Design of gravimetric primary standards for field testing of hydrogen refuelling stations

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Abstract

The Federal Institute of Metrology METAS developed a Hydrogen Field Test Standard (HFTS) that can be used for field verification and calibration of hydrogen refuelling stations. The testing method is based on the gravimetric principle. The experimental design of the HFTS as well as the description of the method are presented here. The HFTS has been tested at METAS with nitrogen gas at -40°C to mimic a refuelling process in the field. Laboratory tests have shown that icing on the pipes of the HFTS have a non-negligible impact on the results. The major uncertainty components have been identified and assigned values. The required expanded uncertainty of 0.3% could be achieved. A detailed uncertainty budget has been presented and shows that the scale is the largest contributor; buoyancy corrections only play a minor role. For the lowest uncertainty measurements, appropriate waiting times or cleaning methods to get rid of icing are required.

1. Introduction

A large hydrogen infrastructure is currently in development across Europe. The industry faces the dilemma that they are required to meet certain measurement requirements set by legislation that currently cannot be followed due to the lack of available methods and standards. One important metrological challenge is the accurate measurement of the amount of delivered hydrogen during refuelling.

Hydrogen refuelling stations look and operate in a very similar way to petrol stations, the only difference being that the delivered amount is given in kg. The SAE J2601 [1] establishes the protocol and process limits for hydrogen fuelling of light duty vehicles so that the vehicle storage tanks don't overheat or overfill. Vehicles are typically refuelled with precooled hydrogen gas from dispensers within 3 min to 5 min from banks of pressurized cylinders. Fuel delivery temperature, the maximum fuel flow rate, the rate of pressure increase and the ending pressure are all parameters that are defined by process limits and are affected by ambient temperature, fuel delivery temperature and initial pressure in the vehicle's hydrogen tank. During a fill, temperature and pressure span wide ranges: pressure can go from 0.1 MPa up to a nominal working pressure of 70 MPa; to allow short filling times, hydrogen can be precooled down to -40°C. Mass flow is determined

by a pressure-ramp rate (PRR) that depend on initial pressure, available volume and temperature. We are thus far from steady conditions and only limited studies of high pressure hydrogen flow meters und transients conditions have been performed [2, 3].

International requirements propose accuracies for meters used in hydrogen refuelling station [4] and only very limited studies have been performed on how to test, inspect and verify such systems under laboratory conditions and in the field.

In the EMPIR Metrology for Hydrogen Vehicles (MetroHyVe) project, this is addressed through the development of several mobile gravimetric standards for field verification as well as an investigation into the use of substitute fluids for laboratory testing or calibration of flow meters used in hydrogen refuelling stations (HRS). In the gravimetric method, the dispensed mass is determined by weighing the amount of delivered hydrogen collected in a pressure vessel on a scale. We chose the gravimetric method because it is a well-known method that should allow obtaining the needed expanded uncertainty of 0.3%, one fifth of the proposed requirement of 1.5% Maximum Permissible Error (MPE) for flow meters.

In the course of the MetroHyVe project, three field test standards are being developed by members of the consortium: Justervesenet, METAS and VSL while CESAME Exadebit is using a standard built by Air Liquide.

In this work, we constructed a Hydrogen Field Test Standard certified for measurements in an environment with explosive atmosphere and performed measurements in the laboratory using nitrogen gas cooled down to -40°C to mimic real conditions as encountered in a HRS. The major uncertainty components have been identified and assigned values. The required expanded uncertainty of 0.3% could be achieved and a field test phase will follow this work.

2. Experimental design

2.1 Mechanical design

The Hydrogen Field Test Standard (HFTS) consists of two 36 L pressure tanks mounted into an aluminium frame. The tanks are type 4 cylinders (carbon fibre-reinforced epoxy with a plastic liner) with a service pressure of 70 MPa (at 15°C), corresponding to a capacity of 1.44 kg H_2 each. The nominal empty mass of each tank is 33 kg with dimensions of 320.8 mm x 910.3 mm. Figure 1 shows the HFTS in its frame resting on its aluminium base plate (1900 mm x 1000 mm). The total weight is around 400 kg. The HFTS alone weighs around 150 kg.

