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An unconventional supersonic liquefied technology for natural gas

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Abstract

We proposed a novel supersonic liquefied technology to liquefy the natural gas to LNG liquids. The Peng-Robinson equation of state combined with a thermodynamic process modeling package was employed to calculate the gas dynamics parameters in the supersonic liquefied process. The method to intensify the liquefied process was also discussed. The results show that natural gas expands to supersonic velocities with leading to the low pressure and temperature of 1611.5 kPa and -118.86 ˚C at the nozzle exit, respectively. The pressure-temperature (P-T) curves remaining in the dense phase region indicates that the supersonic liquefied apparatus can successfully liquefy the natural gas to LNG liquids. A large expansion ratio improves the performance of a supersonic liquefied apparatus since natural gas is expanded further. Inserting a long constant conduit between the Laval nozzle and diffuser deepens the supersonic liquefied process.

Keywords: Supersonic liquefaction; Liquefied natural gas; Laval nozzle

1. Introduction

With the rapid development of the global economy, the demand for energy supply is increasing continuously in the last two decades. In the current energy structure, the fossil fuels, major forms of coal, oil and natural gas, play an important role in the worldwide economy [1-37]. For example, in China more than 90% energy is met with the fossil source [1]. At the same time, it has been anticipated that global CO_2 emissions will continuously rise to 30% before 2030, although with the use of the nuclear and renewable energies [2, 3]. Therefore, alternative fuels should be utilized to simultaneously satisfy the increasing growth of energy demand and the reductions of CO_2 emissions.

Natural gas is gaseous mixture, primarily composed of methane (CH_4), ethane (C_2H_6), propane (C_3H_8) and butane (C_4H_{10}), with some higher alkanes (C_5^+), carbon dioxide (CO_2), hydrogen sulfide (H_2S), nitrogen (N_2) and a small amount of water vapor (H_2O). As a clean fossil fuel, natural gas can reduce the emissions of greenhouse gas compared with other traditional fuels. However, natural gas is not easy to be stored or transported due to its low

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density. For instance, natural gas pipelines are impractical across oceans. Therefore, people use the natural gas in other ways, i.e. compressed natural gas (CNG) and liquefied natural gas (LNG) [4-10]. At normal pressure (101,325 Pa), the natural gas can be condensed into liquids, namely, LNG, when the temperature reaches approximately −162 °C. This liquefied process causes that LNG only takes up approximately 1/600 volume of natural gas in the gaseous state. Hence, LNG can be easily stored and transported owing to the decrease of volume. In typical liquefied natural gas, methane (CH_4) is more than 90%, with some ethane (C_2H_6), propane (C_3H_8) and butane (C_4H_{10}) and a small amount of heavier hydrocarbons (C_5^+). The density of LNG is dependent on the temperature, pressure and composition, and the typical density is about 0.41 kg/L to 0.5 kg/L. From BP's statistical review [11], the growth of natural gas trade was about 10.1% and the LNG shipments grew by more than 22.6% in 2010. At present, 30.5% of global gas natural trade is accounted by LNG. In addition, the natural gas demand increases strongly in China, and most of them is driven by LNG [12, 13]. Therefore, some novel technologies should be developed to improve the natural gas processing.

In this paper, we proposed a novel liquefied technology, namely, a supersonic liquefied apparatus, which condenses natural gas into LNG liquids. The liquefaction parameters of natural gas are analyzed and the numerical experiment to intensify the liquefied processing is performed.

2. Supersonic liquefied apparatus

The principle of the supersonic liquefaction is to utilize the reduction of the stream static pressure and temperature by gas expansion to supersonic velocity. Hence, the key component of the supersonic liquefied apparatus, as shown in Fig. 1, includes a Laval nozzle, which accelerates the natural gas to supersonic speed. The nozzle contains a convergent, throat and divergent section, which forms the subsonic, critical and supersonic zone. The convergent section accelerates the natural gas and the sound velocity is achieved in the throat region. To get this critical condition, the inlet diameter should be greater than $\sqrt{5}$ times of the throat diameter. The length of convergent section should be equal to or larger than the throat diameter. Moreover, the convergent curvature is typically designed to keep the acceleration of the natural gas velocity uniform. In this work, the equation to calculate the convergent curvature is expressed as:

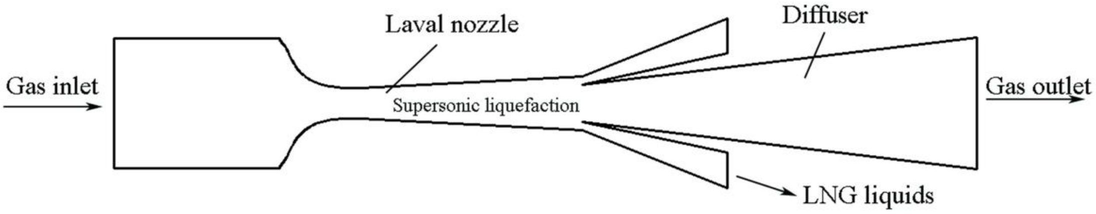

Fig. 1 Schematic diagram of supersonic liquefied apparatus.

$$\left(\frac{D_{cr}}{D}\right)^2 = 1 - \left[1 - \left(\frac{D_{cr}}{D_1}\right)^2\right] \frac{\left[1 - \left(\frac{x}{L}\right)^2\right]^2}{\left[1 + \frac{1}{3}\left(\frac{x}{L}\right)^2\right]^3} \tag{1}$$

where D_1 and Dcr are the inlet and throat diameter, respectively, m. L is the convergent length, m; x is an arbitrary cross section, m; and D is the diameter at an arbitrary cross section of x, m.

The dimensions of supersonic liquefied apparatus are shown in Table 1. The critical cross-section area is 0.001058 m^2. The nozzle inlet and outlet areas are 0.01327 and 0.005255 m^2, respectively.

Table 1. Dimensions of supersonic liquefied apparatus

Parameter	Value	Unit
Nozzle inlet diameter	130.00	mm
Nozzle throat diameter	36.71	mm
Nozzle outlet diameter	81.80	mm
Nozzle converging length	109.56	mm
Nozzle diverging length	564.81	mm
Diffuser outlet diameter	130.00	mm
Diffuser length	579.42	mm
Total length	1373.79	mm

3. Methodology

In this investigation, FLUENT and HYSYS software were combined to solve the mathematical model. FLUENT is a numerical computing solver and HYSYS is a process modeling package. The connection of the two simulation software provides an efficient computational way to investigate the novel supersonic liquefied process. The natural gas properties can be calculated by HYSYS package with common thermodynamic methods. A mathematical model of the natural gas flows based on FLUENT solver was utilized to solve the governing equations, namely, the continuity (mass), momentum and energy equations. The SIMPLE algorithm [15, 16] was used to couple the velocity and pressure field. The standard k-ε model was employed here due to the high Reynolds number in the supersonic liquefied apparatus [17].

3.1. Equation of state

As the supersonic liquefied process is performed in high pressure and low temperature, natural gas property is far from a perfect gas. An equation of state (EOS) plays a significant role in evaluating the gas properties. Errors in pressure and temperature evaluation lead to the poor prediction of the flow structures using the perfect gas model. Therefore, a real gas model should be used to calculate the natural gas thermodynamic properties, including density, viscosity, enthalpy, entropy and so on. So far a lot of equations of state have been proposed to predict thermodynamic properties of pure compounds and mixtures [18-29]. The Peng-Robinson EOS (PR EOS) [21] is the most popular equation of state for natural gas in the chemical industry. A slightly better performance around critical conditions makes the PR EOS somewhat better suited to gas/condensate systems. Hence, the PR EOS is selected for the computational works, which is expressed as follows:

$$P = \frac{RT}{V-b} - \frac{a(T)}{V(V+b)+b(V-b)} \tag{2}$$

in which

$$b = 0.077796 \frac{RT_c}{P_C} \tag{3}$$

and

$$a(T) = \left(0.45724 \frac{R^2 T_C^2}{P_C} \right) \alpha(T) \tag{4}$$

where

$$\alpha(T) = \left[1 + k(1 - \sqrt{T/T_c}) \right]^2 \tag{5}$$

$$k = 0.37464 + 1.54226\omega - 0.26992\omega^2 \tag{6}$$

where P, T and V are the pressure, temperature and volume, respectively. R is the gas constant; the parameter a is a function of temperature; b is constant, k is a constant characteristic of each substance and ω is the acentric factor. Tc and Pc are the critical temperature and the critical pressure, respectively.

