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The gas leakage dynamic flow in nanoporous silica aerogel under different pressure

Xin P. ZHAO, Zeng Y. LI*, He LIU and Wen Q. TAO

* Corresponding author: Tel.: +86 29 82665446; Fax: +86 29 82665445; Email: lizengy@mail.xjtu.edu.cn
Key Laboratory of Thermo-Fluid and Science and Engineering, Ministry of Education
School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an 710049, P.R. China

Abstract The dynamic gas flow in silica aerogel caused by ambient pressure change is an important factor to influence the thermal conductivity performance. Due to the ultrafine pore size, the flow in the silica aerogel is non-continuum. The direct simulation Monte Carlo method is selected to simulate the transient behavior of gas dynamic flow in nanoporous silica aerogel caused by pressure difference. The influences of pressure ratio and porosity on the unsteady dynamic response and the magnitude of the macroscopic parameters are investigated. The results show that the response processes under different pressure ratio and porosity conditions are similar. The effect of the pressure ratio and porosity are mainly reflected on the magnitude of the macro parameters and response time.

Keywords: DSMC, Gas Leakage, Silica Aerogel, Transient Flow

1. Introduction

Silica aerogel is a kind of nanoporous material with high porosity, low density and high specific surface area. Because the nano structure of silica aerogel is open-cell and possesses ultrafine pore size, it has excellent properties on thermal insulation, filtering, chemo catalysis and other aspects [1, 2]. Generally, the nano structure of silica aerogel is similar as pearl-necklace network named "secondary particles" which are bigger primary particles or aggregated by primary particles (1-5nm). The solid skeleton, gas phase and radiation are three main pathways for heat transfer within silica aerogel. In most cases, the gas phase plays a dominant role in the heat transfer process. The gaseous heat transfer in silica aerogel is determined by the pressure, temperature and the material microstructure [2]. The gaseous thermal conductivity in silica aerogel is much lower than that in free space because the motions of gas molecules are constrained seriously by the complex structure of solid framework.

It is reasonable for the gas in silica aerogel to be treated as motionless at a stable environment. However, the gas leakage dynamic flow will happen if there exists pressure difference from the aerogel materials to the working environment [3]. The macroscopic movement of gas may change the thermal insulation performance. Thus, it is necessary and important to study the gas dynamic response characteristics when pressure difference exists. Yet, it is difficult to investigate the dynamic process through experiment.

The aim of this paper is to simulate the influences on the gas flow in silica aerogel when a sudden change of the ambient pressure happens. In the present study, three-dimensional (3D) diffusion-limited cluster-cluster aggregation (DLCA) method is adopted to generate the complex inner structure of silica aerogel [4]. The gas flow and mass transfer in silica aerogel under different pressure difference are predicted based on 3D direct simulation Monte Carlo (DSMC) method [5].

2. Physical Model and Numerical Methods

Within silica aerogels, mesopores (2-50nm) are dominated. Micropores (<2nm) are mainly concentrated in "secondary particles". There also exist a few portion of macropores (>50nm) [1]. Usually, the gas molecules in "secondary particles" are restricted remarkably. Thus, we assume that micropores in the 'secondary particles' have little influence on the gas flow in silica aerogels are ignored. For simplicity, we assume that 'secondary' particles are uniform and with no micro pores in DLCA method.

The flow mechanism of gas in silica aerogel is controlled by viscous ($Kn \ll 1$) or molecular ($Kn \gg 1$) processes depending on the local size and shape of pores [3]. The Knudsen number is defined as

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

where λ is the mean free path of gas in silica aerogel influenced by the number density of gas molecules and the structure of pore, and D is the equivalent pore size.

In most cases, the pore size distributions of silica aerogels are mainly concentrated on 1-100nm [1,2]; the mean free path will be much smaller than that in free space (70nm, air at 300K and 1 atm) considering the influence of solid skeleton. Therefore, the noncontinuum effect needed to be taken into account ($Kn > 0.01$).

2.1 Physical model

Let's consider a porous flat with infinite thicknesses in x and y directions and a certain thickness in z direction which separates the recipient and the environment [6, 7]. The recipient and the environment are separated by a distance L_z in z direction at the pressures $p(z_0) = p(z_L) = p_0$ and the temperature T_0 initially. At the time $t = 0$, the dynamic flow occurs in z direction as the pressure of atmosphere varied Δp suddenly. The temperature of the solid frame T_0 will keep constant. The computational domain is shown in Fig.1. The boundaries in x and y directions are periodic.

