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# Treatment of natural gas with varying $CO_2$ concentration using supersonic flows

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*Abstract*—In order to reveal the supersonic condensation characteristics of carbon dioxide ($CO_2$) in natural gas under low temperature condition, the mathematical models and numerical schemes for the $CH_4$-$CO_2$ mixture gas under supersonic flow condition were established, and the influence of inlet $CO_2$ concentration on the $CO_2$ removal efficiency of Laval nozzle was investigated. The results show that with the increase of $CO_2$ concentration at nozzle inlet, the initial nucleation position is closer to nozzle throat, the maximum nucleation rate and the droplet number decrease but the droplet radius increases significantly, which eventually leads to an increase in liquefaction rate. When the inlet $CO_2$ mole fraction is lower (less than 10%), the $CO_2$ fraction in gas phase is almost 0, which indicates that low-temperature separation technology is practicable for the removal of $CO_2$ from natural gas.

*Keywords—$CO_2$, natural gas, supersonic, condensation*

## I. INTRODUCTION

With the optimization of energy structure and increasing emphasis on environmental issues, the demand for natural gas continues to grow with each passing day. However, natural gas extracted from wellheads often contains a certain amount of carbon dioxide ($CO_2$). The $CO_2$ is a kind of harmful impurity, which will not only cause corrosion to transport pipelines and equipment, but also increase the greenhouse gas emissions [1,2]. Currently, the common decarbonization processes include absorption [3], adsorption [4] and membrane separation [5], which have been used in different situations, respectively. However, these processing methods for natural gas suffer from shortcomings such as high energy consumption, serious secondary pollution and cumbersome equipment.

The supersonic separation technology is an emerging mixture gas separation technology being developed in recent years. The main principle of this technology is that high-pressure gas flows through Laval nozzle, and the refrigeration effect generated by high-speed expansion causes condensable components to condense from mixture gas, thereby achieving liquefied separation of condensable components. Supersonic separation technology has many advantages such as low capital costs, environmental friendliness, and flexible structure. The application of supersonic separation technology in the field of natural gas dehydration [6] and natural gas liquefaction [7-9] has been widely studied theoretically and experimentally. It has great potentials for the remove $CO_2$ from natural gas, which is of great significance to improve and develop the purification technology for natural gas containing $CO_2$ and reduce the treatment cost of natural gas.

In this paper, Laval nozzle structure was designed according to the flow and condensation properties of natural gas containing varying quantities of $CO_2$. Based on gas - liquid two - phase flow governing equations, internal consistent classical nucleation theory (ICCT) model and Gyarmathy's droplet growth model, the mathematical models and numerical schemes of supersonic condensation flow of methane ($CH_4$)-$CO_2$ mixture gas were established ($CH_4$ is the main component of natural gas). The condensation flow characteristics of $CH_4$-$CO_2$ mixture gas under supersonic condition were studied by adding the effect of condensation on governing equations to FLUENT via User-Defined Functions (UDF) and User-Defined Scalar (UDS). The effects of inlet $CO_2$ concentrations on the condensation process and the removal efficiency of $CO_2$ gas were mainly studied, The results will lay a foundation for the application of supersonic separation technology in the field of $CO_2$ removal from natural gas.

## II. DESIGN OF LAVAL NOZZLE

The Laval nozzle adopted in this work is mainly composed of four parts, as shown in Fig. 1. The throat diameter was determined by the flow rate and thermodynamic parameters of the mixture gas. In order to improve the stability of the flow field and reduce the influence of turbulence on the flow field, the convergent section of the nozzle was designed with double cubic curve.

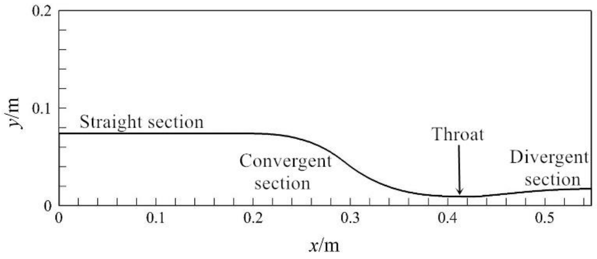

Fig. 1. The structure diagram of the Laval nozzle.

