Numerical simulation of carbon dioxide removal from natural gas using supersonic nozzles

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Abstract. Supersonic separation is a technology potentially applicable to natural gas decarbonation process. Preliminary research on the performance of supersonic nozzle in the removal of carbon dioxide from natural gas is presented in this study. Computational Fluid Dynamics (CFD) technique is used to simulate the flow behavior inside the supersonic nozzle. The CFD model is validated successfully by comparing its results to the data borrowed from the literature. The results indicate that the liquefaction of carbon dioxide can be achieved in the properly designed nozzle. Shock wave occurs in the divergent section of the nozzle with the increase of the back pressure, destroying the liquefaction process. In the supersonic separator, the shock wave should be kept outside of the nozzle.

1. Introduction

Natural gas plays a more and more important role in the worldwide energy supply network. When burned as fuel for energy, natural gas emits less harmful pollutants to the environment than other fossil fuels. Natural gas is a gaseous mixture of hydrocarbons and various contaminants when produced from the reservoir. The impurities such as water vapor, carbon dioxide, hydrogen sulphide, and heavier hydrocarbons must be removed from the natural gas before it is brought to the market. Conventional technologies such as absorption and adsorption may have good separation performance, but they need relatively large equipment, complicated system and consequently high investment [1]. Meanwhile, they may have a negative influence on the environment due to the use of chemicals [2]. Supersonic separator has emerged as a promising technology to eliminate some of the disadvantages mentioned above [3]. It is a compact device capable of separating condensable components from gaseous mixture by combining the principles of adiabatic cooling and cyclonic separation [4]. The Laval nozzle is a key part of the supersonic separator. Supersonic flow is obtained in properly designed converging-diverging nozzle, resulting in low pressure and temperature. Researches on the flow behavior in the supersonic nozzles have been reported in voluminous literature [5-9]. However, the investigation on the performance of supersonic nozzles in separating carbon dioxide from natural gas is deficient at present. The flow behavior of methane-carbon dioxide mixture inside the supersonic nozzle is investigated and the influence of back pressure on the performance of the supersonic nozzle is discussed in this work.

2. Nozzle structure
The Laval nozzle contains three sections: convergent section, throat and divergent section. The converging contour is calculated by a cubic polynomial equation, which can be expressed by equation (1). The structure parameters of the nozzle employed in this work are shown in Table 1.

\[
\frac{D - D_t}{D_t - D} = \begin{cases} 
1 - \frac{1}{X_m^3} \left( \frac{x}{L} \right)^3 & \frac{x}{L} \leq X_m \\
\frac{1}{(1 - X_m)^3} \left( \frac{x}{L} \right)^3 & \frac{x}{L} > X_m
\end{cases}
\]

where \( x \) is the distance from the inlet, \( D, D_t \) and \( D_s \) are the diameters at \( x \), inlet and throat, respectively. \( L \) is the length of the convergent section. \( X_m = 0.52 \) is the relative coordinate of the junction point between the front section and the back section.

**Table 1.** Geometry size of supersonic nozzle.

| Parameter             | Value (mm) |
|-----------------------|------------|
| Inlet diameter        | 50.00      |
| Throat diameter       | 11.00      |
| Outlet diameter       | 12.00      |
| Converging length     | 33.80      |
| Diverging length      | 17.86      |

3. Mathematical model

3.1. Governing equations

In a supersonic nozzle, the gas flow characteristics can be depicted by partial differential equations, including mass, momentum, and energy equations, as described in equations (2), (3) and (4).

\[
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_j} (\rho u_j) = 0 
\]

\[
\frac{\partial}{\partial t} (\rho u_j) + \frac{\partial}{\partial x_j} (\rho u_j u_j) = -\frac{\partial p}{\partial x_j} + \frac{\partial}{\partial x_j} \left[ \mu \left( \frac{\partial u_j}{\partial x_j} + \frac{\partial u_j}{\partial x_j} - \frac{2}{3} \frac{\partial u_j}{\partial x_j} \right) \right] + \frac{\partial}{\partial x_j} \left( -\rho u_j u_j \right) 
\]

\[
\frac{\partial}{\partial t} (\rho E) + \frac{\partial}{\partial x_j} (\rho u_j E + u_j p) = \frac{\partial}{\partial x_j} \left( k_{eff} \frac{\partial T}{\partial x_j} + u_j \tau_{eff} \right) 
\]

where \( \rho \) is the density, \( t \) is the time, \( u \) is the velocity, \( p \) is the pressure, \( \mu \) is the viscosity, \( \delta_{ij} \) is the Kronecker delta, \( E \) is the total energy, \( k_{eff} \) is the effective thermal conductivity, \( \tau_{eff} \) is the effective viscous stress.