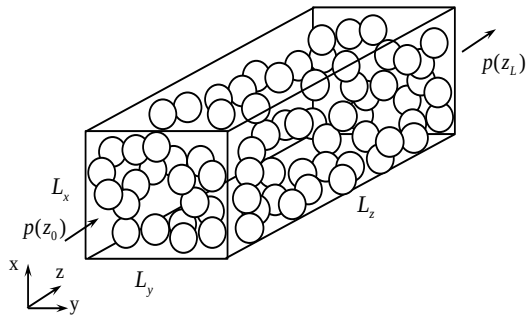

Fig.1 The model of the domain

The parameters are normalized to render the problem dimensionless.

$$z = \frac{z'}{L_z}; \qquad n = \frac{n'}{n_0}; \qquad p = \frac{p'}{p_0};$$

$$t = \frac{t'}{\lambda_0 / \upsilon_0}; \qquad M = \frac{M'}{M_0}; \qquad w = \frac{w'}{\upsilon_0}$$

where the mean free path $\lambda_0 = kT_0 / \sqrt{2}\pi d^2 p_0$, the most probable molecular speed $\upsilon_0 = \sqrt{2kT_0 / m}$, k is the Boltzmann constant, d is the molecular diameter, m is the molecular mass and M_0 is the mass flow rate when silica aerogel is removed at the same pressure boundaries.

2.2 DLCA method

Silica aerogel is a type of classical porous material prepared by small nanoscale silica particles aggregation and then dried and the process is called sol-gel. There are a variety of aggregation models such as BA (ballistic aggregation), RLA (reaction-limited aggregation), DLA (diffusion-limited cluster aggregation), and DLCA (diffusion-limited cluster-cluster aggregation) which have been developed to simulate this process [4, 8].

Here, we adopt the off-lattice DLCA method to simulate the solid frame of silica aerogel [4]. Initially, a certain number of particles which determined by the porosity of a given structure are placed random in the simulation region. Then, a particle or a cluster is picked random and moved a small distance in a random direction. The moved particle or cluster will be detected whether it collides with other particles or clusters. The colliding particles or clusters will aggregate a bigger cluster and move together. If not, another

particle or cluster will be chosen and moved. The process will be repeated until all the particles merge into one cluster. The size of the simulated particles can be uniform or not. In current work, uniform 'secondary particles' which represent bigger particles or clusters formed by small particles aggregation are used. The structure generated is shown in Fig.2.

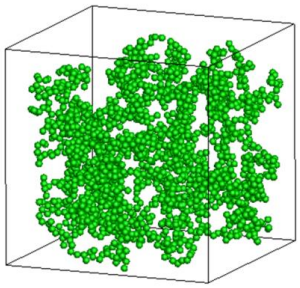

Fig.2 The structure of silica aerogel generated by DLCA

The shape of the particles in our simulations is treated as cubic, and the size is 6.5nm in our simulations. However, it should be emphasized that the neglect of micropores will lead the calculated specific surface area smaller compared with the real structure due to the fact that the micropores are not considered. The mean pore size can be given by

$$D = \frac{4}{S}\left(\frac{1}{\rho_{skeleton}} - \frac{1}{\rho_{bulk}}\right) \tag{2}$$

where S is the specific area, $\rho_{skeleton}$ is the density of porous medium and ρ_{bulk} =2200 Kg/m^3 is the density of silica. Also, the calculated values of the mean pore size will be bigger than the real structure.

