Study on variation of thermodynamic parameters in vehicle-mounted LNG gas cylinder under static state

Si Huang¹, Tiankun Yi¹,³, Jiawei Guo¹, Gang Yang² and Li Xia²

¹ School of Mechanical and Automotive Engineering, South China University of Technology, Guangzhou, Guangdong 510641, China
² Guangdong Institute of Special Equipment Inspection and Research, Foshan, Guangdong 528000, China
³ E-mail: 707774238@qq.com

Abstract. To study the variation regularities of the mass, evaporation, temperature, and pressure of LNG in the gas cylinder with time, LNG was used as the working medium, and three specifications of LNG gas cylinders (150L, 330L, and 450L) were selected for heat transfer calculation. For the liquefied medium in the LNG gas cylinders, a saturated homogeneous model was adopted. For the heat transfer calculation of the adiabatic layer of LNG gas cylinders, a multilayer insulation structure model of radiation and heat conduction was adopted. According to the thermodynamic relations in the saturated state, the relations between the temperature and the physical parameters of the liquefied medium were fitted. The calculation method and calculation model were verified by the experimental data of liquid nitrogen for three specifications of LNG cylinders (150L, 330L, and 450L). According to the calculation model, the variation regularities of LNG thermodynamic parameters with time were obtained at a normal temperature. In the initial period of evaporation, the increasing temperature rate of LNG, the evaporating rate of LNG, and the pressure-boost rate of LNG in the gas cylinder are slow, and then these rates increase. The final mass of boil-off gas (BOG) produced by the phase change finally reaches 0.68% of the total mass of the medium in the gas cylinder.

Nomenclature

| Symbol | Definition                          |
|--------|------------------------------------|
| A      | Area [m²]                          |
| a      | Accommodation coefficient [0.9 for air] |
| B      | Length of outer container cylinder [m] |
| b      | Length of inner container cylinder [m] |
| C      | Total emission coefficient [W·m⁻²·K⁻⁴] |
| Cₐm    | Emission coefficient of adiabatic layer [0.1 W·m⁻²·K⁻⁴ for aluminum foil] |
| Cₐ∅    | Emission coefficient of black body [5.67 W·m⁻²·K⁻⁴] |
| Cₐr    | Emission coefficient of gas cylinder [0.45 W·m⁻²·K⁻⁴ for stainless steel] |
| cₚ     | Specific heat of medium [J·kg⁻¹·K⁻¹] |
| D      | Gas cylinder diameter [mm]         |
| d      | Diameter of glass fiber [1μm]      |
| M      | Molecular weight [0.029 kg·mol⁻¹ for air] |
| m      | Mass of medium [kg]                |
| n      | The number of aluminum foil shields. |
**1. Introduction**

Liquefied natural gas (LNG), as clean energy, is used as a fuel for automobiles with its low-carbon and environmentally friendly characteristics. The LNG fueling system is composed of gas cylinders, carburetors, pressure reduction valves, and other components. LNG is stored in cryogenic gas cylinders in liquid form. With the transfer of external heat, the LNG will evaporate and the pressure will increase accordingly. When the internal pressure of the gas cylinder reaches the popping pressure, the pressure reduction valve will automatically open to reduce the pressure to ensure the safety of the gas cylinder [1].

