

HYDROGEN TANK FILLING EXPERIMENTS AT THE JRC-IE GASTEUF FACILITY

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ABSTRACT

Storage of gases under pressure, including hydrogen, is a well-known technique. However the use in vehicles of hydrogen at pressures much higher than those applicable in natural gas cars still requires safety and performance studies with respect to the verification of the existing standards and regulations. The JRC-IE has developed a facility, GasTeF, for carrying out tests on full-scale high pressure vehicle's tanks for hydrogen or natural gas. Typical tests performed in GasTeF are static permeation measurements of the storage system and hydrogen cycling, in which tanks are fast filled and slowly emptied using hydrogen pressurised up to 70 MPa, for at least 1000 times according to the requirements of the EU regulation on type-approval of hydrogen-powered motor vehicles. Moreover the temperature evolution of the gas inside and outside the tank is monitored using an ad-hoc designed thermocouples array system. This paper reports the first experimental results on the temperature distribution during hydrogen cycling tests.

INTRODUCTION

The storage of gases under pressure, including hydrogen, is a well-known technology. Nonetheless, the use of hydrogen tanks in vehicles and in particular the challenge of using very high pressures, demands stringent regulations supported by safety and performance studies. Requirements to qualify storage systems for on-road passenger vehicles have been under development primarily by the SAE International and ISO committees (respectively SAE-J2579 [1] and ISO15869 [2]). Recently, European Regulations have been published and implemented [3, 4] and at present there is a common effort under the auspices of the UN-ECE for the establishment of a Global Technical Regulation for Hydrogen Fuelled Vehicles (GTR HFV) which comprises the requirements of SAE and ISO [5]. The push to develop safety standards for hydrogen storage well before commercialization is driven by two factors. First, by acknowledging that on-road safety is of highest priority, stringent requirements are being developed a-priori, without waiting for any lessons learned from on-road experience. Second, taking into account that this sector is exposed to intense commercial competition, innovation cycles are short and new technologies are introduced regularly into the marketplace. These short cycles required innovation friendly storage requirements in the regulations to support a quick qualification process for novel technologies featuring reliable and durable performance under broad conditions of use. Setting up qualification test campaigns with conditions that take into account historical failure mechanisms is helpful but not sufficient. Instead a comprehensive approach to define extreme conditions of on-road vehicle service is required. Storage systems must then function both under the stresses of normal vehicle operation and under externally imposed stresses.

Today's state of the art for hydrogen storage comprises 35 MPa (350 bar) and 70 MPa (700 bar) compressed gas tanks. Carbon fiber fully wrapped-reinforced tanks are already in use in prototype hydrogen-powered vehicles. Two types of inner liners are typically used: metal (aluminium, Cr-alloys) in "Type 3" storage pressure vessels and high molecular weight polymer in "Type 4" tanks, as described in ISO 158692. The application of such materials comes from the need of guaranteeing impermeability of the inner liner to the hydrogen molecules whilst having the tank being as lightweight as possible. These pressure vessels must be able to withstand a temperature range between -40 °C and 85 °C in normal conditions including filling and discharging and these temperature specifications are also applied to the tank materials.

The overall objective for the fill process of these high pressure tanks calls for reasonably short fill duration, while at the same time exceedingly high temperatures in the tank must be avoided [6]. It is these two factors that drive the engineering of the tank filling process. Shortening the fill duration will lead to higher temperatures inside the tank - due to the near-adiabatic compression of the gas. In aiming to ensure a complete fill of the tank higher gas temperatures during filling have to be compensated with a higher filling end pressure (pressure overshoot). Bearing all these effects in mind, three major risks can be identified for the fast filling process:

- Over-pressure when the tank is exposed to a pressure higher than the permitted one.
- Over heating when the tank is exposed to a temperature higher than the maximum allowed value.
- Over filling which can occur in cold countries when the ambient temperature is low. In that case the pressure overshoot usually used for fast filling can lead to high storage pressure but low filling temperature. If the tank is subsequently exposed to higher temperature during the day the pressure may reach unexpected values.

The JRC-IE facility GasTeF is an EU reference laboratory that has been designed to carry out performance verification tests of full-scale high pressure vehicle tanks for hydrogen or natural gas or of any other high-pressure components, such as valves and pipes. In particular the following experiments on both Type 3 and Type 4 tanks are performed:

- Fast-filling cycling, in which storage tanks are fast filled and slowly emptied using hydrogen pressurized up to 70 MPa, in the range of 500 to 5000 times to simulate their lifetime in a road vehicle. During the cycling process the tank is monitored for leaks and permeation rates using gas chromatography.
- Static permeation measurements as a function of time on tanks filled to 70 MPa and temperatures to 85 °C.
- Temperature profile measurements inside the tanks to validate computational fluid dynamic (CFD) simulations of the temperature distribution when filling and emptying at various rates.