2.2 DSMC method

Because the motions of molecules are restricted by solid skeleton and the scale of the pores close to the mean free path, the flow regime in silica aerogel is non-continuum. The flow status can be described by the Boltzmann equation. Considering the complicated inner structure of the computational domain, it is difficult to solve the Boltzmann equation directly. Whereas, it is convenient to reflect the influence of the skeleton structure for DSMC method which is based on the direct simulation of the particles' movements. In DSMC method, the number density, temperature and the mean flow velocity are needed for the opening boundary [8]. However, the pressure cannot be used directly. In the calculation, only the inlet pressure and temperature and the outlet pressure are given. According reference [9], the inlet velocity can be acquired:

$$w_{j,in} = w_j + \frac{p_{in} - p_j}{\rho_{j,in} a_{j,in}} \tag{3}$$

For outlet,

$$\rho_{j,e} = \rho_j + \frac{p_{j,e} - p_j}{(a_j)^2} \tag{4}$$

$$w_{j,e} = w_j + \frac{p_j - p_e}{\rho_{j,e} a_{j,e}} \tag{5}$$

$$T_{j,e} = \frac{p_e}{\rho_{j,e} R} \tag{6}$$

where j is the grid number, in represents the inlet, e denotes the outlet and R is the gas constant.

A modified version which is suitable for calculating the flow in porous medium is developed based on the traditional Bird's code. Because the scale of the grids used to discern the solid skeleton is too small to satisfy the statistical principle of DSMC, we generate two sets of grids in our modified technique. One is used to trace the movements of the molecules, and the other is utilized to statistically calculate the macroscopic parameters. The time step in the simulations are 0.01 τ or 0.1 τ according to the time needed for the system to reach the steady state, where $\tau = \lambda_0 / \upsilon_0$. The results are ensemble averaged until the needed macro parameters are acquired [10-12]. The mass flow rate through the flow cross-section is calculated by $M = \sum_{j=1}^{N} m n_j w_j / N$, where N is the total number of cells in the cross sections.

3. Results and Discussion

The dynamic flow in the nanoporous silica aerogel has been studied at different pressure

ratio of outlet pressure p_e to initial pressure p_0 defined as $PR = p_e / p_0$ (PR=0.1, 0.25, 0.5 and 0.75) and porosity (φ =0.827, 0.870, 0.904, 0.938 and 0.956). And the mean pore size according to equation (2) is 29.4nm, 40.6nm, 56.5nm, 89.4nm, 125.7nm as the porosity increasing. The initial pressure p_0 keeps 101325 Pa in all cases. The temperature of the solid frame is also kept T_0 during the simulations. Because of the needed time for the systems to reach steady, the combined time step 0.01 τ or 0.1 τ are adopted in the simulations to reduce the amount of calculation. For convenience, all the results are presented by dimensionless macro parameters. In the following parts, we will analyze the typical transient solutions of macroscopic properties from the initial state to steady state first. Then, the influences of pressure ratio and the porosity of the structure will be studied. The results are calculated at the same simulation region.