Over the years, many scholars carried out various researches on the medium in cryogenic storage tanks. In terms of theoretical models, Rotenberg [2] proposed a saturated homogeneous model, which assumed that the temperature of the gas phase and liquid phase in the tank is equal, and the system pressure is the saturated pressure of the liquid phase. Hochstein [3] proposed an integral model to calculate the pressure of the medium and regarded the liquid phase as a saturated homogeneous medium and the gas phase as an overheating gas. Gursu [4] proposed a homogeneous surface evaporation model, which assumed that the cryogenic medium is in a thermal equilibrium state. It can accurately predict the pressure-boosting regularity and the heat leakage in the storage tank. In terms of
numerical calculations, basing on the mass and energy conservation equations, Qu et al. [5] developed a code for calculating the generation rate of BOG (boil-off gas) and temperature in LNG storage tanks, and verified the accuracy of the calculation method through experimental data. Sharafian et al. [6] carried out heat transfer analysis and calculation to study the effect of initial temperature of LNG, vacuum degree of adiabatic layer, and thermal conductivity of adiabatic layer on storage time. Huerta et al. [7] derived the analytical solutions for isobaric evaporation in equilibrium and non-equilibrium states and studied the effect of initial filling rate on heat transfer, boil-off rate, and temperature of BOG. In terms of experimental research, Li et al. [8] used liquid nitrogen as the working medium to study the effect of the initial liquid level height on the heat leakage of the cryogenic storage tank through experiments, and the research showed that the heat leakage has a cubic relationship with the variation of the liquid level. Hulsbosch et al. [9] used liquid nitrogen as the working medium to study the pressure-boosting regularity of cryogenic storage tanks under fire situations through experiments. Kang et al. [10] studied the pressure-boosting regularity of the medium in the tank under different filling rates through experiments, and the study showed that tanks with higher filling rates have a faster rate of pressure increase.

In terms of heat transfer calculation research on adiabatic layer, Resis [11] and Xie et al. [12] carried out heat transfer calculations on the adiabatic layer, and they considered the radiation heat transfer, solid heat conduction, and convective heat transfer. Shen et al., [13] and Zhu et al. [14] carried out heat transfer calculations on the adiabatic layer, and they considered the radiation heat transfer of the adiabatic layer and the heat conduction of solid and gas. When Zheng et al. [15] and Wang et al. [16] calculated the heat transfer of the adiabatic layer, they also considered radiation heat transfer, solid and gas heat conduction, and verified the accuracy of the calculation method through experiments.

The above studies are mainly aimed at the variations of the thermodynamic parameters of the liquefied medium in the cryogenic storage tank. There is no universal calculation method yet, and the lack of experimental data also makes the verification of the theoretical model difficult. Therefore, in this study, a saturated homogeneous model was adopted for the liquefied medium in the LNG gas cylinder. A multi-layer insulation structure model of radiation and heat conduction was adopted for the heat transfer calculation of the adiabatic layer. First, the accuracy of the calculation method was verified by the pressure-boosting experiment of liquid nitrogen. Then LNG was used as the working medium to study the variation regularities of the mass, evaporation, temperature, and pressure of LNG in the gas cylinder with time, and provide technical support for the safety of the vehicle-mounted LNG gas cylinder.

2. Models

2.1. Vacuum multilayer insulation
Three universal LNG gas cylinders were selected as the research objects. Table 1 lists the basic parameters of the LNG gas cylinders. As shown in Figure 1, the LNG gas cylinder is a multi-layer vacuum insulation structure, which wraps the multi-layer aluminum foil on the outer surface of the inner container to achieve the purpose of effective thermal insulation.

| Table 1. Basic parameters of three types of LNG gas cylinders. |
|-----------------|-----------------|-----------------|-----------------|
| Product type    | CDPW500-150     | CDPW500-330     | CDPW600-450     |
| $V$ (L)         | 150             | 330             | 450             |
| $\phi$          | 90%             | 90%             | 90%             |
| B (mm)          | 1085            | 1410            | 1565            |
| b (mm)          | 980             | 1270            | 1410            |
| $D_1$ (mm)      | 410             | 533             | 575             |
| $D_2$ (mm)      | 430             | 559             | 624             |
| $D_3$ (mm)      | 449             | 579             | 646             |
Figure 1. Schematic diagram of gas cylinder.