These tests are part of a more general effort aimed at assessing from a safety point of view existing and future regulation in the field, and in particular the measures in the implementing European regulation on type-approval of hydrogen vehicles [4].

This paper describes the facility in general and specifically the experimental set-up developed for the direct measurements of the temperature evolution inside the tank during the filling phase. Part of the paper is the presentation of the results of the first experiments, which were performed to test the functionality of the facility and to commission it. These first results allow a qualitative comparison with already published results and the definition of requirements for the future experimental campaigns.

GASTE F DESCRIPTION

GasTeF consists of a half-buried concrete bunker with an attached gas storage area in open air. The Fig. 1 shows a schematic of the facility. The utilized gases (helium, hydrogen, methane, etc) are stored in standard bundles of bottles at a pressure of around 28 MPa. From here the gas enters a two-way piston compressor, which is able to reach hydrogen pressures up to almost 90 MPa. The filling consists of two phases: in the first, the compressor is bypassed and the tank is filled directly from the gas storage. In the second phase, the compressor takes over and raises the pressure in the tank to the required value. The same two phases occur when emptying of the tank, then the gas flows in the other direction to re-fill the storage bottles. This results generally in a non-linear filling profile. The fill rate given in this paper represents the average value calculated considering the total time required for reaching the final pressure.

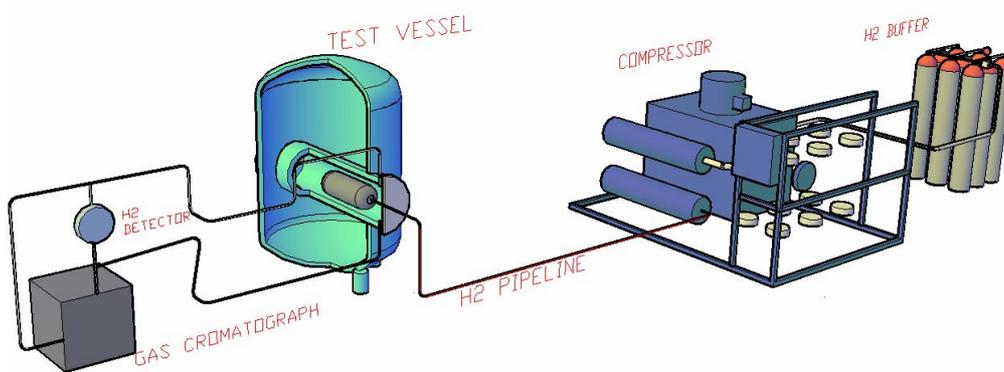


Figure 1. Schematic of GasTeF facility

The fuel tanks are horizontally placed inside a sleeve, which contains an inert gas (helium or nitrogen) and serves as a chamber for determining hydrogen permeation through the test tank walls, see Fig. 2. The hydrogen concentration in the sleeve is measured with a gas chromatograph and two hydrogen detectors, one for low concentrations up to 2000 ppm and the other for concentrations up to 10 %.

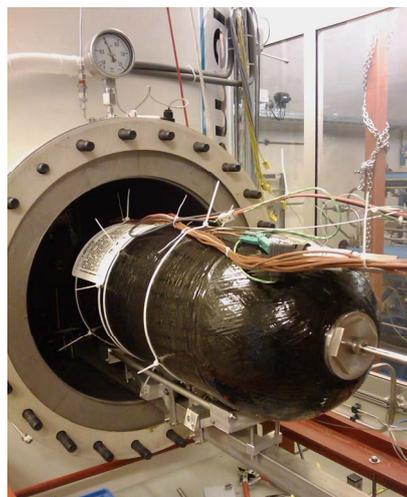


Figure 2. Instrumented tank being inserted in the sleeve

Prior to and during tests with hydrogen, the bunker is closed and the internal oxygen concentration is reduced to less than 1% by purging with nitrogen gas to prevent any occurrence of ignition created in the event of hydrogen leakage. As the purging procedure requires several hours, a sequence of tests with helium is made before the bunker is closed to identify and mitigate any flaws with the experimental set up. As such, the tightness of the connections, the operation of the thermocouples and pressure transducers and the behavior of the tank under high pressures are checked. In normal operation the facility runs fully automatically and the tests are operator supervised from a control room situated in an adjacent building. A more detailed description of the GasTeF facility and its equipment can be found in [7].

