

Rapid Compressed Hydrogen Filling System for Fuel Cell Vehicle

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ABSTRACT

A compressed gas system previously utilized in natural gas vehicles (NGV) was utilized in the Honda FCX. Rapid filling tests of the high-pressure hydrogen tanks of both Type 3 (Aluminum liner) and Type 4 (High Density Polyethylene) were carried out using compressed hydrogen gas. Type 3 tanks showed better performance. This result can be explained in terms of the difference in the heat diffusion coefficient between aluminum and HDPE. The shortest filling time from 1 MPa to 34.4 MPa to enable a gas temperature increase (ΔT) of 45 °C to be observed using Type 3 tanks with a total volume of 156.6 dm³ was found to be 3.6 min., almost the same as the refueling time of present gasoline vehicles.

1. Introduction

The hydrogen storage methods proposed for use with a direct hydrogen fuel cell vehicle (FCV), up to this time, included compressed gas, liquid hydrogen, and hydrogen-absorbing alloy^{(1), (2)}. The present development project focused on the type of compressed gas storage system that had proven performance in natural gas vehicles. Two types of lightweight compressed gas tanks exist for use on vehicles, one with aluminum and the other with plastic as the liner material. The natural gas automobile standard NGV2-2000 defines the former as Type 3 and the latter as Type 4. The Honda Civic GX natural gas passenger car uses a Type 4 tank, which for a given tank volume, is lighter in weight than a Type 3 tank. The present project approached development from the perspective of rapid filling.

It is desirable, in terms of passenger car usability, for FCV filling time to be equal to the refueling time of current gasoline vehicles. The Interface Working Group of the SAE

Fuel Cell Standards Committee is studying a standard that sets the upper limit for gas temperature during filling at 85 °C. The High Pressure Gas Safety Law of Japan stipulates an upper limit for gas temperature while stored in a container at 40 °C. Therefore, the gas temperature increase (ΔT) during filling must not be allowed to exceed 45 °C, which is the difference between these two temperatures.

This report will describe the results from the study of gas behavior during the rapid filling of tanks with compressed hydrogen, simulations of filling behavior, and the factors that affect hydrogen filling mass.

2. Rapid Filling Test with Compressed Hydrogen at 34.4 MPa

Figure 1 shows a filling system from a hydrogen filling station to a high-pressure hydrogen tank (hereafter referred to as a tank). Tests were carried out on a pair of tanks placed side by side, using both a pair of Type 3 and a pair of Type 4

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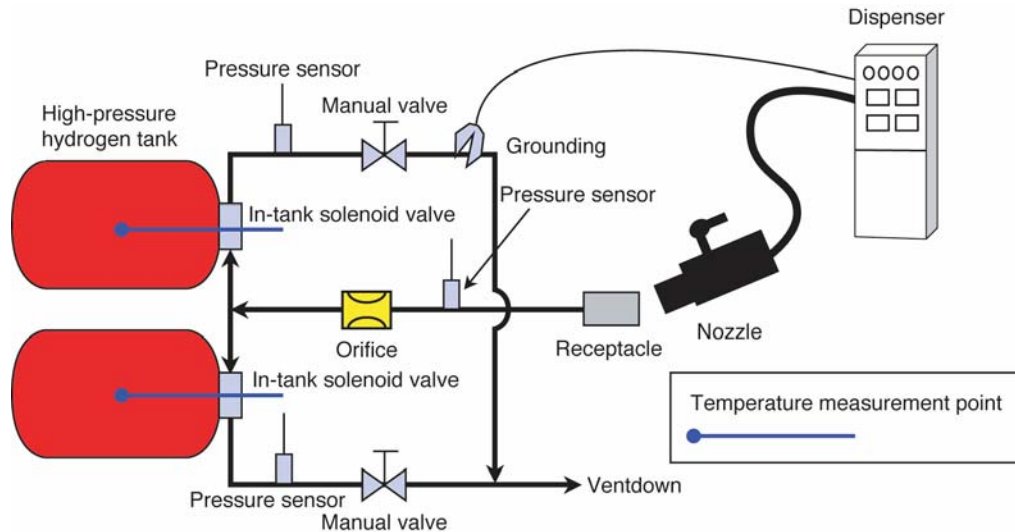