The HFTS is equipped with two 27 cm long Pt 100 probes inserted at one end of each tank and two digital pressure transducers with a 100 MPa range. Additional Pt 100 probes are mounted on the HFTS to monitor temperature in the tubing and around the scale. Passive pressure gauges are also mounted before the tanks. A Coriolis mass flow meter is also part of the HFTS and can be placed in series with the piping leading to the tanks for monitoring or eventual calibration purposes.

The frame is mounted on a 300 kg scale with 0.1 g resolution for gravimetric measurements. The weight of the frame can be lifted from the scale by a load removal system activated by a hand pump. The complete system (HFTS + scale) is placed on an aluminium base plate, which can be lifted with a forklift and placed into a van for transport.

Accompanying the HFTS are a secondary ESD plastic frame to protect the scale from the environment (shown in Figure 2), a mobile data acquisition system with laptop and a 4 m tall stainless steel vent stack with support for venting the hydrogen gas in the field after a fill. During transport, the HFTS's load is removed from the scale and held in place by locking nuts. A detailed description of the operating instructions is part of the internal documentation of the HFTS.

Figure 1: HFTS resting on its base plate.

Figure 2: HFTS with housing, here partially closed. The open space between base plate and housing is 25 cm. The housing is not leak tight.

2.2 Flow scheme

Figure 3 shows the Piping and Instrumentation Diagram (P&ID) of the HFTS. The system is composed of three lines: an inlet line connected to the hydrogen dispenser, a purge line to flush the system with N_2 and an outlet line for blowing the tanks down. The components located in the blue box are part of the frame that is being weighed on the scale. The hydrogen from the dispenser enters the HFTS through a nozzle as mounted on a car and is guided into the tanks. The gas can pass through the Coriolis mass flow meter, depending on the position of the needle valves V-4, V-1 and V-5. After filling, the tanks are emptied through a vent stack after passing through a cascade of pressure reducing valves PR-1 and PR-2 located after the needle valve V-9.

The base plate accommodates the piping for the load removal system (in red in Figure 3) as well as the piping for flushing and purging the tanks of all hydrogen gas before transport. Several nozzles placed around the frame allow flooding the ESD

housing with an inert gas during the measurements to prevent eventual icing on the pipes.

All the piping in contact with hydrogen is made of medium pressure ¼" tubing, NPT and FK series fittings and valves in 316-stainless steel.
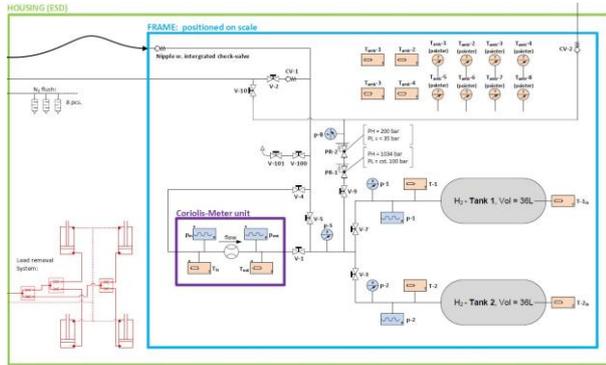

Figure 3: HFTS Piping and Instrumentation Diagram.

2.3 Electrical scheme and Data acquisition

The HFTS will store high-pressure hydrogen during field-testing and is therefore considered as equipment in an environment with explosive atmosphere (ATEX Zone 2). This puts some constraints on the design of the electrical scheme and data acquisition (DAQ) system as well as on the choice of sensors.

Figure 4 shows the electrical scheme of the HFTS. The components in the coloured part are located in the explosive atmosphere zone and are all certified. This includes the temperature probes, the pressure sensors, the scale and the Coriolis mass flow meter. These last two instruments are considered as non-arcing and are connected to the DAQ system through their own transmitters or readout modules. The remaining sensors are connected through terminal boxes to safety barriers located in the DAQ rack outside the ATEX Zone. An earth monitoring system (in yellow in Figure 4) guarantees that the HFTS and its DAQ system are continuously grounded with the hydrogen refuelling station to prevent electrostatic charges as ignition sources.