3.2. Numerical method

The flow behavior in the Laval nozzle is simulated by computational fluid dynamics (CFD) modeling using Fluent software. The density based solver and \( k-\omega \) turbulence model are employed in this study as they are applicable to compressible flow.

According to the flow characteristic of the supersonic compressible flow, pressure boundary conditions are imposed for the inlet and outlet. The total pressure, static pressure, total temperature, turbulence parameters, and species are fixed for the inlet and outlet. No-slip and adiabatic boundary conditions are posted to the wall.

The unstructured meshes are generated and a fine grid scheme is employed in the boundary layer. Grid independence is investigated for the designed nozzle before the simulation is carried out. The second
order upwind scheme is employed for the flow equations, and the first order upwind scheme is applied to the turbulent kinetic energy and specific dissipation rate. The solution is considered as converged when the residuals drop below $10^{-6}$ for the energy and $10^{-3}$ for the others, while the mass flow rate relative error between the inlet and outlet is less than 0.05%.

4. Results and discussion

4.1. Model validation

The model is validated by the work of Arina [10]. The working fluid is air. The inlet temperature and pressure are 288 K and 0.1 MPa, respectively. The outlet pressure is 83% of the inlet pressure. The same nozzle geometry, inlet and outlet conditions are used in the validation study. The pressure and density distribution along the nozzle axis are presented in figure 1. As can be seen from figure 1, the simulation results are in excellent agreement with the data in Arina’s work and the discontinuous changes of fluid properties caused by shock wave are captured accurately by the simulation, which illustrates that the model discussed above is suitable to simulate the flow behavior inside the Laval nozzle.

![Figure 1](image.png)

Figure 1. Comparison of pressure and density distribution along the nozzle axis.

4.2. Nozzle flow behavior

The flow field in the supersonic nozzle is simulated based on the above numerical method. A binary mixture of methane (70 mole %) and carbon dioxide (30 mole %) is chosen as the working fluid in this study. Total pressure and total temperature at the inlet are set to be 8 MPa and 303 K, respectively. The pressure at the outlet is not specified as the flow will be supersonic at the exit. The pressure-temperature variation is shown in figure 2, where the phase envelope of the gas mixture is also plotted. As figure 2 shows, the pressure-temperature curve enters the two-phase region as the stream flows through the supersonic nozzle, which illustrates that the liquefaction of carbon dioxide can be achieved.
4.3. Effect of back pressure

The back pressure at the exit has a great influence on the flow behavior of the gas stream in the nozzle. A shock wave may occur in the divergent section of the nozzle under certain conditions, which will change the flow from supersonic to subsonic. The effect of back pressure on the flow behavior is simulated, under the conditions that the inlet parameters remain constant (8 MPa and 303 K). Figure 3-4 show the phase diagram and the pressure-temperature variations at different back pressure. It is clear that the presence of the shock wave prevents the gas from expanding and leads to the abrupt change in the pressure and temperature, which will cause the evaporation of the condensed carbon dioxide. It also can be seen that with the decrease of the back pressure, the gas flow expands more and the temperature and pressure are lower before the occurrence of the shock wave. As indicated in figure 4, when the back pressure is 5.6 MPa, the pressure-temperature curve is kept outside of the two-phase region and the flow will remain in a single phase all though the nozzle. In the supersonic separator, the shock wave should be enforced into the diffuser and the condensed liquid should be separated from the natural gas before the occurrence of the shock wave.

5. Conclusion

The performance of the Laval nozzle in removing carbon dioxide from the natural gas is simulated by Fluent. Due to the expansion in the Laval nozzle of supersonic separator, it is possible to cool the gas to a sufficient extent that the liquefaction of carbon dioxide can occur. Shock wave occurs in the divergent section of the supersonic nozzle with the increase of the back pressure. The emergence of the shock wave significantly impairs the expansion process, destroying the liquefaction condition.
Therefore, the back pressure should be reasonably controlled to avoid the emergence of the shock wave in the nozzle.

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