## III. MATHEMATICAL MODEL AND NUMERICAL SCHEMES

### A. Mathematical model

In the $CH_4$ - $CO_2$ mixture gas, $CH_4$ is carrier gas and $CO_2$ is condensable gas. The governing equations were established considering the slip between the different phases, which include continuity, momentum and energy equations, presented as Eqs. (1)-(3). The liquid governing equations were expressed in Eqs. (4)-(6) [10,11].

$$\frac{\partial \rho_{\mathrm{v}}}{\partial t}+\frac{\partial}{\partial x_j}\left(\rho_{\mathrm{v}} u_j\right)=S_{\mathrm{m}} \tag{1}$$

$$\frac{\partial}{\partial t}\left(\rho_{\mathrm{v}} u_i\right)+\frac{\partial}{\partial x_j}\left(\rho_{\mathrm{v}} u_j u_i\right)= \tag{2}$$

$$-\frac{\partial p_{\mathrm{v}}}{\partial x_i}+\frac{\partial}{\partial x_j}\left[\mu\left(\frac{\partial u_j}{\partial x_i}+\frac{\partial u_i}{\partial x_j}-\frac{2}{3}\delta_{\mathrm{ij}}\frac{\partial u_j}{\partial x_j}\right)\right]+\frac{\partial}{\partial x_j}\left(-\rho_{\mathrm{v}}\overline{u_i' u_j'}\right)+S_{\mathrm{u}}$$

$$\frac{\partial}{\partial t}\left(\rho_{\mathrm{v}} E\right)+\frac{\partial}{\partial x_j}\left(\rho_{\mathrm{v}} u_j E+u_j p_{\mathrm{v}}\right)=\frac{\partial}{\partial x_j}\left(k_{\mathrm{eff}}\frac{\partial T}{\partial x_j}+u_i \tau_{\mathrm{eff}}\right)+S_{\mathrm{h}} \tag{3}$$

$$\frac{\partial}{\partial t}\left(\rho_{\mathrm{v}} E\right)+\frac{\partial}{\partial x_j}\left(\rho_{\mathrm{v}} u_j Y\right)=S_{\mathrm{Y}} \tag{4}$$

$$\frac{\partial}{\partial t}\left(\rho_{\mathrm{v}} N\right)+\frac{\partial}{\partial x_j}\left(\rho_{\mathrm{v}} u_j N\right)=J \tag{5}$$

$$r_{\mathrm{d}}=\left(\frac{3Y}{4\pi\rho_{\mathrm{l}} N}\right)^{\frac{1}{3}} \tag{6}$$

where $S_m$, $S_u$, $S_h$ are the source terms of continuity, momentum and energy equations, respectively; $u$ is the gas velocity; $p$ is the pressure; $\mu$ is the gas viscosity; $\delta_{ij}$ is the Kronecker delta; $u'$ is the velocity fluctuation; $E$ is the tatal energy; $k_{eff}$ is the effective thermal conductivity; $T$ is the temperature of mixture; $\tau_{eff}$ is the effective stress tensor; $N$ is the droplet number; $J$ is the nucleation rate.

Spontaneous condensation of gas has typical characteristics of gas - liquid imbalance. The ICCT model derived by Girshick [12,13] was adopted to calculate the nucleation, and the Gyarmathy's model [14] was used to calculate the droplet growth in this paper.

### B. Numerical schemes

The computational fluid dynamics (CFD) software package ANSYS Fluent 16.0 was employed as the numerical simulation tool. The $k$-$\omega$ turbulence model was applied since the gas flow in Laval nozzle is compressible flow under high-speed condition [15,16]. The equations were discretized by the second-order upwind scheme. In order to consider the effect of phase transition on the governing equation, the governing equations of liquid phase was added by UDS, and the source terms were added to FLUENT using UDF written by C language.

Based on the flow characteristics of flue $CH_4$-$CO_2$ mixture gas in Laval nozzle, both nozzle inlet and outlet are set as pressure boundaries, the $CO_2$ concentration at the inlet is changed according to the calculation requirements.