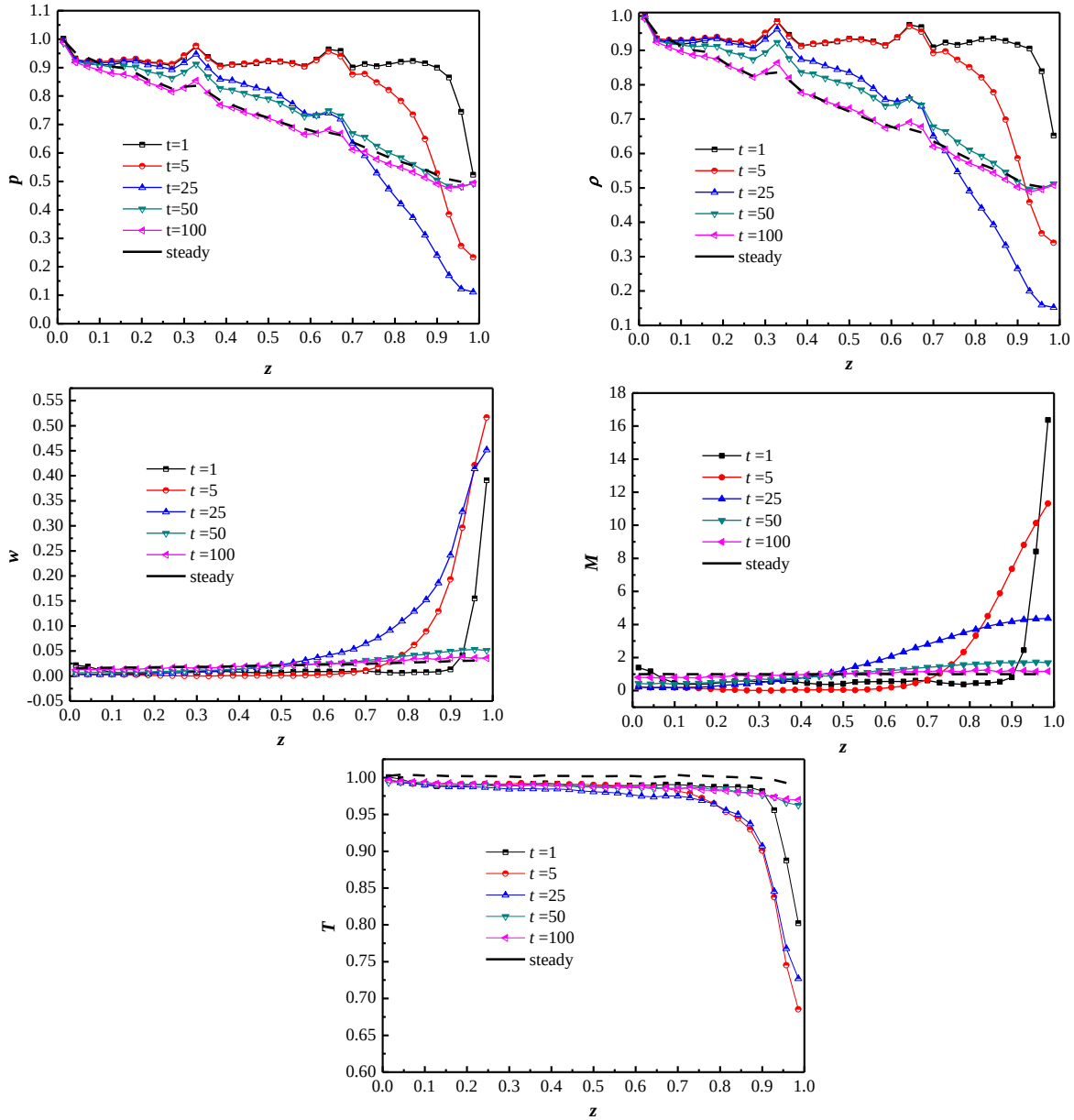

Fig.3 Distributions of dimensionless pressure p, density ρ, velocity w, temperature T, mass flow rate M at PR=0.5, φ =0.938

Fig.3 shows the transient processes of cross-section averaged macro-parameters pressure p, density ρ, velocity w, temperature T, and mass flow rate M distributions along the coordinate z after the environment pressure decreases at $t = 0$ (PR=0.5, φ =0.938). The steady parameters are presented by the solid line. From the represented density profiles, we can see that the immediate pressure-drop will drive a number of gas molecules to escape from the nano-porous medium to the environment. This tendency will propagate to the upstream rapidly as time goes on. The directional gas molecules movement will cause the macro mass flow rate inside the silica aerogel and the fluctuations of other parameters like pressure, temperature and velocity in the whole computational region.