EXPERIMENTAL RESULTS

The fast filling experiments presented in this paper have been performed using a 29 liter type 4 tank, with a plastic liner and a wrapping of carbon and glass fiber reinforced epoxy composite with a length of 828 mm and 235 mm diameter. The nominal working pressure is 70 MPa.

The tank has been instrumented with twelve thermocouples arranged as depicted in Fig. 3. Four thermocouples are located at the outer surface and the other eight are placed inside the tank. The thermocouples 7 and 8 are inserted through the gas inlet opening. The other thermocouples are positioned inside the tank by means of a special array designed at JRC-IE, see Fig. 4. The thermocouple array is introduced at the bottom of the tank and can be axially displaced so that the distance of the thermocouples to the gas inlet can be adjusted. The thermocouple number 5 acts as reference for all temperature measurements and remains therefore in a fixed position even if the array's location is adjusted. Outside temperatures are measured by other thermocouples of which the one at the top outer surface of the tank, T Top, is usually placed adjacent to the reference thermocouple 5. All thermocouples have 1 mm diameter and are of type K, capable of measuring in a range of -200 to 1250 °C with an uncertainty of 2.2 °C.

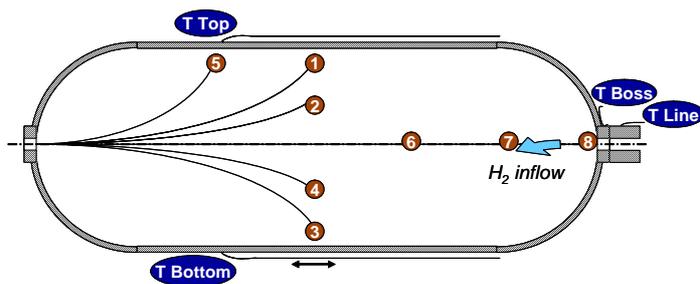


Figure 3. Position of the thermocouples

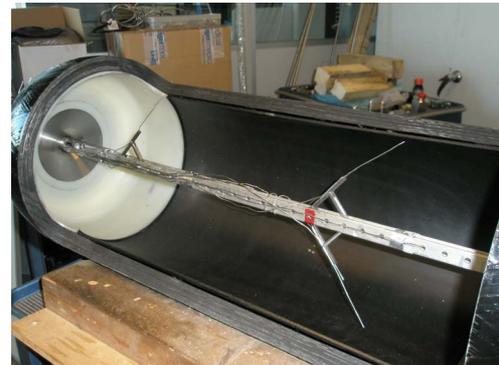


Figure 4. GasTeF thermocouple array

The gas pressures at the inlet/outlet and inside the tank are measured using pressure transducers. To avoid excessive accuracy errors due to the big pressure range, the utilized transducers are split in pressure classes: one is calibrated for the low, the other one for the high end of the pressure range. For the high range pressure transducer (0 to 70 MPa) the error is over 5 % for pressures lower than 10 MPa and only 0.64 % at 70 MPa whereas for the transducer calibrated for low pressures, 0 – 2.5 MPa, the error is 0.4 %. The time interval for pressure and temperature data logging is 0.6 seconds.

Tests have been carried out with the thermocouple array being placed at three different axial positions given by the distance of the thermocouples 1, 2, 3, 4 and 6 from the gas inlet. Within this paper, experiments have been performed in three different positions named A, B and C. For example, the thermocouple 1 was located at 200, 340 and 433 mm from the inlet; these three distances represent the A, B and C cases. As said above, the thermocouple 5 is used as reference and stays approximately at the same position.

Tables 1 to 3 indicate the position of all the thermocouples in the A to C cases. For each position at least three fast filling cycles on different days have been made with the aim of using the results as input/validation for the CFD modeling of the fast filling performed by C. Galassi et al. [8]. Fig. 5 gives an example of one hydrogen cycle performed with the thermocouples in position B. Long-term temperature evolution measurements have been carried out after the cycling sequences in positions B and C mainly to check the time required to stabilize the pressure and temperature of the gas inside the tank. Both fast filling and long term temperature evolution tests has been performed with the sleeve at room temperature.