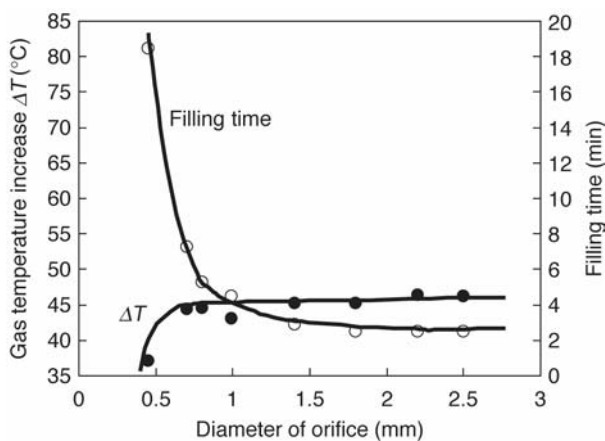
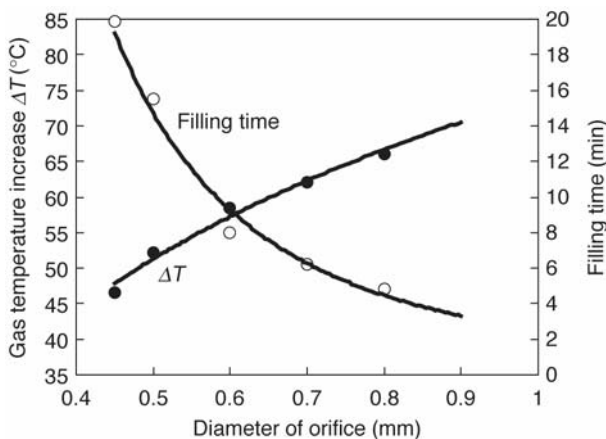
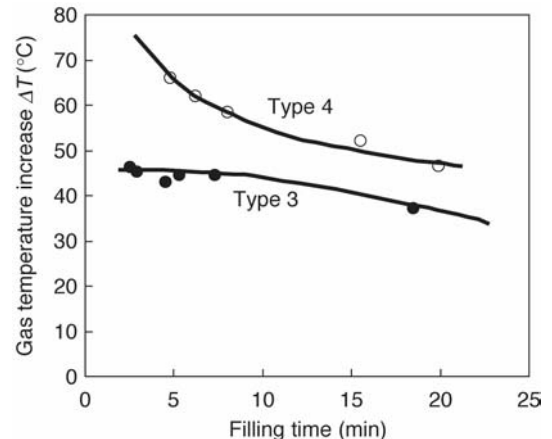
Fig. 1 Schematic illustration of high-pressure hydrogen supply system

tanks. One series of tests was carried out at ambient temperature, with filling speed controlled by an orifice installed before the tanks. The tests were conducted with a hydrogen pressure inside the tanks of 1 MPa (all pressures in

this report are gauge pressures) before filling as an initial condition.

Figures 2 and 3 show the gas temperature increase (hereafter ΔT) as the orifice diameter was varied and its relationship to the filling time for both the Type 3 and Type 4 tanks. The Type 4 tank had a high density polyethylene (hereafter HDPE) liner. For both tank types, the time required for filling decreased as the diameter of the orifice was increased, but ΔT showed a tendency to increase. Although the Type 4 tank is lighter weight than the Type 3 tank, it was found that ΔT showed greater change relative to the change in filling time for the Type 4 tank. With the Type 3 tank, on the other hand, it was found that although the filling time grew shorter when the orifice diameter was 0.7 mm or larger, the ΔT showed almost no change.