The DAQ system is shown in Figure 5 and acquires data from the scale and the temperature and pressure sensors. All cables can be plugged or unplugged from the HFTS to eliminate torqueing of the scale during weighing. The Coriolis mass flow meter is connected through a dedicated transmitter that is not part of the DAQ system.
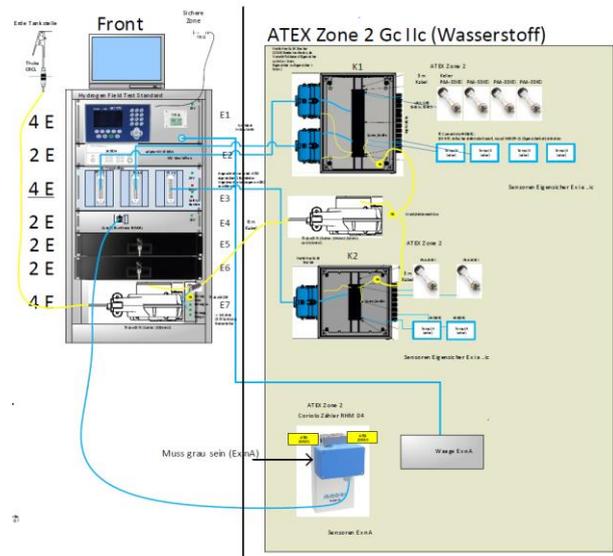

Figure 4: Electrical connections from the DAQ system to the ATEX Zone 2.

Figure 5: DAQ system with the various components (top to bottom): scale display, ambient conditions monitoring, multimeter and connection cables to the HFTS.

3. Description of the method

The HFTS is used to measure the mass of gas delivered into a vehicle. The method will be presented here.

The dispensed mass into the HFTS is calculated by:

$$m_{H2} = m_2 - m_1 \qquad (1)$$

Where *m* is the true mass and the subscripts denote the mass of the HFTS before and after the filling, respectively. The mass indicated by the

scale needs to be buoyancy corrected and for that we need the volume of the tank and of the frame. The HFTS tank volume is a function of pressure and temperature and is given by:

$$V_{tank} = V_0 \cdot (1 + 3 \cdot \alpha \cdot \Delta T) \cdot (1 + \lambda \cdot \Delta P) \quad (2)$$

where $V_0 = 59.5$ L is the external tank volume at ambient conditions with no internal pressure , $\alpha = 2.0 \cdot 10^{-6}$ °C^{-1} the linear thermal expansion coefficient, $\lambda = 2.2 \cdot 10^{-10}$ Pa^{-1} the pressure expansion coefficient, ΔT and ΔP are the difference of the temperature and pressure from the reference values, respectively. If we take into account a maximum temperature difference of 80 °C, one obtains a volume correction factor of 1.00048. The pressure expansion coefficient has been determined experimentally during a refuelling up to 70 MPa and from manufacturer's data. The thermal and pressure expansion coefficients are very similar to values published elsewhere [3] and have been assigned an uncertainty of 10% (k=1). Taking a maximum pressure difference of 87.5 MPa gives a correction factor of 1.01925. Thermal expansion will be completely neglected in the remaining description of the method as its correction factor is much smaller compared to the pressure correction. The external volume of the tank has been given an uncertainty of 5 L (k=1). The volume of the frame has been determined using the CAD drawing of the HTFS (70 L) and has been assigned an uncertainty of 5 L (k=1).

From Equations (1) and (2), we can now calculate the dispensed mass into the HFTS corrected for buoyancy and apparent mass reading from the scale:

$$m_{H2} = (W_2 - W_1) \cdot \left(1 - \frac{\rho_0}{\rho_N}\right) \quad (3)$$
$$+V_0 \cdot [\rho_{air2} \cdot (1 + \lambda \Delta P_2) - \rho_{air1} \cdot (1 + \lambda \Delta P_1)] + V_{frame} \cdot (\rho_{air2} - \rho_{air1}),$$

where W are the readings of the scale and the subscripts denote the reading before and after the filling, respectively. The factor $\left(1 - \frac{\rho_0}{\rho_N}\right)$ turns apparent mass into true mass where $\rho_0 = 1.2 \frac{kg}{m^3}$ and $\rho_N = 8000 \frac{kg}{m^3}$ are the densities of air and stainless steel at reference conditions, ρ_{air} is the density of the air around the scale and the tanks before and after the fill and is calculated using the formula by Giacomo [5].

To obtain a feeling for orders of magnitude, a complete fill in the HFTS corresponds to 2.9 kg of hydrogen gas in the tanks at a pressure of 70 MPa. This yields a volume expansion of 0.92 L for each tank. Under identical ambient conditions before and after the fill, we obtain a buoyancy correction of 2.12 g (0.08%) for both tanks. The term due to the volume of the frame only plays a role if ambient conditions change.