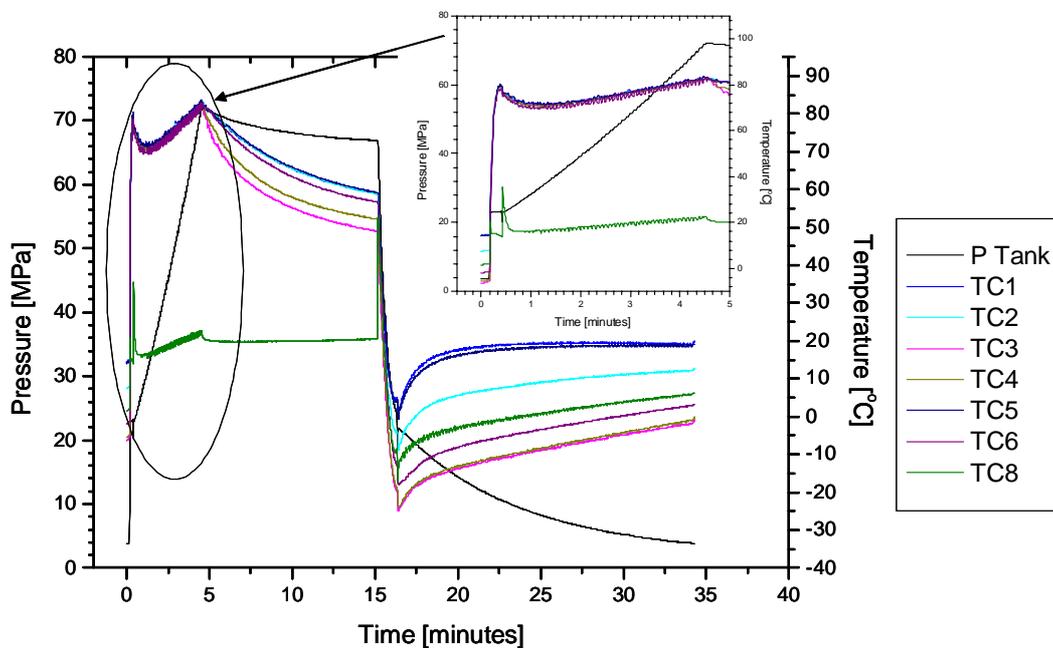


Figure 5. Hydrogen fast filling cycle

The results of the fast filling tests are shown in the Table 1, Table 2 and Table 3 for both helium and hydrogen. As it can be seen in there, several starting and end pressures (P_i and P_f values are relative) have been considered and different fill rates were achieved. T_i and T_f relate to the initial and the final temperatures measured by the reference thermocouple TC-5, while T_{top} tank is the temperature reached at the top outer surface of the tank and T_{boss} is the temperature measured at the tank boss, as shown in the Fig. 3.

Table 1. Results for the thermocouple array in Position A: TC-1 and TC-3 at 200 mm from inlet, TC-2 and TC-4 at 250 mm, TC-6 at 175 from inlet and TC-5 at 500 mm from gas inlet and 15.5 mm from liner

Position A					Internal TC-5		External	
Gas	Pi [MPa]	Pf max [MPa]	Duration [min]	Fill rate [bar/s]	Ti [°C]	Tf [°C]	T top max [°C]	T boss max [°C]
Helium	9.9	69.44	5.14	1.89	33.0	101.7	47.3	40.5
Helium	9.47	69.22	5.17	1.98	30.3	100.4	47.0	35.9
Helium	9.44	69.4	3.42	2.7	29.8	106.5	46.2	34.1
Helium	2.24	69.57	3.43	3.02	11.7	113.9	42.3	33.3
Helium	3.9	69.65	2.51	3.85	28.7	119.1	45.4	29.9
Helium	3.86	69.27	3.94	3.45	29.8	119.1	46.6	29.3
Helium	2.22	69.24	3.12	3.48	22.3	126.3	48.1	31.1
Helium	3.97	69.14	2.56	5.19	31.5	124.0	51.1	34.9
Helium	3.9	69.59	3.13	3.39	34.9	122.0	47.7	39.1
Helium	3.9	69.59	2.10	5.05	35.6	127.4	51.8	35.0
Helium	3.88	71.91	3.16	3.45	36.5	124.9	53.4	35.0
Helium	3.88	71.23	9.99	1.22	34.6	94.9	53.2	42.7
Helium	3.86	72.13	4.33	2.50	32.3	115.1	52.3	38.2
H ₂	3.86	72.1	5.11	2.19	19.2	83.3	45.2	34.7
H ₂	2.3	72	5.15	2.21	18.2	91.8	49.0	38.9
H ₂	2.39	72.25	4.20	2.68	26.9	96.8	51.7	41.8
H ₂	3.87	71.91	3.57	2.87	8.9	81.3	40.9	30.6
H ₂	0.03	72.26	4.65	2.93	20.6	101.3	53.4	43.7
H ₂	0.04	72.01	3.46	3.19	21.2	101.6	49.2	41.6