## IV. RESULTS AND DISCUSSION

The numerical methods established above were used to simulate the condensation process of $CH_4$-$CO_2$ mixture gas. The calculation conditions are as follows: the inlet temperature of the mixture gas is 273.15 K, the inlet pressure of the mixture gas is 6 MPa, and the inlet $CO_2$ mole fractions are varied between 10% and 20%. The condensation parameters of $CO_2$ in the Laval nozzle are shown in Figs. 2-6.

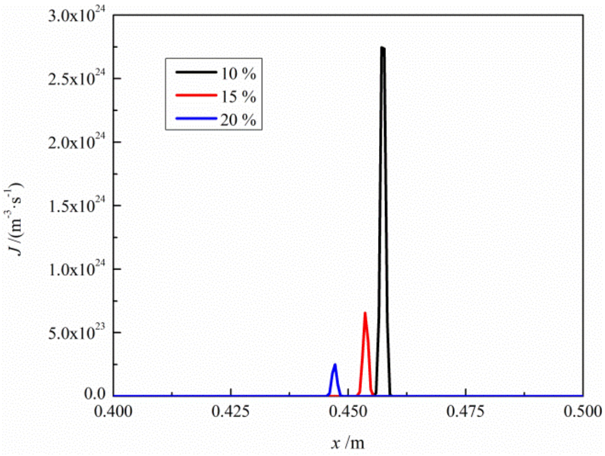

Fig. 2. The nucleation rate distribution of $CO_2$ in the Laval nozzle.

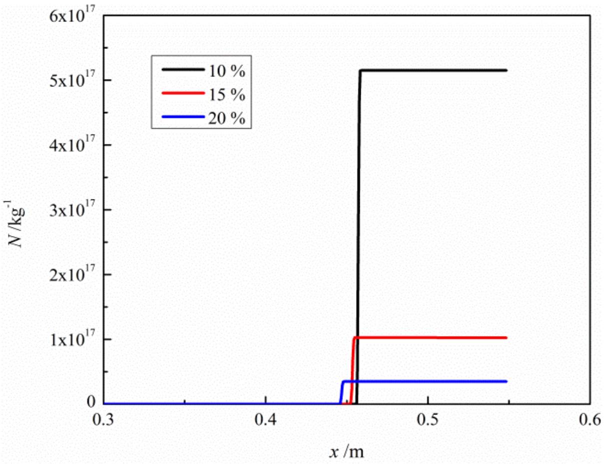

Fig. 3. The droplet number distribution of $CO_2$ in the Laval nozzle.

Fig. 2 and Fig. 3 show the nucleation rate and droplet number distributions with different gas composition. As we can see, with the increase of the inlet $CO_2$ concentration, the nucleation position (Wilson point) is advanced, the maximum nucleation rate and droplet number decrease obviously. When the inlet $CO_2$ mole fraction rises from 10% to 20%, the maximum nucleation rate decreases from $2.65\times10^{24}$ $kg^{-1}s^{-1}$ to $2.47\times10^{23}$ $kg^{-1}s^{-1}$, and the droplet number decreases from $5.12\times10^{17}$ $kg^{-1}$ to $3.42\times10^{16}$ $kg^{-1}$.

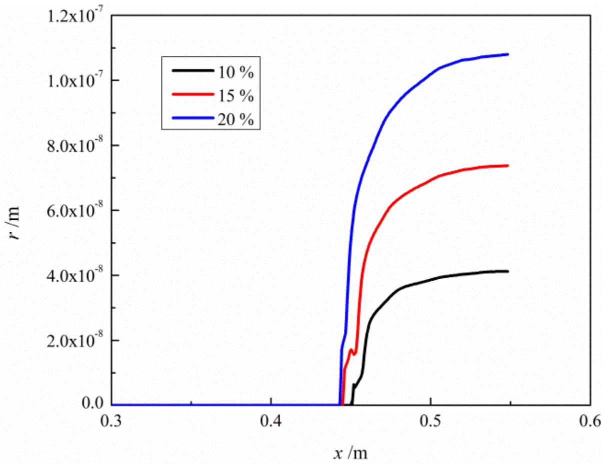

Fig. 4. The droplet radius distribution of $CO_2$ in the Laval nozzle.