Figure 4 organizes the findings for both tanks regarding ΔT during filling and the filling time based on the results in Fig. 2 and Fig. 3. Considering that a larger ΔT is associated with a lower filling efficiency, the Type 3 tank has the advantage in rapid filling. In order to keep ΔT below 45 °C, the Type 3 tank allows rapid filling in five minutes or less, but the Type 4 tank requires slow filling that takes 20 minutes or more.

Fig. 2 Effect of diameter of orifice on gas temperature increase (ΔT) and filling completion time (Type 3)Fig. 3 Effect of diameter of orifice on gas temperature increase (ΔT) and filling completion time (Type 4)Fig. 4 Relationship between filling time and gas temperature increase (ΔT) for Types 3 and 4 high-pressure hydrogen tanks

3. Influence of Initial Hydrogen Pressure Inside the Tank on ΔT

On the vehicle side, refueling could be envisioned to take place at a variety of different residual hydrogen pressure levels. Therefore tests were carried out with the hydrogen pressure varied over 0 – 15 MPa before filling, and the ΔT was determined for each residual hydrogen pressure. The tests were carried out with a 2.5-mm diameter orifice installed on the filling line into the Type 3 tank, and a 0.7-mm diameter orifice on the Type 4 tank.

The results, as shown in Fig. 5, indicate a tendency for both tanks to have a lower ΔT when the hydrogen pressure in the tanks before filling is higher. The amount of change (slope) of ΔT for increasing hydrogen pressure in the tanks before filling was found to be greater in the Type 4 tank than in the Type 3 tank. In the case of the Type 4 tank, as will be explained below, the heat diffusion coefficient of the HDPE is low, so a larger proportion of the heat that is generated by polytropic adiabatic compression is stored in the hydrogen gas.

Consequently, the Type 4 tank is thought to be affected more than the Type 3 tank by the initial hydrogen pressure inside the tank.

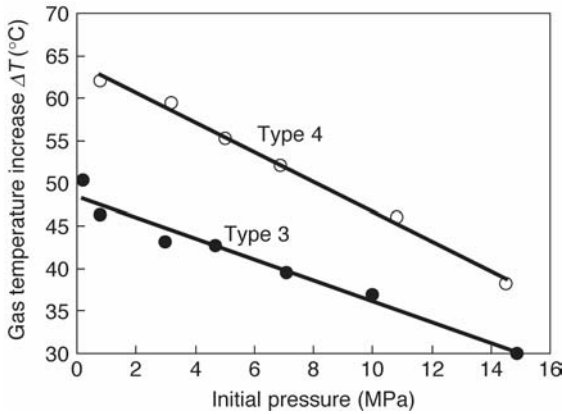


Fig. 5 Dependence of gas temperature increase (ΔT) of Types 3 and 4 high-pressure hydrogen tanks during filling on initial pressure

4. Influence of Ambient Temperature during Filling on ΔT

Similar to that described in the preceding section, refueling could be envisioned to take place under a variety of different temperature conditions on the vehicle side. Therefore the influence of ambient temperature during filling on ΔT was investigated. These tests were carried out only with the Type 4 tank, for which ΔT is larger during filling. The tests were carried out with a 0.7-mm diameter orifice installed and hydrogen pressure in the tank of 1 MPa.

As shown in Fig. 6, the results from testing with ambient temperatures over the range of 4 – 39 °C showed that a higher ambient temperature tends to result in a higher gas temperature peak, while ΔT is not dependent on ambient temperature but remains constant at 62 °C. It was inferred, therefore, that similar results would be obtained with the Type 3 tank.