Table 2. Results for the thermocouple array in Position B: TC-1 at 340 mm from inlet and TC-3 at 343, TC-2 and TC-4 at 364 mm, TC-6 at 314 from inlet and TC-5 at 515 mm from gas inlet and 27.5 mm from liner

Position B					Internal TC-5		External	
Gas	Pi [MPa]	Pf max [MPa]	Duration [min]	Fill rate [bar/s]	Ti [°C]	Tf [°C]	T top max [°C]	T boss max [°C]
Helium	3.96	72.09	5.03	2.27	18.1	101.5	49.6	36.3
Helium	0	72.18	4.39	2.97	20.5	100.1	52.3	37.8
Helium	4	72.05	3.39	3.11	21.8	106.9	54.9	39.8
Helium	3.92	72.24	3.37	3.15	18.3	104.9	52.3	37.6
Helium	3.91	72.18	3.36	3.17	20.7	106.8	53.8	38.7
H ₂	0.03	72.06	5.51	2.05	20.6	93.9	53.3	46.0
H ₂	0.44	72.02	5.17	2.26	22.8	115.8	54.8	45.2
H ₂	2.14	72.21	5.07	2.28	19.8	92.8	51.9	52.1
H ₂	3.89	71.94	4.37	2.46	3.7	77.2	40.9	31.0
H ₂	3.86	72.06	4.33	2.49	13.9	82.9	46.3	35.1
H ₂	3.84	72.12	4.33	2.50	18.9	85.7	49.2	37.7
H ₂	0.02	72.06	4.7	2.92	20.6	99.3	52.6	43.5
H ₂	2.18	71.97	3.70	3.73	24.1	97.5	52.5	40.8
H ₂	2.37	72.31	4.2	2.88	21.1	96.7	51.6	43.2

Table 3. Results for thermocouple array in Position C: TC-1 at 433 mm from inlet, TC-2 at 478 mm, TC-3 at 418 mm, TC-4 at 464 mm and TC-6 at 393 from inlet and TC-5 at 505 mm from inlet and 18 mm from liner

Position C					Internal TC-5		External	
Gas	Pi [MPa]	Pf max [MPa]	Duration [min]	Fill rate [bar/s]	Ti [°C]	Tf [°C]	T top max [°C]	T boss max [°C]
Helium	14.95	16.18	11.32	0.02	4.7	13.4	33.6	16.4
Helium	14.98	69.06	7.48	1.15	7.1	71.7	42.0	24.4
Helium	14.96	69.07	7.46	1.16	5.5	70.3	34.6	24.9
Helium	14.73	69.06	8.29	1.17	27.9	86.4	26.0	36.5
H ₂	14.94	16	10.36	0.02	10.0	16.6	32.5	21.5
H ₂	2.3	29.94	42.6	0.11	14.1	37.5	30.0	22.9
H ₂	2.23	29.72	5.17	0.87	15.5	89.4	38.6	23.2
H ₂	15.14	69.02	7.20	1.28	20.7	73.4	33.2	26.3
H ₂	15	69.2	6.53	1.31	26.5	77.1	42.5	29.9
H ₂	14.93	69.13	6.50	1.32	19.1	68.3	33.3	26.8
H ₂	14.93	69.08	6.50	1.32	15.8	65.2	40.9	28.6
H ₂	14.99	69.13	6.47	1.33	16.0	68.3	37.8	28.3
H ₂	14.98	69.18	6.44	1.34	11.4	64.5	35.6	27.1
H ₂	2.32	29.76	1.39	2.75	20.8	87.0	30.9	25.5
H ₂	0.03	29.89	1.42	2.9	21.4	93.0	34.9	25.8
H ₂	14.97	69.01	1.45	5.13	23.8	86.0	40.6	28.9
H ₂	14.92	69.42	1.42	5.32	19.5	82.5	38.5	28.5
H ₂	15.01	69.44	1.39	5.46	33.9	95.3	44.1	28.9
H ₂	2.38	29.95	0.41	6.63	21.7	98.6	32.2	24.1

DISCUSSION

As can be seen in Fig. 5 following the first sharp pressure increase, the highest temperatures are reached in the upper part of the tank, see TC-1 and TC-5, due to the buoyancy effect. Nonetheless local temperature differences with respect to the average gas value are in the range of 5 to 10 °C. The lowest temperatures are obtained near the inlet, TC-6, and at the inlet, TC-8. In trying to assess the temperature stresses on the tank liner, thermocouple TC-5 has been used for further data analysis.