For a multi-component mixture, such as natural gas, mixing laws are used to calculate the parameters a and b. In this work, the Van Der Waals mixing rules [30, 31] were applied to obtain the equation of state parameters for a mixture from those of the pure components. The mathematical expressions of this mixing rule can be written,

$$a = \sum_{i=1}^{n} \sum_{j=1}^{n} x_i x_j \sqrt{a_i a_j} (1 - k_{ij}) \tag{7}$$

$$b = \sum_{i=1}^{n} x_i b_i \tag{8}$$

where x is molar fraction; n is total number of the gas components; k_{ij} is the binary interaction parameter between components i and j.

4. Results and discussion

4. 1. Verification of computational model

Some published papers have investigated the fluid dynamic characteristics in the Laval nozzle. In this section, our numerical technique is validated with the recent available data before we apply it to our designed supersonic separator for liquefied natural gas. The geometry used in Arina's investigation [32], is shown in Fig. 2. The equation to calculate the nozzle dimensions is as follows.

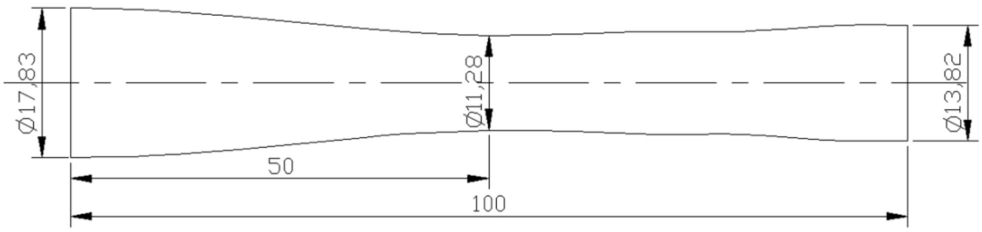

Fig. 2 Nozzle geometry in Arina's work.

$$\begin{cases} A(x) = 2.5 + 3\left(\dfrac{x}{x_{th}} - 1.5\right)\left(\dfrac{x}{x_{th}}\right)^2 & x \le x_{th} \\ A(x) = 3.5 - \dfrac{x}{x_{th}}\left(6 - 4.5\dfrac{x}{x_{th}} + \left(\dfrac{x}{x_{th}}\right)^2\right) & x \ge x_{th} \end{cases} \tag{9}$$

where $A_{\text{throat}} = 100$ mm^2, length $x_{\max} = 100$ mm and the throat placed at $x_{\text{th}} = 50$ mm.

The working fluid was air. The inlet total temperature and pressure were 288 K and 10^5 Pa, respectively. The exit pressure is assigned of 83049 Pa. The Redlich-Kwong equation of state was employed to predict the dynamics parameters of gas fluids in this study. The same geometry and condition were utilized to compare the fluid dynamics characteristics in the Laval nozzle. Fig. 3 shows the pressure distribution in the Laval nozzle in our numerical simulation and Arina's work. It can be seen that the same flow behavior were obtained and the shockwave position was accurately captured by our simulation method. Therefore, our numerical results agree with Arina's results well.

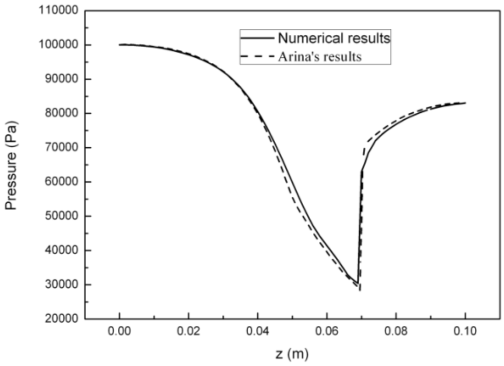

Fig. 3. Pressure profiles for nozzle flow.

4. 2. Grid independence test

In the computational simulation, the mesh of the geometry has an important effect on the accuracy and stability. For grid independence, we should start with coarse, moderate and fine grid to get grid independent solution. Three simulations had been done with a total of 19,823, 91,977 and 180,050 quadrilateral cells for the supersonic liquefied apparatus with same boundary conditions. The Mach number at the nozzle exit is shown in Table 2. It can be seen that there is little difference between these simulations results. Even 19,823 cells provide a sufficient grid independency. However, the calculation was performed with 91,977 cells for exactly judging the shock wave positions.