There are three forms of heat transfer in the vacuum multilayer insulation: (1) Radiant heat transfer in the vacuum layer. (2) Gas heat conduction in the vacuum layer. (3) Solid heat conduction in the adiabatic layer [17]. To simplify the calculation, the equivalent thermal conductivity $\lambda_e$ is introduced to characterize the performance of the adiabatic layer, which can be estimated as [18]:

$$\lambda_e = \lambda_r + \lambda_v + \lambda_c$$  \hspace{1cm} (1)

The thermal conductivity $\lambda_r$ of radiation in the vacuum layer can be estimated as [19]:

$$\lambda_r = \frac{C(T + T_m)(T^2 + T_m^2)\delta_v}{100'}$$  \hspace{1cm} (2)

$$C = \frac{I}{C_v + \frac{I}{C_m}}$$  \hspace{1cm} (3)

The thermal conductivity $\lambda_v$ of the gas in the vacuum layer can be estimated as [20]:

$$\lambda_v = a\delta p_v^{\gamma+1} \left( \frac{R}{8\pi MT_m} \right)^{1/4}$$  \hspace{1cm} (4)

According to the experimental data of the relations between thermal conductivity, mechanical pressure and density, the empirical formula for the thermal conductivity $\lambda_c$ of the solid in the adiabatic layer can be estimated as:

$$\lambda_c = 0.12 \times 10^{-7} \lambda_r \left[ d (1-u) p_v^\gamma \right]^{1/2}$$  \hspace{1cm} (5)

The temperature distribution in the vacuum multilayer insulation is so significant. From this, we can infer the heat transfer relations between radiant heat conduction, gas heat conduction, and solid heat conduction. The simplified temperature distribution in the vacuum multilayer insulation can be estimated as [21]:

$$T_i = \left[ T^4 + \frac{k}{n+1} \left( T_r^4 - T_i^4 \right) \right]^{1/4}$$  \hspace{1cm} (6)

If it is assumed that the aluminum foil shields are arranged equidistantly as shown in Figure 1, and the thermal boundary layer of the adiabatic layer in the middle position of jacket space. The temperature $T_m$ of the thermal boundary layer of the adiabatic layer can be estimated as:
When the ambient temperature $T_s$ is 298K and the vacuum $P$ for jacket space is 0.02Pa, Figure 2 shows the relations between the equivalent thermal conductivity $\lambda_e$ and the temperature $T$ of medium in the gas cylinder, which are calculated by Equations (1) - (7). As shown in Figure 2, the equivalent thermal conductivity $\lambda_e$ increases slightly with the increase of the temperature $T$ in the gas cylinder. The equivalent thermal conductivity $\lambda_e$ increases with the increase of the volume of the gas cylinder. When the gas cylinder volume is 330L and the vacuum $P$ for jacket space is 0.02Pa, Figure 3 shows the relations between the ambient temperature $T_s$ and the equivalent thermal conductivity $\lambda_e$. As shown in Figure 3, the equivalent thermal conductivity $\lambda_e$ increases with the increase of the ambient temperature $T_s$, and the increasing trend accelerates slightly.

$$T_w = \left( \frac{1}{2} T_s + \frac{1}{2} T_1 \right)^{1/4}$$  \hspace{1cm} (7)

2.2. Calculation method

Because of the small volume of the LNG gas cylinders studied, the medium quickly reaches thermal equilibrium, and temperature stratification is not easy to form. Therefore, the saturated homogeneous model was used to calculate the heat transfer of these gas cylinders. According to the law of thermodynamics, there is the following relation in $\Delta t$ time interval [22]:

$$Q = (c_{pl} m_i + c_{pg} m_g) \Delta T = \Phi \Delta t$$  \hspace{1cm} (8)