As it can be noted from the Tables 1 to 3, the results are coherent with the experimental findings reported in [9] and [10]. The main factors affecting the temperature rising during fast filling are the fill rate and the initial pressure. Longer fill times produce lower final gas temperatures, similarly the higher the initial pressure in the tank the lower final gas temperature. As provided in Table 2, the temperature at the external tank surface can reach up to 55 °C for a filling of an empty tank with a 2.26 bar/s (226 kPa/s) average rate.

A comparison between helium and hydrogen with regards to the temperature development during fast filling is made in Fig. 6 and Fig. 7. Final internal temperatures obtained with helium (filled triangles in Fig. 6) are normally higher for the same fill rate than with hydrogen (open triangles in Fig. 6) except in the cases where the fill process starts with an empty tank. On the other hand the gas type does not significantly influence the maximum external temperature reached at the tank surface.

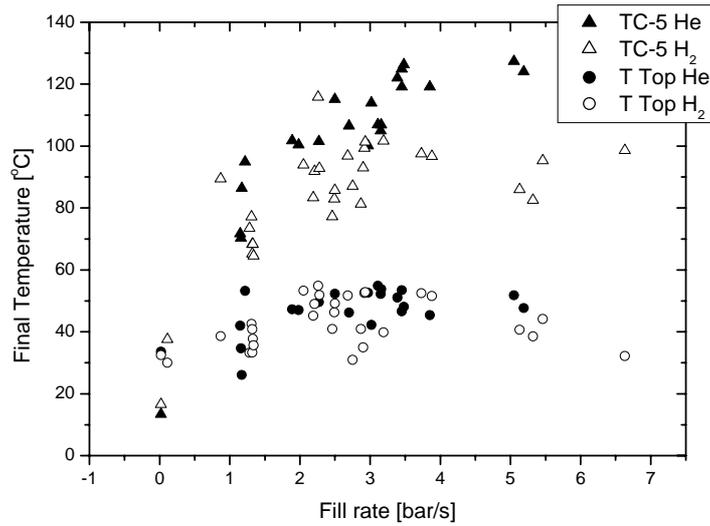


Figure 6. Summary of all the filling experiments performed with helium or hydrogen. The final temperatures of thermocouple 5 and of the thermocouple located on the external tank surface (“T Top”) are given as function of the average fill rate. Data correspond to different initial and final tank pressures

Fig. 7 shows the temperature increase achieved in the reference position (see the position of TC-5 in the Fig. 3) as a function of the fill rate. For low fill rates both helium and hydrogen are comparable. At higher fill rates the 19.3% higher thermal conductivity of hydrogen becomes important and leads to a separation of the trend curves and lower valued at the hydrogen side.

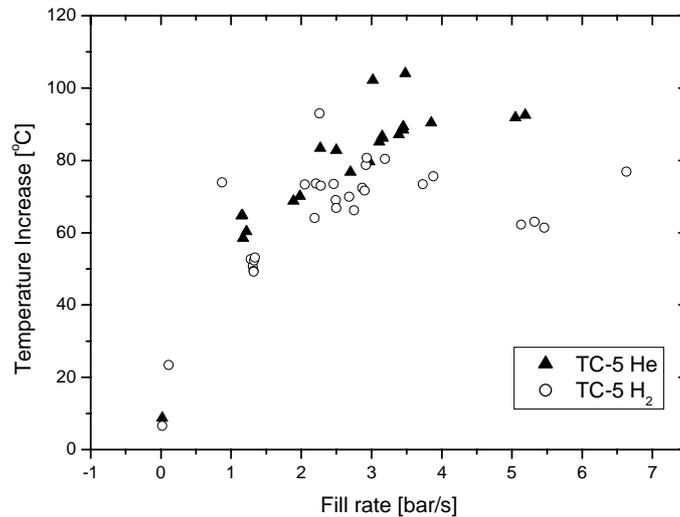


Figure 7. Temperature increase as a function of the fill rate for helium and hydrogen

Fig 8 shows of the temperature evolution as a function of the fill rate and the final pressure. It is observed that in certain cases very high temperatures are obtained after a full filling of an empty tank (e.g. pressure changes from 0 to 72 MPa at an average rate of 2.2 bar/s). This phenomenon has also been found in the experiments performed by Kim et al. [11]. Although these spikes lead to temperatures outside of the established 85 °C limit they are short-lived and disappear approximately 10 minutes after the filling process has been finished.

