The effect of defueling rate on the temperature evolution of on-board hydrogen tanks

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ABSTRACT

During the driving of a fuel cell car, the expansion of the hydrogen along the emptying of the high pressure storage tank produces a cooling of the gas. The hydrogen vessel can experience a fast depressurization during acceleration or under an emergency release. This can result on the one hand in exceeding the low safety temperature limit of ~40 °C inside the on-board compressed hydrogen tank and on the other hand in the cooling of its walls. In the present paper, defueling experiments of two different types of on-board hydrogen tanks (Type III and Type IV) have been performed in all the range of expected defueling rates. The lowest temperatures have been found on the bottom part of the Type IV tank in very fast defuelings. For average driving conditions, in both types of vessels, the inside gas temperature gets closer to that of the walls and the tank would arrive to the refuelling station at a temperature significantly lower than the ambient temperature.

INTRODUCTION

Following the declarations of the EU and G8 leaders, carbon dioxide emissions must be reduced by 80 per cent (from 1990 levels) by 2050. The targets are to stabilize the atmospheric carbon dioxide at 450 parts per million and to keep the increase in global temperature below 2 °C relative to its pre-industrial level [1]. To achieve these goals, the road transport sector would require a decarbonisation in up to a 95% [2].

Fuel cell electric vehicles (FCEV) provide the benefits of electric vehicles combined with the functionality of a combustion engine car. FCEV can be refuelled in 3–5 min using a fuel hose similar to the one used in a conventional fuel station. Moreover, they have autonomy for hundreds of kilometres before they need refuelling. Expected vehicle range per full fuelling is taken to be greater than or equal to 500 km (300 miles). The user convenience and the zero emission of FCEVs make them a clean alternative for personal transportation [3].

Consequently, many car manufacturers have stated their intention to commercialize fuel cell powered vehicles in the 2015/2020 timescale. In 2009 for example, seven of the largest car manufacturers in the world – Daimler, Ford, General Motors, Honda, Hyundai-Kia, Renault–Nissan and Toyota – signed a letter of understanding addressed to the oil and energy industries and government organizations. The letter indicated their intent to commercialize a significant number of FCEVs from 2015 [4]. Nowadays, some of these vehicles are already on the market: the Toyota Mirai [5] and the Hyundai ix35 [6].

Hydrogen is an energy dense fuel by mass, higher than conventional fuels. However, volumetric energy densities are much lower. At present, the most common method of storing hydrogen on-board in land vehicles is as a compressed gas,
either at 35 MPa or, more commonly, at 70 MPa. The hydrogen storage system contains all components that form the primary high pressure periphery for containment of stored hydrogen. The key functions of the hydrogen storage system are to receive hydrogen during fuelling, store the hydrogen until needed and then discharge the hydrogen to the fuel cell system for use in powering the vehicle. Constituents of a typical compressed hydrogen storage system include the high pressure hydrogen storage tank and all other components as the check valve, the shut-off valve and the thermally-activated pressure relief device (TPRD) [7].

Most high pressure hydrogen storage tanks used in fuel cell vehicles consist of two layers: an inner liner that prevents gas leakage/permeation and an outer layer that provides structural integrity. The liner is made of metal in a Type III tank and of plastic polymer in a Type IV. The outer layer is usually made of resin impregnated fibre reinforced composite which is wrapped over the liner. The on-board hydrogen storage system must fulfil specific performance based and technical design requirements [7–9] which are collected in regulations and standards for hydrogen powered motor vehicles.

The storage system must be able to supply a sufficient mass flow rate of hydrogen to the fuel cell system to meet the required power demand at acceptable pressures and temperatures under all driving conditions. The average fuel consumption of a hydrogen powered midsize crossover Sport Utility Vehicle (SUV) [10] can go from about 0.1 to 0.2 g/s for a city drive (30 km/h and 50 km/h; corresponding to about 10 kW fuel cell power) to 0.4–0.6 g/s for a highway drive (at 100 km/h and 120 km/h and about 30 kW fuel cell power). However, peaks of hydrogen consumption can be expected during acceleration; e.g. 0.7–0.8 g/s for an easy acceleration and 1.5–1.8 g/s for full-throttle acceleration. Moreover, the tank might also experience a very fast depressurization in an emergency hydrogen release from a TPRD which is designed to vent the entire contents of the vessel rapidly in a few seconds [7].