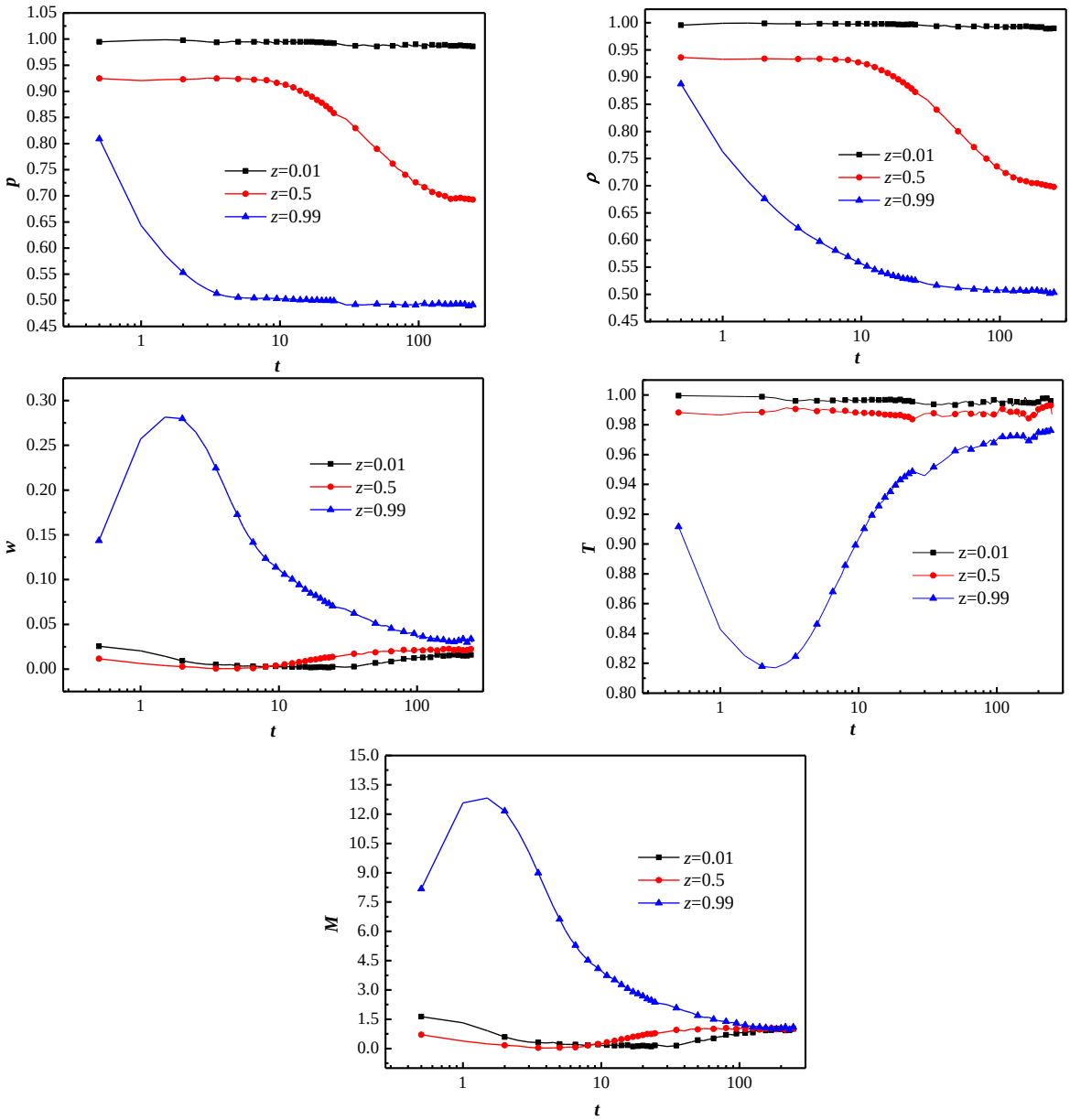

Fig.4 Time evaluation of dimensionless pressure p, velocity w, temperature T, mass flow rate M at PR=0.5, φ =0.938

The unsteady process about the macro cross-section averaged parameters are predicted at three locations in z direction (z=0.01, z=0.5, z=0.99) as shown in Fig.4 (PR=0.5, φ=0.938). Combining the variation trend of macro parameters in Fig.3 and Fig.4, we can divide the dynamic response into non-regular regime and regular regime like the classification of unsteady heat transfer: in non-regular regime, the distributions of the macro parameters are controlled by the initial conditions and the influences of the initial condition disappears gradually as time increasing; in regular regime, the distributions of the macro parameters are controlled by boundary conditions. For the case in Fig.3 and Fig.4, the non-regular regime is from $t\approx0$ to $t\approx10$. In this stage, the immediate pressure drop mainly influence the region near the outlet and the magnitude of the macro parameters varies quickly at the outlet (z=0.99). In the regular regime, the fluctuation continues to spread; the influences of propagations begin to be reflected in the whole domain ($t\approx10$~100). And the system reaches a new stationary status at the end of the regular regime.

Fig.5 compares the dynamic responses of macro parameters at different pressure ratio varying from 0.1 to 0.75 with the same structure (φ=0.938) at the outlet (z=0.99). The transient behavior of the dynamic process is similar. However, the non-regular regime increases as the PR decreasing and the magnitude of the macro parameters has higher fluctuations at the outlet in our simulations. Table.1 shows the rough time t of the system needed to reach steady flow states at different PR. From the specific values, we can see that the total time of the establishment for the system also increases as the PR decreasing.