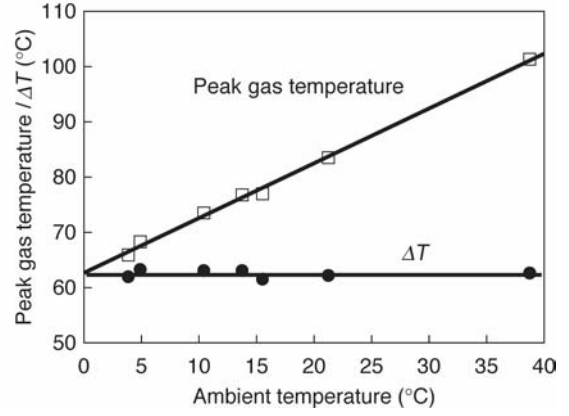


Fig. 6 Effect of ambient temperature on peak temperature and gas temperature increase (ΔT) for Type 4 high-pressure hydrogen tanks

5. Filling Behavior of Hydrogen at 34.4 MPa Pressure in the Type 3 Tank

The Type 3 tank had a total volume of 156.6 dm³. When filling this tank so that the hydrogen pressure inside the tank rose from 1 MPa to 34.4 MPa, the minimum filling time at which ΔT would remain at 45 °C or lower was found to be approximately 3.6 minutes. Figure 7 shows the changes of pressure and ΔT over time.

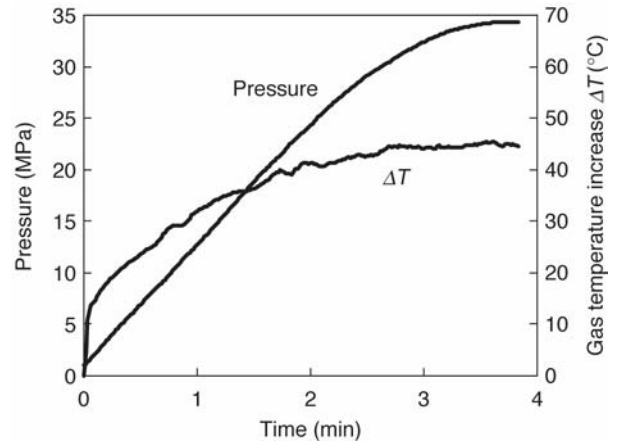


Fig. 7 Filling characteristic from 1 to 34.4 MPa using a Type 3 high-pressure hydrogen tank with a total volume of 156.6 dm³

Pressure inside the tank increased in a straight line with respect to time up to the vicinity of 20 MPa. After that, the rate of pressure increase showed a tendency to gradually slow. The reason that the rate of pressure increase slows in this way is that the critical pressure of 18 MPa with respect to the filling pressure of 34.4 MPa is exceeded. The critical pressure can be derived using the below formula⁽³⁾.

$$P_c = P_i \{2/(\kappa+1)\}^{\kappa/(\kappa-1)} \quad (1)$$

Here, P_c is the critical pressure, P_i is the initial filling pressure, and κ is the specific heat ratio (in the case of hydrogen, 1.409).

The gas temperature inside the tank shows a sharp increase upon initial filling, after which the rate of temperature increase tends to slow over the course of filling time. This is the result of the decrease in compression ratio as filling occurs, the increase in the heat content of the gas itself, and the occurrence of thermal diffusion to the various materials making up the tank.

6. Simulation of Filling Behavior

Newhouse *et al.* conducted natural gas filling tests with different types of tanks and compared their filling efficiency. Their tests used a Type 1 tank of steel, a Type 3 tank with a thin-walled aluminum liner (3.4 mm thick), a Type 4 tank with HDPE liner, and a Type 4 tank with a nylon 11 liner. From these results, these tanks can be ranked by filling efficiency as follows: Type 1 > HDPE Type 4 = Type 3 > nylon Type 4. Tests were also carried out using a Type 3 tank with a thick-walled aluminum liner (5.6 mm thick) and a Type 4 tank with HDPE liner, showing that the Type 3 tank has a higher filling efficiency⁽⁴⁾. This suggests that filling efficiency varies with the thickness of the aluminum liner.