The transfer of hydrogen, which covers the filling and the emptying of hydrogen storage vessels, implies compression and expansion of the gas, which produces temperature variations. During the fast refuelling, the increase of the internal energy of the gas inside the tank (consequence of the work done to compress the gas) produces a temperature increase. Similarly, during the driving, the decrease of the internal energy of the gas produces its cooling. The hydrogen mass inside the tank is determined by the pressure and temperature. In order to take both parameters into account, the State of Charge (SOC) of a compressed hydrogen tank, given by Equation (1), has been defined as the ratio between the density of the gas at a given temperature and pressure and the full tank density (at 15 °C and the Nominal Working Pressure, NWP, which for our case is 70 MPa) [11].

\[
\text{SOC} = \frac{\rho_{\text{H}_2}(P, T)}{\rho_{\text{H}_2}(\text{NWP}, 15 \degree C)} \times 100.
\] (1)

Standards and regulations for on-board compressed hydrogen tanks have established that safe operational conditions, including filling and defuelling, must respect the temperature limit between −40 °C and +85 °C [7–9]. The SAE J2601 standard [11] establishes the protocol for hydrogen fuelling of light duty vehicles. For non-communication case, the temperature of the vehicle storage system at the onset of fuelling is not available to the dispenser. This temperature is normally assumed to be equal to the ambient temperature. However, the storage system can be warmer or colder than ambient temperature at the start of refuelling. The reasons could be several; e.g. the vehicle parking location, the position of the storage system on the vehicle, the driving distance and speed to the refuelling point. In the last version of the standard, industry-wide consensus has been reached on the definition of “soak” as the temperature deviation from ambient of the vehicle storage system and “Hot Soak” and “Cold Soak” zones are specified as a function of the ambient temperature.

The GasTeF facility is a reference laboratory of the European Commission Joint Research Centre where pre-normative research on full-scale high pressure hydrogen tanks is performed in support to European Union policies [12]. In the last years, many experimental campaigns have been carried out in GasTeF to analyse on-board hydrogen tanks behaviour during refuelling. A description of the facility and some of the last obtained results can be found in Refs. [13–16]. In addition, the experimental results are complemented with computed fluid dynamics analysis of the phenomena taking place in the tank with a model developed at JRC by means of an ANSYS® CFX [17,18].

Although the refuelling of hydrogen is already a studied hydrogen transfer process, there are very few published data related to the behaviour of on-board hydrogen tanks during defuelling. In the work presented in this article, a series of discharge experiments of commercial hydrogen storage tanks (one Type III and one Type IV) have been performed in GasTeF. The chosen defuelling rates cover all the expected driving conditions; from a steady state city drive to the vehicle’s maximum fuel-demand rate. Temperatures of the gas inside the tank have been monitored to study their spatial distribution. The temperatures at the outer surface have also been measured to study the temperature evolution through the walls and to determine its relation to the inside gas temperature.

**Experimental conditions**

In Table 1, the characteristics of the two 70 MPa nominal working pressure on-board hydrogen storage tanks used in this study are given. One is a Type IV with 29 L capacity and the other is a Type III with 40 L capacity. Similarly to our previous experimental studies [13–16], each tank has been instrumented with 8 thermocouples (TC) and with four resistance temperature detectors (RTD). The pressure has been measured using a pressure transducer (PT) placed at the rear. In all cases, a 3 mm diameter hydrogen injector has been used. As depicted in Fig. 1, the TCs (labelled TCI to TC8) measure the temperature of the gas at different positions. The thermocouples have been placed inside the tank by means of a tree-shape array introduced through the rear. The RTDs (labelled TTop, TBottom, TFront and TRear) have been placed on the tank wall to measure the temperature of the external surface and of the bosses. The positions selected to compare the evolution of the inside gas temperature with the evolution
of the temperature at the external surface of the tank on top and bottom are highlighted with an oval in Fig. 1.

Both Type III and Type IV tanks were filled to 77 MPa. The defueling started once the full tank temperature was equilibrated with the surrounding environment at 25°C. The criterion for thermal equilibrium was a difference of ±2°C to the targeted value in all thermocouples and RTDs. During this thermal equilibration, the cooling of the gas inside the tank results also in a drop of pressure reaching values below 70 MPa. As a result, the State of Charge (SOC) at the beginning of the discharge ranged between 90% and 100% in the different experiments performed.