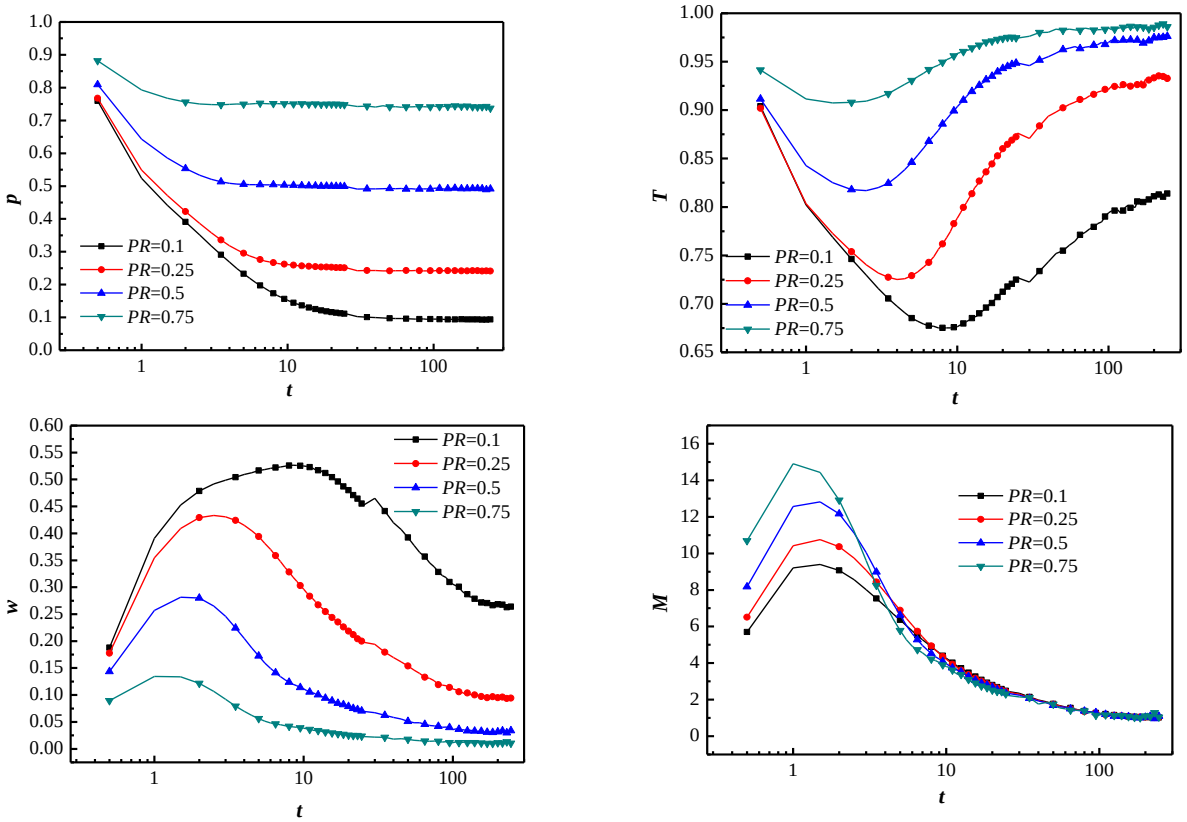

Fig.5 Time evaluation of dimensionless pressure p, velocity w, temperature T, mass flow rate M at different PR, z=0.99

Table1. The approximate time of the system needed to be steady (p_0 =101325, φ =0.938)

PR	*t*
0.75	160
0.5	170
0.25	210
0.1	220

In Fig.6, the influence of porosity on the macro parameters at the outlet is predicted at same pressure ratio (*PR*=0.5). With the decrease of porosity, the mean size of the pores and channels becomes smaller and the flow resistance of the gas flow also becomes stronger due to more frequently collisions between gas molecules and solid skeleton. Thus, the values of the macro parameters in the domain with lower porosity are smaller. However, due to the weaker fluctuation, the non-regular regime decreases when the structure porosity is lower. In general, the overall impact of the porosity on the transient behavior is like the pressure ratio as can be observed. Tabel.2 shows the approximate time of the system needed to reach the steady states. We can see that the overall trend of the needed time for the system to reach steady state is increasing as the porosity decreases. And there exists fluctuation, the specific information need further research.