Simulations were then conducted to investigate the influence of the thickness of the aluminum liner on ΔT . When filling with compressed hydrogen, the tank surface radiates heat, so that the perfectly adiabatic compression (in which PV^κ —where P is pressure, V is volume, and κ is the specific heat ratio, which in the case of hydrogen is 1.409—is a constant) does not take place. Moreover, since the temperature inside the tank changes, isothermal change (in which PV is a constant) does not take place. This is, therefore, a polytropic change as expressed by $PV^n = \text{Constant}$. The results in Fig. 7 and the physical properties of the materials in Table 1 were then used in an unsteady heat transfer model to determine the polytropic index n . Accurate approximation was found to be possible when $n = 1.2$. The filling behavior of four types of tanks was then simulated using this value. The four tanks were (1) Type 4 tank with HDPE, (2) Type 3 tank with liner 7 mm thick, (3) Type 3 tank with liner 3 mm thick, and (4) Type 3 tank with liner 1 mm thick. Their filling efficiencies were calculated from those results. The simulation was carried out using the same constant conditions of 10-minute filling time and 35 MPa filling pressure throughout. As shown in Table 2, the results

indicate that ΔT is higher and filling efficiency is lower for thinner aluminum liners. These findings support the test results from Newhouse *et al.*⁽⁴⁾. Contrary to their findings, however, the filling efficiency of Type 3 tanks with liners 3 mm and 1 mm thick was found to be higher than that of the Type 4 tank.

Table 2 Comparison of filling efficiency obtained from the simulation method

	ΔT (°C)	Filling efficiency(%)
Type 4	54	86.8
Type 3 (Liner thickness : 7 mm)	40	89.9
Type 3 (Liner thickness : 3 mm)	46	88.6
Type 3 (Liner thickness : 1 mm)	51	87.5

Figure 8 shows the distribution in the hydrogen gas, the various tank parts, and the outside air of thermal energy generated by polytropic compression when the temperature of the hydrogen gas is at its highest. In the case of the Type 3 tanks, it became clear that the heat content of the liners is lower and the temperature of the hydrogen gas is higher when the liners are thinner.

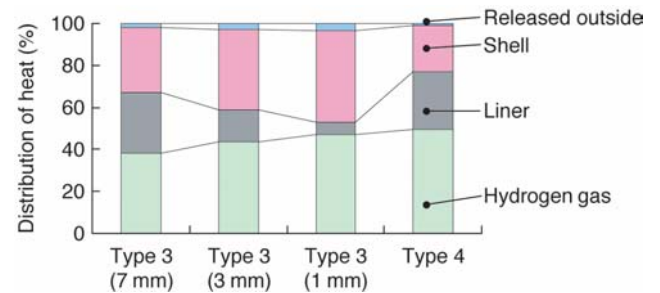


Fig. 8 Distribution of heat generated by the rapid filling of compressed hydrogen gas for various types of tanks

Table 1 Mechanical properties of high-pressure hydrogen tank components

	Aluminum	HDPE	CFRP*
Density (kg m ⁻³)	2680	955	1700
Specific heat (kJ kg ⁻¹ K ⁻¹)	0.896	2.1	0.9
Heat conductive coefficient (W m ⁻¹ K ⁻¹)	180	0.41	6.5
Heat diffusion coefficient (mm ² s ⁻¹)	74.96	0.2	4.25

*)Carbon Fiber Reinforced Plastic

The conspicuous differences in filling behavior that can be recognized even though the heat generated during filling is the same in Type 3 and Type 4 tanks is inferred to be caused by the physical properties of the liner materials. As shown in Table 1, the heat diffusion coefficient α of aluminum is 375 times that of HDPE, and a greater proportion of the heat generated during filling is thought to be distributed to the liner and carbon fibers. It is presumed that the percentage distribution of thermal energy to the hydrogen gas itself is reduced as a result, so that ΔT during filling is lower in Type 3 than in Type 4 tanks.

$$\alpha = \lambda / (\rho \cdot C_p) \quad (2)$$

Here α is the heat diffusion coefficient, λ is the thermal conductivity coefficient, ρ is the density, and C_p is the specific heat at constant pressure.