The different discharge speeds were controlled with the compressor which empties the tank by placing back the hydrogen to the hydrogen reservoir of the facility. The compressor empties the tank to 5 MPa at a constant pressure ramp rate. When the pressure inside the tank is below 5 MPa, the compressor is not anymore able to reduce the same amount of hydrogen at the same speed and the discharge rate lowers down significantly. In the performed tests, the tanks were emptied down to 2 MPa. In Fig. 2, the measured pressure profile and the calculated SOC and Mass Flow Rate (MFR) profiles during a fast discharge (in less than 20 min) of the Type III tank are shown. For the comparison of the internal and external temperatures and in order to properly identify the effect of different discharge rates, only the first part of the discharge, down to 20% SOC (pointed on Fig. 2 with dotted line) has been considered. For this first linear part, the Average Mass Flow Rate (AMFR) has been calculated considering the total time required for reaching the final mass. The values of the mass flow rate (MFR) and SOC given in this paper have been calculated with the measured temperature and pressure values and using the Redlich-Kwong equation of state for real gases which is used to predict accurately hydrogen properties in a wide range of temperature and pressures [19]. It has to be mentioned that 20% SOC is not foreseen to be largely surpassed during the driving of a hydrogen car; in some of the FCEVs already in the market at 10 MPa (which at 15°C temperature corresponds to a 20% SOC) the indicator of refuelling is lighted on [6].

### Results and discussion

#### Effect of the type of tank

Fig. 3 shows the evolution of the internal and external temperatures during a defueling of Type III (a) and Type IV (b)
tanks. In both cases, the emptying has been performed at a high AMFR value of 1.8 g/s. The mass flow rate is the same in both cases but due to the smaller volume of the Type IV tank, the discharge takes place in shorter time. As it has already been mentioned, during the discharge, the gas expansion produces a cooling of the gas inside the tank. Moreover, due to the buoyancy effect, warmer gas moves upwards resulting in a vertical gas temperature gradient [14]. Along the emptying, heat exchange occurs between the warmer environment and the colder gas through the walls of the tank. As a consequence, the external surface temperature is considerably reduced during the emptying. On top of that, there exists a temperature difference between the top and the bottom tank external surfaces due to the vertical temperature gradient of the gas. The effect of this heat exchange is especially visible towards the end of the discharge, when the MFR becomes smaller and the gas mass in the tank is considerably reduced. Under these circumstances an increase of the gas temperature is experienced (in Fig. 3 this effect is visible after approximately minute 12 in Type III tank and after minute 8 in Type IV).

Comparing the two types of tanks, Fig. 3 shows that, for similar discharge rates, the gas reaches lower temperatures in the Type IV than in the Type III. This behaviour is related to the different thermal properties of the materials of each tank. The thermal diffusivity of the aluminium alloy liner on Type III tank is much higher to that of the polyethylene on Type IV [14,20]. The effect of the higher thermal diffusivity of the Type III tank is also visible on its external surface which experiences a bigger temperature decrease than in Type IV.

Effect of the discharge rate

The evolution of the temperature of the gas and of the external surface on top (TC3 and TTop) and bottom (TC1 and TBottom) of the tanks (as shown in Fig. 1) has been compared for different discharge rates. In Fig. 4, the evolution of these temperatures in the Type IV tank has been plotted against the SOC (%) for two extreme mass flow rate values (1.8 g/s and 0.2 g/s). In the fast emptying (Fig. 4a), the temperature decrease of the gas is very big, with final values in the range from −18 °C to −55 °C, while the external surface experiences only a very small decrease of approximately 5 °C. The slow emptying shown in Fig. 4b occurs in longer time which allows for a higher heat exchange. The external surface temperatures experience a drop of more than 15 °C in this case. The faster the discharge, the bigger is the vertical temperature gradient inside the tank (higher is the difference between TC3 and TC1). This effect has been observed in both tank types.