The main component of LNG is methane. According to the thermodynamic relations of methane in the saturated state [23-24], the relations between liquid specific heat $c_{pl}$, gas specific heat $c_{pg}$, pressure $p$, and heat of vaporization $r$ and temperature $T$ were regressed in the temperature range of 105K~165K:

$$\begin{align*}
c_{pl} & = 3.370 \times 10^4 - 990.301 T + 12.086 T^2 - 6.557 \times 10^{-2} T^3 + 1.348 \times 10^{-4} T^4 \\
c_{pg} & = 4.881 \times 10^6 - 1511.677 T + 18.314 T^2 - 9.874 \times 10^{-2} T^3 + 2.015 \times 10^{-4} T^4 \\
p & = 1.869 - 5.628 \times 10^{-2} T - 5.246 \times 10^{-7} T^2 + 8.473 \times 10^{-10} T^3 + 6.739 \times 10^{-13} T^4 \\
r & = 1.460 \times 10^8 + 1.528 \times 10^7 T - 201.412 T^2 + 1.124 T^3 - 2.540 \times 10^{-3} T^4
\end{align*}$$  \hspace{1cm} (9)

The heat leakage rate $\Phi$ of vacuum multilayer insulation can be estimated as [25]:

$$\Phi = \lambda_e A \frac{(T - T_s)}{\delta}$$  \hspace{1cm} (10)

The calculated area $A$ can be calculated according to the geometric average area [26]:

$$A = \sqrt{A_1 A_2}$$  \hspace{1cm} (11)
\[ \begin{aligned}
A_1 &= \pi D_1 b + 1.3802 \frac{D_1^2}{2} \\
A_2 &= \pi D_2 B + 1.3802 \frac{D_2^3}{2}
\end{aligned} \] (12)

Substituting Equation (10) into Equation (8) to obtain the following relationship:

\[ \lambda_c A \left( T - T_s \right) \Delta t = \left( c_p m_l + c_p m_g \right) \Delta T \] (13)

Using the forward-difference method to discretize the Equation (13), and using the average value of the initial temperature \( T_i \) and the final temperature \( T^{i+1} \) to calculate the temperature \( T \) on the left side of the Equation (13):

\[ \lambda_c A \frac{T_i + T^{i+1}}{2} - T_s \Delta t = c_p m_l (T_i - T^{i+1}) + c_p m_g (T_i - T^{i+1}) \] (14)

From Equation (14), the final temperature \( T^{i+1} \) of the medium in the gas cylinder can be expressed as:

\[ T^{i+1} = \frac{\lambda_c A (T_i + T^{i+1})/2 - T_s \Delta t}{2 \delta (c_p m_l + c_p m_g) + \lambda_c A \Delta t} \] (15)

Therefore, the final boil-off rate \( \alpha^{i+1} \), the mass of the gaseous medium \( m_g^{i+1} \), and the mass of the liquid medium \( m_l^{i+1} \) have the following relationship:

\[ \alpha^{i+1} = \frac{\lambda_c A \left( T_i + T^{i+1} \right)/2 - T_s}{r^l m_l^i} \] (16)

\[ m_g^{i+1} = \alpha^{i+1} m_l^i, \quad m_l^{i+1} = m_l^i - m_g^{i+1} \] (17)

The known conditions include gas cylinder volume \( V \), filling rate \( \phi \), and ambient temperature \( T_s \). The initial conditions are:

\[ m_l^0 = \rho_0^0 V \phi, \quad T^0 = 112.15K, \quad p^0 = 0.1MPa \]

![Figure 4. Calculation procedure.](image-url)
Because the popping pressure of the reduction valve is 1.6MPa, the calculation will be terminated when the pressure of the medium in the gas cylinder reaches this pressure value. The calculation procedure is shown in Figure 4.

2.3. Experimental verification of liquid nitrogen
In order to verify the accuracy of the calculation method, three specifications of LNG gas cylinders (150L, 330L, and 450L) were selected for the pressure-boost experiment. Figure 5 shows the schematic diagram of the LNG gas cylinder experiment device. Liquid nitrogen was used as the experimental medium, because of its abundant resources, easy liquefaction, relatively low price, stability, safety, and easy operation.