7. Comparison with Rapid Filling of Natural Gas (methane)

During rapid filling with methane, the temperature drops conspicuously in the vicinity of the methane inlet in the in-tank solenoid valve upon initial filling. The temperature of the gas in the center of the tank, however, does not drop as much as at the inlet. Figure 9 shows one example of that filling behavior. This is the result of Joule-Thomson expansion (isoenthalpic change) that occurs when high-pressure methane flows into the container from the pressure accumulator at the filling station. After approximately 0.1 minutes of filling time, the temperature of the inflowing gas then tends to rise. This suggests that the temperature drop caused by Joule-Thomson expansion (endothermic heat) is exceeded by the temperature rise (exothermic heat) resulting from the energy generated by compression of methane. Thus the behavior of the gas temperature during rapid filling with high-pressure methane differs significantly from the case of hydrogen, and this is caused by the difference in the amount of temperature change undergone by the two gases in connection with Joule-Thomson expansion.

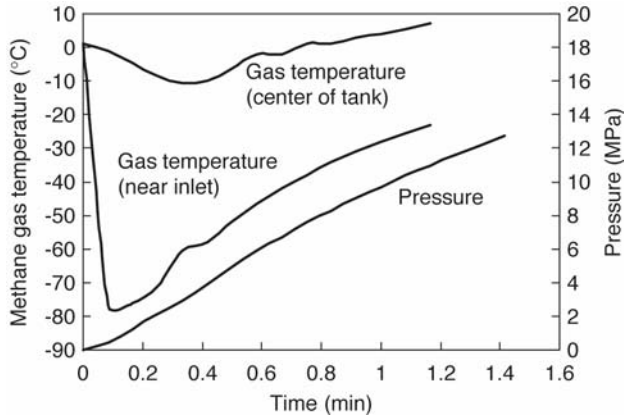


Fig. 9 Changes in gas temperature at the center and near inlet of the tank, and pressure with methane gas filling time

Joule-Thomson expansion is the phenomenon whereby a temperature change occurs in a certain volume of gas under high pressure in a constant flow because the gas passes through a valve or orifice. There are cases when the temperature will rise because of the Joule-Thomson expansion and cases when it will drop, and the boundary temperature between those two cases is known as the inversion point. The inversion point varies with the kind of gas and its pressure. In the case of air at around normal pressure, for example, it is 487 °C, and for hydrogen -72 °C. Above this temperature, passage through a valve or orifice causes the temperature to rise, and below this temperature, it will drop. The drop in temperature results from a decrease in internal energy to the extent of the work done against the intermolecular force. In the case of methane, for example, according to the temperature-entropy curve⁽⁵⁾, when methane at 27 °C and 20 MPa undergoes isoenthalpic change to 1 MPa, then a temperature drop of approximately 80 °C will occur. In the case of hydrogen, however, according to the temperature-entropy curve⁽⁵⁾, hydrogen in the same pressure region undergoes almost no temperature change even if Joule-Thomson expansion occurs. As a result, the kind of temperature drop observed in the initial stage of filling with methane does not occur, and the gas temperature inside the tank rises over time.

8. Investigation of Hydrogen Filling Mass

Under constant temperature and pressure, hydrogen behaves nearly as an ideal gas, but its behavior diverges from the ideal under lower temperatures or higher pressures. This is caused by intermolecular interaction, whereby the intermolecular force distance becomes shorter under higher pressure, so that the repulsive force increases. As a result, for an actual gas V and ideal gas V_{id} at the same temperature and pressure, the volume ratio $Z = V/V_{id}$ will be greater than 1. Z is known as the compression coefficient, and it describes the deviation of an actual gas from an ideal gas.