The decrease of the local gas temperatures (ΔTgas = Tgas_Full − Tgas_20%SOC) has been considered for all the defueling experiments performed, as well as the decrease of the tank surface temperatures (ΔTsurface = Tsurface_Full − Tsurface_20%SOC) for the top and bottom positions. Fig. 5 shows the results for a Type III tank while the results for a Type IV can be found in Fig. 6. As the discharge rate increases, with less time for heat exchange from the warmer environment towards the gas, ΔTgas increases while the ΔTsurface decreases in absolute values. The minimal temperature limit of −40 °C established by standards and regulations [7–9] is largely exceeded on the bottom for discharges with AMFR larger than 0.8 g/s. It must be noted that this minimal temperature limit applies to the materials of the tank, to avoid ageing or damage effects from too low temperatures, and not to the gas temperatures. Considering that the external surface of the tank experiences only a maximal decrease of 20 °C, it can be concluded that only a limited part of the wall thickness (the gas phase in contact with the internal surface of the liner) is affected by extreme low temperature. However, the measurement of local temperatures in the liner and at the liner-composite interface and along the thickness of the composite wrapping would be important to gain deeper insight in the thermal behaviour of the tanks.

From the results above, it cannot be excluded that a liner of a type IV tank does not experience a temperature lower than −40 °C under extreme emptying conditions. To avoid this occurring, tank manufacturers require that the storage

![Fig. 3 – Evolution of the gas and the external surface temperatures during a discharge at an AMFR of 1.8 g/s of a Type III 40 L tank (a) and a Type IV 29 L tank (b).](image-url)
The system is designed in such a way that defueling in less than 1.5 h during driving does not occur. Also, in case of the more rapid defueling before regular maintenance, the emptying procedure prescribes a methodology aimed at avoiding the attainment of temperatures below 40 °C. Therefore, extreme defueling rates are not expected during the operative life of a storage system. Nevertheless, temperatures even lower than those measured in this study might be expected during emergency releases triggered by the automatic opening of the thermally activated pressure release device [7].

![Fig. 4](image)

**Fig. 4** – Evolution of the gas and external surface temperatures on top and bottom during the discharge of the Type IV tank (from Full tank down to 20% SOC) at mass flow rates of 1.8 g/s (a) and 0.2 g/s (b).

![Fig. 5](image)

**Fig. 5** – Temperature decreases of the gas (a) and external surface (b) on top and bottom of the Type III tank for different discharge rates from Full tank down to 20% SOC.

![Fig. 6](image)

**Fig. 6** – Temperature decrease of the gas (a) and external surface (b) on top and bottom of the Type IV tank for different discharge rates from full tank down to 20% SOC.
Comparison of gas and surface temperature evolution

To establish a possible empirical relation between gas and tank surface temperatures, the data presented so far has been further elaborated in Fig. 7 where the ratio $\Delta T_{\text{gas}}/\Delta T_{\text{surface}}$ has been calculated. In both tank types, this ratio has been found similar for top and bottom position of the tank surface. In Fig. 7 the values of this ratio have been plotted against the average mass flow rate. For Type III tank, the evolution has been found to be linear with the discharge rate with a regression coefficient $R^2$ greater than 0.99. The high heat transfer coefficient of the metal liner and the good contact of the liner with the composite wrapping can be a reason for this behaviour. For Type IV tanks, a second order polynomial regression was needed to attain a value of $R^2$ greater than 0.99. The equations fitted to experimental data measured on top of the tanks are also shown in Fig. 7.

The slow discharge rates investigated in this study correspond to the expected average driving conditions of a fuel cell car. These ‘realistic’ values (highlighted by the oval in Fig. 7), are characterized by a value of the $\Delta T_{\text{gas}}/\Delta T_{\text{surface}}$ ratio of approximately 2. This means that for defueling rates in the range of 0.2–0.4 g/s, the decrease of the temperature at the surface of the tank is indicatively the half of the decrease of the internal gas temperature. Therefore, in the case of a continuous emptying from 100 to 90% SOC down to about 20% SOC, the car would arrive at the refuelling station with its internal gas temperature. Consequently, in the case of a continuous emptying from 100 to 90% SOC down to about 20% SOC, the car would arrive at the refuelling station with its hydrogen storage system at a temperature of at least 20 °C lower than the ambient temperature. This initial tank temperature will affect the final temperature, and consequently the final SOC, reached inside the storage system during the refuelling.

Under extreme acceleration conditions or under an emergency release triggered by the pressure release device, very low temperatures might be reached inside the tank. In some cases, the design temperature limit of $-40$ °C might be exceeded, while the external surface of the tank does not see almost any change of temperature. However, lower temperatures are expected at the interface gas-liner, at the interface liner-composite and along the thickness of the composite wrapping.

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References