Table 2. Grid independence tests

Case number	Number of cells	Mach number	Error (%)
1	19 823	1.9837	0
2	91 977	1.9833	0.020

4. 3. Liquefied parameters of natural gas

The gas dynamics parameters in the supersonic liquefied apparatus are computationally simulated with the standard k-ε model and Peng-Robinson equation of state. Figs. 4-6 depict the Mach number, static pressure and temperature in the liquefied process. It can be seen that

Laval nozzle accelerates natural gas to supersonic velocities with a Mach number of 2.074. This expansion of gas leads to the low pressure and temperature. The average static pressure and temperature are 1,611.5 kPa and -118.86 ˚C at the nozzle exit, respectively. The natural gas starts to condense to LNG liquids when the gas pressure drops down to the cricondenbar and the temperature becomes lower than the condensation temperature, and if there is enough time for condensation.

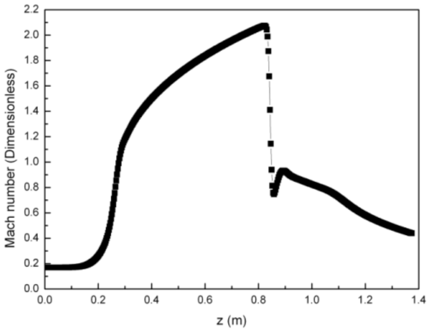

Fig. 4 Mach number.

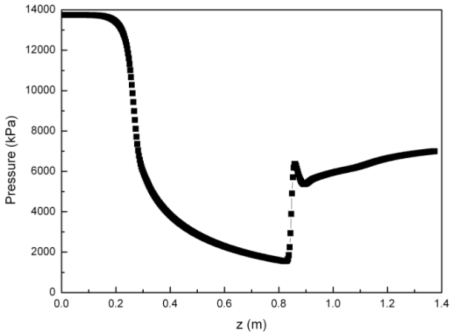

Fig. 5. Static pressure.

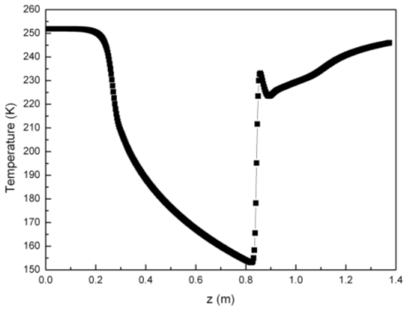

Fig. 6. Static temperature.

The phase envelope of natural gas, composed of 96.044% CH_4, 2.98% C_2H_6 and 0.976% C_3H_8, is calculated by HYSYS, as shown in Fig. 7, where the variation of the temperature with the pressure for the supersonic liquefaction are plotted. Fig. 7 indicates that the pressure-temperature lines remain in gas-liquid coexistence region even can penetrate into the dense phase region. Therefore, the supersonic liquefied apparatus can successfully liquefy the natural gas into LNG liquids using the supersonic expansion characteristics.

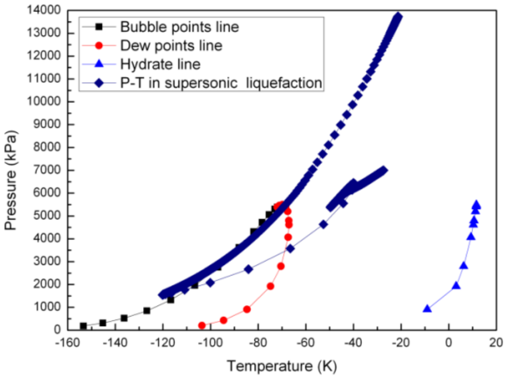

Fig. 7. Phase envelope and P-T in supersonic liquefaction.

Moreover, there is a shock wave in the supersonic liquefaction, leading to the discontinuous and irreversible changes in the thermodynamics properties of fluids. For instance, the gas speed changes from supersonic velocity to subsonic velocity, as seen in Fig. 4. The condensed LNG liquids should be separated from the natural gas mixture before the emergence of the shock wave. Therefore, the shockwave is enforced into the diffuser instead of in the Laval nozzle for a supersonic liquefied apparatus. Otherwise, the condensed LNG liquids re-evaporate into the gas stream due to the high temperature as a result of the shockwave in the nozzle.

