It can be described briefly as follows. The mass fraction gradient reaches its maximum magnitude at a point right after the end of splitter. This maximum magnitude also increases as the mixer becomes smaller. As a result, the continuum-based simulation method, which utilizes constant diffusion coefficients, overpredicts an increase in the magnitude of diffusion flux. However, the severe local rarefaction at the regions experiencing severe gradients has resulted in considerable reduction in the effective diffusion coefficients. This can limit the increase of diffusion mass fluxes at the regions near the end of splitter. This can be deduced from the curves appeared in Fig. 7. Contrary to the continuum-based simulation method, which predicts a considerable increase in the magnitude of diffusion mass fluxes at $x/H_{in}\sim 0$ for the smaller mixers, the molecular-based method predicts a much less increase in this region as the mixer size decreases.
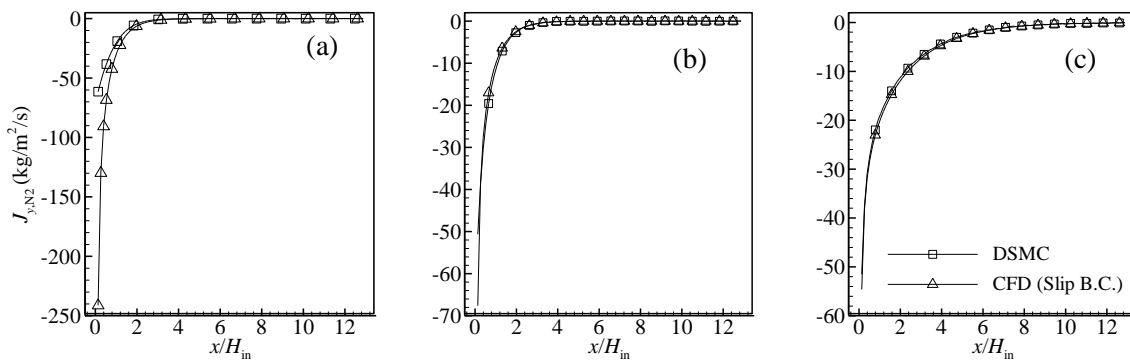

Fig. 7: The variations of y-component of the diffusion mass flux across the centerline along the mixing channel; (a) $H_{in}=0.04$, (b) $H_{in}=0.2$, and (c) $H_{in}=1.0$ µm.

5. Conclusion

The flow and mixing of two gases in micro/nanoscale channels were simulated using the continuum-based incorporated with slip/jump boundary conditions and molecular-based simulation methods. It was observed that for the micromixers working in the slip flow regime, both simulations methods would predict almost the same solution behaviors.

However, in more rarefied conditions occurring in nanomixer cases, the results of continuum-based method could deviate considerably from the results of the molecular-based method, where the latter one is treated as a more accurate one. From the fluid flow perspective, the continuum-based method would under-predict the velocity magnitudes and the mass flow rate through the mixers. From the mixing perspective, a weaker back diffusion is predicted by the continuum-based method. An interesting "dilute-to-dense" mass diffusion phenomenon is observed near the walls of nanomixer cases working in highly rarefied conditions, which cannot be described via using the first-order constitutive laws incorporated with the present continuum-based solver. Finally, the effect of local rarefaction effects on the weakening of diffusion mechanism was observed in this study. It was due to the severe mass fraction gradients

Acknowledgements

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