The main steps of the experiment are as follows:
First, connecting the port for vacuum pumping to an external vacuum system for vacuuming while observing the vacuum gauge. In order to reduce the convection and heat conduction of the residual gas in the adiabatic layer, it is necessary to ensure that the vacuum for the jacket space is less than 0.02 Pa during the experiment, and the flange of the port for vacuum pumping shall be leak-checked before vacuuming.

Then putting the LNG gas cylinder on the electronic weighing machine, connecting all accessories as shown in Figure 5, and filling the 150L gas cylinder with liquid nitrogen and left standing to obtain the thermal equilibrium. The initial filling rate is determined to be 90% by weighing. After filling, closing the liquid inlet and outlet valves.

After the filling was completed, starting to measure the pressure of the medium in the gas cylinder. When the ambient temperature is 298K, according to the actual situation of the experimental gas cylinder, selecting the atmospheric pressure gauge to measure the relevant parameters and recording experiment data as the pressure gauge increases by one scale.

After the reduction valve opening for the first time, the experiment was completed. Then filling the 330L and 450L gas cylinders with liquid nitrogen, repeating the above steps at a full rate of 90%, and carrying out multiple experiments.

![Figure 5. Experimental apparatus.](image)

1-Molecular Sieve. 2-LNG gas cylinder. 3-Thermal Insulated Material. 4-Stop Valve. 5-Port for Vacuum Pumping. 6-Port for liquid filling. 7-Reduction valve. 8-Multimeter/Data Acquisition System. 9-Computer. 10-Atmospheric Pressure Gauge. 11-Vacuum Ionization Gauge. 12-Vacuum Gauge. 13-Electronic Weighing machine.

Experiment 1 and Experiment 2 both used liquid nitrogen as the medium. Two pressure-boosting experiments were carried out on the three specifications of LNG gas cylinders in Table 1 to obtain the regularity of pressure variation with time.

As shown in Figure 6, the calculated results are relatively close to the experimental data, and the variation regularities of the pressure are consistent: the pressure in the gas cylinder increases slowly in
the initial period, but the pressure rises faster with the increase of time. At the same time, the pressure and time are approximately the cubic power relationship. Small gas cylinders have a smaller thermal capacity. The absorbed heat will cause great changes in its state, and the pressure increases more quickly.

Figure 6. Curve of the pressure $p$ in the gas cylinder with time $t$. 
3. Discussion

3.1. The relations between the mass of the LNG and the time

With the heat transfer of external heat, the LNG in the gas cylinder will evaporate continuously, and the mass of the BOG will increase continuously. Figure 7 is the variation curve of the non-dimensional mass of the LNG \( m_l^* (m_l^* = m_l / m_l^0) \) with time \( t \). As shown in Figure 7, the mass of the LNG \( m_l \) in the gas cylinder shows a downward trend with time. When the volume of the gas cylinder decreases, the evaporation of LNG accelerates. When the gas cylinder volume is large, LNG evaporates slowly.

Figure 8 is the variation curve of the non-dimensional mass of the BOG \( m_g^* (m_g^* = m_g / m_g^0) \) with time \( t \). As shown in Figure 8, from the initial state to the maximum working pressure of the gas cylinder, the mass of the BOG \( m_g \) finally reaches 0.68% of the total mass of the medium.

![Figure 7. Curve of the non-dimensional mass \( m_l^* \) of the LNG with time \( t \).](image7)

![Figure 8. Curve of the non-dimensional mass \( m_g^* \) of the BOG with time \( t \).](image8)

3.2. The relations between the temperature of the LNG and the time

Figure 9 shows the calculated variation curve of the temperature \( T \) of the LNG in the gas cylinder with time \( t \). As shown in Figure 9, from the initial state to the maximum working pressure of the gas cylinder, the temperature \( T \) of the LNG rises from 112.15K to 160K. The temperature \( T \) of the LNG in the gas cylinder rises slowly in the initial period, and the temperature rises faster with the continuous evaporation of LNG. When the volume of the gas cylinder decreases, the rising rate of the temperature \( T \) of the LNG increases. When the gas cylinder volume is large, the temperature \( T \) of LNG rises slowly.