Fig. 8 stresses the observation that the temperature rise during fast filling is notably influenced by the fill rate (e.g. shown by the triangles in Fig. 8) and the initial tank pressure (observed for 72 MPa fillings). One item worthwhile to mention and something we will have a closer look into with the next experiments is the observation that long fill times lead to a greater dispersion of the temperature increase in the filling experiments. As such the 15 MPa final pressure values with the low fill rate between 0.1 and 0.9 bar/s (and an accordingly long fill time) feature a temperature range of ca. 50°C, while the companion with the higher fill rate of 2.9 bar/s (and a shorter fill time) shows a temperature range of only 8°C between the two values. This trend is also observable for higher final pressure values but not as dominant as for the lower ones.

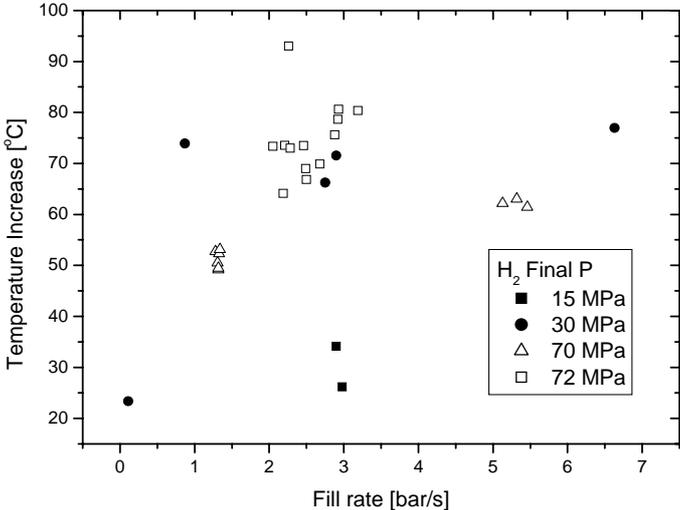


Figure 8. Increase of gas temperature as a function of the fill rate for different hydrogen final filling pressures

Fig. 9 depicts the temperature increase at TC-5 versus the duration of the overall filling process. As one would expect, the general trend is that a shorter filling duration will lead to a higher temperature build up inside the tank. Interesting to note however is that the data points are aligned along a trend line, which might be of second or third order. This alignment is independent of the pressure change and the initial pressure value, indicated in the figure by data points with different symbols. At this point the trend line does not provide a high confidence level; more data are needed for durations between 500 to 2000 seconds and particularly 2500 seconds.

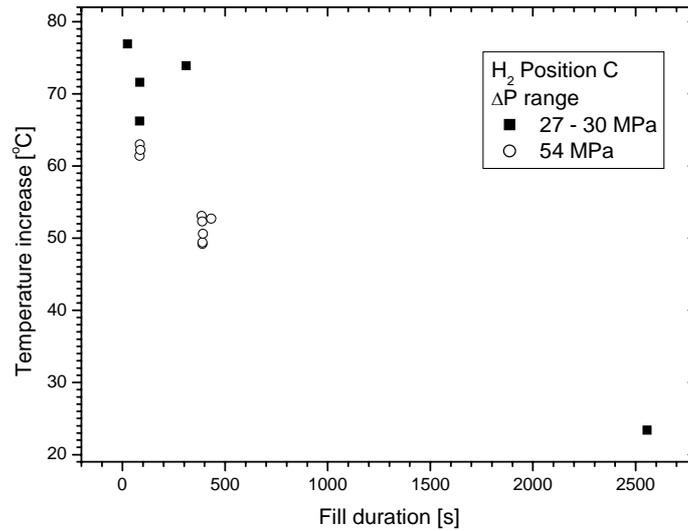


Figure 9. Increase of gas temperature as a function of the fill duration for different pressure increments

As said before, temperature evolution in the tank has been measured also during long-term static pressure experiments following a filling test. The main goal of these experiments was the assessment of the pressure decay and the time required for stabilizing gas temperature after filling. An additional objective is the mapping of local temperatures evolution to serve as validation for modeling work by computational fluid dynamics. During these experiments also the hydrogen concentration in the sleeve has been recorded, to assess the amount of hydrogen permeated from the tank and the joints. Table 4 summarizes the main findings. Note that more than 10 hours are required for the internal gas temperatures to converge to the equilibrium temperature.