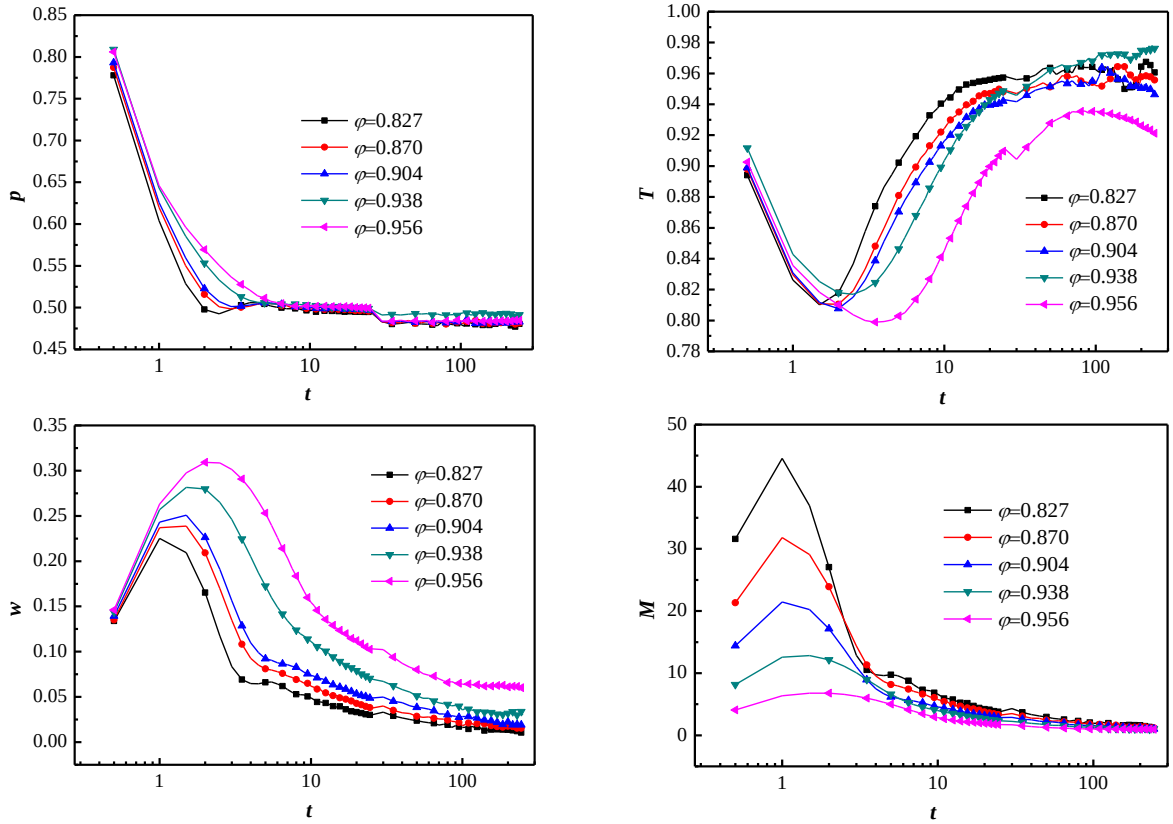

Fig.6 Time evaluation of dimensionless pressure p , velocity w , temperature T , mass flow rate M at different porosity φ , z=0.99

Table2. The approximate time of the system needed to be steady (p_0 =101325, *PR* =0.5)

φ	*t*
0.827	420
0.870	330
0.908	340
0.938	170
0.95	105

3. Conclusions

A modified direct simulation Monte Carlo method is used to calculate the gas leakage dynamic flow in nanoporous silica aerogel. The random nanoporous structures of silica aerogel with different porosity are generated by diffusion limited cluster aggregation

method. The flow is simulated at different pressure difference and porosity. Our simulations show that the response processes of transient behavior at different pressure ratio and porosity are similar. And the influences of the pressure and structure porosity are mainly reflected on the magnitude of the macro parameters and the response time. For pressure ratio, a lower value would lead a higher magnitude and longer response time. For porosity, the overall trend is that the lower porosity cause weaker macro movement. However, the response time is longer. The obtained results will be used to analysis the influence on the thermal conductivity caused by macro mass flow.

Acknowledgements

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