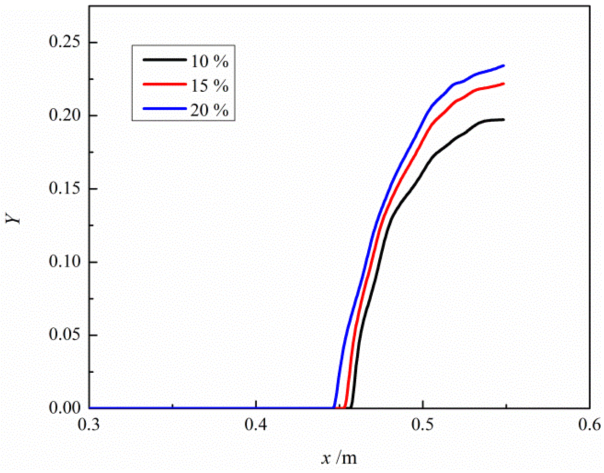

Fig. 5. The liquefaction rate distribution of $CO_2$ in the Laval nozzle.

The distributions of the droplet radius and liquefaction rate are presented in Fig. 4 and Fig. 5. As seen, when the condensation occurs, droplet radius and liquefaction rate increase rapidly in the early stage. This is because after the nucleation rate and the droplet number reach the peak, a large number of vapor molecules will agglomerate on the surface of the droplets, which makes the droplet radius and liquefaction rate increase rapidly. After this stage, the droplet radius and liquefaction increase slowly, reaching the maximum value at the outlet of Laval nozzle. In addition, the droplet radius and liquefaction rate increase with the increase of inlet concentration of $CO_2$. When the mole fraction of $CO_2$ at nozzle inlet rises from 10% to 20%, the maximum droplet radius and the maximum liquefaction rate increase from $3.95\times10^{-8}$ m and 18.86% to $1.07\times10^{-7}$ m and 23.05%, respectively.

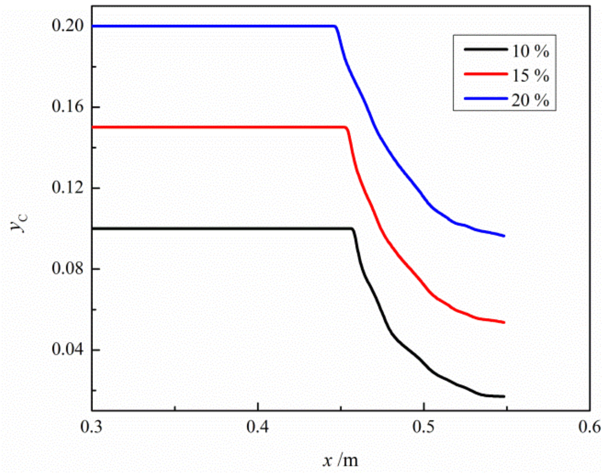

Fig. 6. The $CO_2$ fraction in gas phase distribution in the Laval nozzle e.

Fig. 6 shows the concentration distribution of $CO_2$ in gas phase, it can be seen that the concentration of $CO_2$ in the gas phase at the nozzle outlet is higher with the higher inlet $CO_2$ concentration, even though the radius of droplets is larger and liquefaction rate is higher, more $CO_2$ remains in the gas phase at the nozzle outlet. When the inlet $CO_2$ mole fraction is lower (less than 10%), the $CO_2$ fraction in gas phase is almost 0, which indicates that low-temperature separation technology is practicable for the treatment of natural gas with low $CO_2$ concentration.

## V. CONCLUSION

In this paper, an innovative technology to separate $CO_2$ from natural gas by using Laval nozzle was proposed. The mathematical models and numerical schemes for natural gas containing $CO_2$ were established, and the influence of inlet $CO_2$ concentration on the removal efficiency of $CO_2$ in the Laval nozzle was investigated. The conclusions are as follows:

(1) With the increase of inlet of $CO_2$ concentration, the nucleation position moves forward to the throat. When the inlet $CO_2$ concentration increases from 10% to 20%, the maximum nucleation rate and the droplet number decrease but the droplet radius increases significantly, which eventually leads to an increase in liquefaction rate.

(2) When the concentration of $CO_2$ at the Laval nozzle inlet is low (especially less than 10%), the $CO_2$ fraction in gas phase is almost 0, which indicates that low-temperature separation technology is practicable for the treatment of natural gas with low $CO_2$ concentration.

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