Table 4. Pressure and temperature results for long-term static pressure experiments

Fill rate [bar/s]	P start [MPa]	P end [MPa]	Duration [h]	T initial [°C]	T final [°C]	Time to stabilize T [h]	T ambient [°C]
2.19	72.04	57.13	26.22	91.78	20.05	17	-2.3
2.28	72.2	51.97	29.20	92.8	19.84	10	4.5

The evolution of tank pressure and temperature as well as the concentration of hydrogen in the sleeve is depicted in Fig.10. As soon as the filling finishes, the temperature sharply decreases due to the heat transfer from inside the tank to its outer surface that is at much lower temperature (maximum 55 °C) since the sleeve is kept at room temperature. As temperature decreases the pressure does as well and hence it takes ten hours to reach equilibrium values.

The amount of hydrogen permeated to the sleeve increases during the period in which the temperature is sharply falling. In the case of Fig.10, the instantaneous hydrogen concentration reaches a maximum value of 1700 ppm after 4.5 hours and then decreases following an exponential trend.

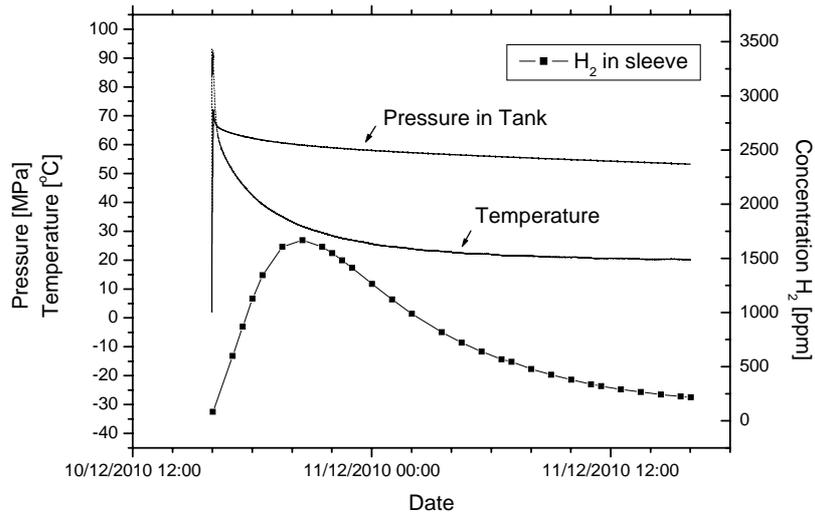


Figure 10. Evolution of pressure, temperature and hydrogen concentration during static permeation test

The GasTeF results are applied to validate existing and future standards and are used as input to pre-normative research for the development and improvement of performance characterization methodologies for high-pressure hydrogen storage.

A point of concern is that the findings presented in this paper show how the maximum temperature during the filling process of a type 4 tank can exceed the maximum temperature of 85 °C, established in the current technical regulations and standards. Based on the experiments this 85 °C seems too limiting for hydrogen filling processes involving high pressures in a short period of time and, in particular, when the hydrogen tank is almost empty. The major question is whether this maximum temperature is appropriate for the material used in the tank. If the temperature overshoot is happening for a short amount of time only –such as in 10 minutes spike- then the higher gas temperature might not even affect the tank liner, as the low thermal conductivity of hydrogen will limit the heat transfer to the plastic liner, leading to great temperature differences between the gas and the liner.

CONCLUSIONS

The temperature evolution in different fast filling conditions has been measured by means of a dedicated designed thermocouple array that is inserted in the tank. The results are in good agreement with those found in literature and serve as validation for the computed fluid dynamic modeling of the fast filling process, also performed at the JRC-IE. In this regard one of the next steps is to place the thermocouples touching or as close as possible to the tank internal surface in order to obtain accurate measurements of the liner temperature during filling and emptying.

Future work will focus on optimizing the fill process in terms of temperature evolution, filling time and stored hydrogen mass. Also foreseen is an improvement of the pressure regulation system so that the mass flow rate during filling and emptying can be tuned.

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