Figure 10 shows the relationship between pressure and density of hydrogen at 15 °C in ideal and actual states. At pressures of approximately 10 MPa or above, the density of the actual gas begins to diverge from that of the ideal gas, showing a lower value than the ideal gas. Figure 11 shows the changes of pressure in hydrogen at 34.4 MPa and 15 °C when the temperature is changed. The pressure-temperature property of hydrogen gas in the actual state demonstrates behavior virtually identical to that in the ideal state.

Based on the relationship of Fig. 10 and Fig. 11, the isothermal hydrogen filling mass at an ending pressure of 35 MPa and the rating temperature of 15 °C, and the actual hydrogen filling mass with the filling behavior of Fig. 7 were derived as 3.75 kg and 3.32 kg, respectively. Therefore, the filling efficiency is 88.5%.

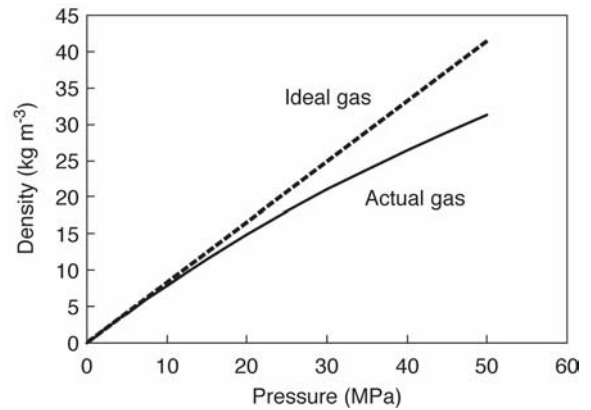


Fig. 10 Relationship between pressure and density of hydrogen gas in ideal and actual states

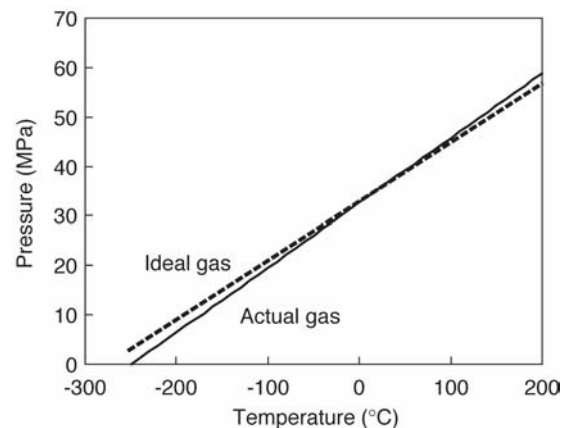


Fig. 11 Pressure-temperature property of hydrogen gas in ideal and actual states

9. Summary

A compressed hydrogen storage system was developed for FCV use and the below findings were obtained. The findings are being utilized in the Honda FCX, which represents the world's first practical application of this technology.

- (1) Rapid filling tests of Type 3 and Type 4 tanks with 34.4 MPa compressed hydrogen showed a conspicuous difference in ΔT during filling, and demonstrated that ΔT is smaller for the former type. This is due to the fact that the heat diffusion coefficient of aluminum is 375 times greater than that of HDPE. Based on these results, the Type 3 tank was adopted.
- (2) Higher residual hydrogen pressure inside the tank means greater heat content, so that ΔT will be lower. Higher ambient temperature means higher maximum temperature of the gas inside the tank, but ΔT remains constant.
- (3) When using a Type 3 compressed hydrogen tank with a total volume of 156.6 dm³ and filling it from an initial hydrogen pressure of 1 MPa to 34.4 MPa, the minimum time in which the gas temperature increase ΔT would reach 45 °C or less from the initial temperature was 3.6 minutes. This is more or less equal to the refueling time of a gasoline vehicle.

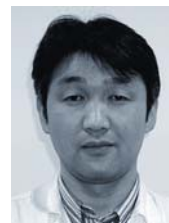
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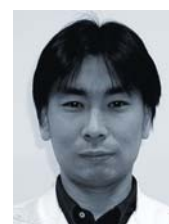
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