![Figure 9. Curve of the temperature \( T \) of the LNG with time \( t \).](image9)
3.3. The relations between the pressure of the LNG and the time

Figure 10 shows the calculated variation curve of the pressure $p$ of the LNG in the gas cylinder with time $t$. Through comparison, it is found that the pressure rise trends of different types of gas cylinders are consistent: the pressure $p$ of the LNG rises slowly in the initial period, and the pressure $p$ of the LNG rises faster with the continuous evaporation of LNG. This is because when the pressure $p$ of the medium increases, the corresponding latent heat of vaporization and specific heat will decrease, and the pressure variation ($dp/dt$) will increase when the same heat is absorbed. Besides, in the initial period, gas and liquid coexist, and the absorbed heat is used to provide the energy for the latent heat of gasification. When the pressure $p$ rises to a certain level, the medium enters the supercooled liquid state, and the absorbed heat supplies the energy for the phase change. Absorption of the same heat for the later period of the supercooled liquid can cause more obvious changes in state. Small gas cylinders have a smaller thermal capacity. The absorbed heat will cause a greater change in its state, and its pressure will rise more quickly. Also, when the gas cylinder volume is 150L, the gas cylinder can maintain approximately 229.02 hours at a normal temperature. When the gas cylinder volume is 330L, it can maintain 334.52 hours at a normal temperature. When the gas cylinder volume is 450L, it can maintain about 525.72 hours at a normal temperature.

![Figure 10. Curve of the pressure $p$ of the LNG with time $t$.](image)

4. Conclusions and perspectives

In this study, LNG was used as the working medium, and three universal gas cylinders were selected for heat transfer calculation. The following conclusions are summarized:

(1) As the external heat transfer causes the LNG in the gas cylinder to evaporate, the mass of the BOG will continue to increase. The mass of LNG in the gas cylinder shows a downward trend over time. When the volume of the gas cylinder is small, the evaporation of LNG is accelerated. When the gas cylinder volume is large, LNG evaporates slowly. The mass of BOG produced by the phase change continues to increase, eventually reaching 0.68% of the total mass of the medium in the cylinder.

(2) From the initial state to the maximum working pressure of the gas cylinder, the temperature of the LNG in the gas cylinder rises from 112.15K to 160K. The temperature of the LNG rises slowly in the initial period, and its temperature rises faster with the rapid evaporation of LNG. The rising rate of the temperature of the LNG in the gas cylinder increases with the decrease of the gas cylinder volume.

(3) The pressure of the LNG in the gas cylinder rises slowly in the initial period, and it rises faster with the continuous evaporation of LNG. When the volume of the gas cylinder is small, the pressure in the gas cylinder rises faster. Also, when the gas cylinder volume is 150L, the gas cylinder can maintain approximately 229.02 hours at a normal temperature. When the gas cylinder volume is 330L,
it can maintain approximately 334.52 hours at a normal temperature. When the gas cylinder volume is 450L, it can maintain about 525.72 hours at a normal temperature.

This model fully considers the radiation of the adiabatic layer and the gas heat conduction in the vacuum layer, and it can accurately predict the pressure of small LNG gas cylinders. However, this model does not consider the convective heat transfer caused by the sloshing of the LNG gas cylinder during the movement. The heat transfer of the neck, support, and pipes has not been considered, so it can be further studied in future work. However, in the total heat transfer of LNG gas cylinders for automobiles, the heat transfer of the adiabatic layer is still dominant, which accounts for more than 80% of the total heat leakage. At the same time, the effect of gas leakage on the heat transfer of gas cylinders can be considered in future work.

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