4. 4. Effects of expansion ratio on the supersonic liquefied process

Expansion ratio is defined as the ratio of the nozzle exit section area to the nozzle throat area. The effects of expansion ratio on the supersonic liquefied process are shown in Fig. 8. The increases of the expansion ratio result in the lower pressure and temperature to deepen the supersonic liquefied process of natural gas. When the expansion ratio is 1.14, it is a shallow cooling, and the natural gas cannot be liquefied into LNG liquids in the supersonic apparatus. Increasing the expansion ratio to 1.52, the pressure-temperature lines pass over the dew point line and go into gas-liquid coexistence region. If the expansion ratio is greater than 2.50, the pressure-temperature lines will go across the bubble point line and get into the dense region. In the deep cryogenic processing, the natural gas can be liquefied to LNG production. That is, a large expansion ratio improves the performance of a supersonic liquefied apparatus since natural gas is expanded further.

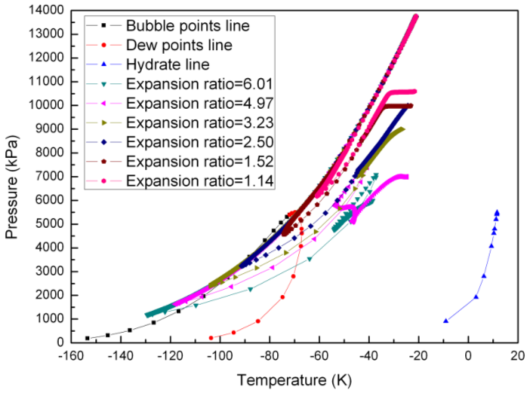

Fig. 8. Effects of expansion ratio on the supersonic liquefied process.

4. 5. Numerical experiment to intensify the liquefied process

In a supersonic liquefied apparatus, a Laval nozzle achieves the adiabatic cooling of natural gas. The isentropic expansion efficiency is general more than 80%. In the static apparatus, the natural gas speed reaches a supersonic velocity, leading to a extremely short residence time of millisecond. However, as mentioned in the above sections, it needs enough time to condense the natural gas to LNG liquids in the supersonic conditions. Here, a constant cross area duct was inserted between the Laval nozzle and the diffuser to increase the residence time of natural gas in the low pressure and temperature, which seems favorable for the condensation. The length of the constant conduit is represented by the length-diameter ratio (L/d), which is the ratio of the conduit length, L, to the nozzle exit diameter, d. Fig. 9 shows that the constant cross area conduit creates a long residence time in low pressure and temperature for natural gas condensation. For instance, the residence time is increased by 1 ms, which can intensify the liquefied process in supersonic velocity, when the length of the constant conduit is 8 times of the nozzle exit diameter. But on the other hand, the static temperature will increase slowly due to the presence of the friction loss in the constant channel. The LNG liquids, only laid in the lower temperature, may re-evaporate into the gas phase due to the rise of the temperature, which will decrease the supersonic liquefied efficiency. Therefore, we suggest that the length of the constant cross area conduit is about 8 times of the nozzle exit diameter for deepening the supersonic liquefied process.

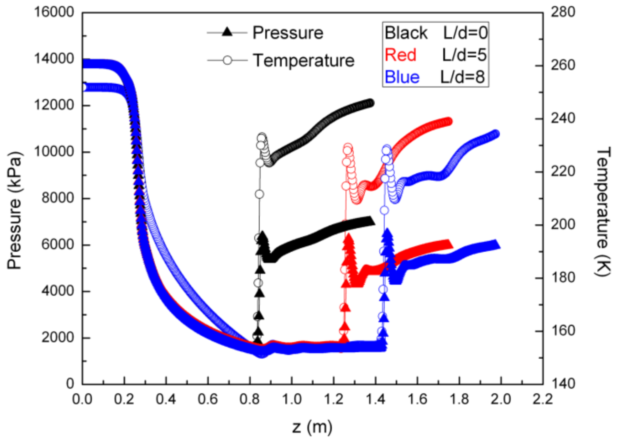

Fig. 9. Effects of the constant conduit on the supersonic liquefied process.

5. Conclusions

A novel methodology was presented to liquefy natural gas to LNG liquids in supercritical conditions. In the supersonic liquefied apparatus, a Laval nozzle expands the natural gas to supersonic velocities, leading to the low pressure and temperature. When the pressure and temperature are below the cricondenbar and the condensation temperature, the natural gas can be condensed to LNG liquids with enough time. However, the high temperature occurs due to the emergence of a shock wave in the divergent section of the Laval nozzle. The rise of the static temperature induces the re-evaporation of liquefied liquids to the disadvantage of the production of LNG. Two methods were proposed to intensify the liquefied process including expanding the expansion ratio and inserting a long constant conduit between the Laval nozzle and diffuser.

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