Laboratory tests were performed to reproduce field tests as closely as possible. The aim was to elaborate a testing procedure and practice using the HFTS before going into the field.

All laboratory measurements were performed with nitrogen gas from a bundle as a 5.5 MPa gas source to fill the HFTS. Before entering the HFTS, the gas was cooled down to -40 °C by a heat exchanger to reproduce the temperature conditions of hydrogen as delivered by a HRS. Pressure and temperature in the tanks was monitored continuously during the fill. The frame was always enclosed or partially enclosed by the housing, the latter geometry leads to a better air circulation around the scale.

To perform the measurements, we followed several steps:

1. Disconnect all the cables and hoses from the frame, lower the HFTS onto the scale and weigh the empty HFTS, record the ambient conditions
2. Lift the HFTS from the scale, connect all sensors and measure tank pressure and temperature as well as temperature of the air in the frame
3. Connect the gas source to the HFTS inlet and fill the tanks. During the fill, all sensors are monitoring and recording data.
4. Disconnect all the cables and hoses from the frame and lower the HFTS onto the scale
5. Wait until scale reading stabilises and record value
6. Lift the HFTS from the scale and connect all sensors. Connect the vent stack and blow down the gas.

4. Results

Typical results for the pressure rate and temperature profiles for a fill up to 4 MPa are shown in Figure 7. The pressure ramp rate (PRR) during the fill is around 1.14 MPa/min. During this period, temperature in the tanks increased due to compression heating while temperature of the tubing decreased below the freezing point of water due to the cold nitrogen gas flow. The air temperature around the scale remains more or less constant. After the fill, all temperatures converge more or less slowly toward the current ambient condition.
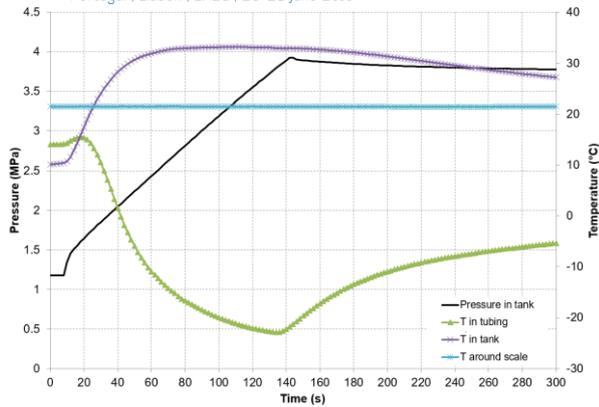
Figure 6: Pressure (in black) and temperature profiles from the HFTS during a fill, PRR = 1.14 MPa/min.

Figure 7 shows the temperature profile around the scale starting 60 s after the fill during 1 hour with the HFTS housing closed. Due to the temperature increase in the tanks, heat is transferred to the air around the scale, which then returns to ambient temperature after around 40 minutes. This can be considered as a worst case because of the poor air circulation around the HFTS and yields a maximum drift of 0.3 °C.
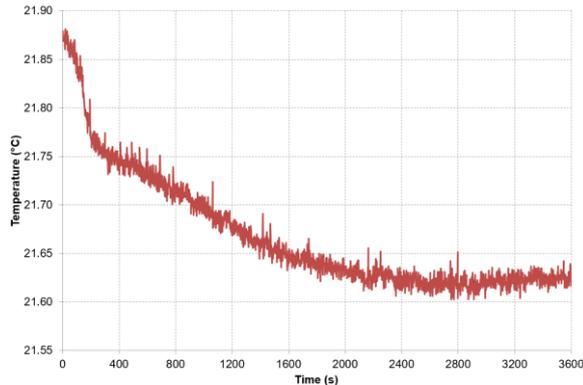

Figure 7: Temperature profile around the scale after a fill with closed housing.

The cold gas flowing through the HFTS causes part of the humidity present around the HFTS to condense and freeze on the pipes. This quantity of ice will be weighed but should not be part of the determination of the mass of dispensed gas. Figure 8 shows the scale reading profile shortly after the fill with a closed and a partially closed housing (opening of 25 cm). We notice an increase in mass due to condensation that reaches a maximum before a slow decrease because of melting and evaporation, eventually reaching a constant scale value. The maximum quantity of lost mass seems to be independent of the position of the housing and amounts to more or less 7 g in our case.
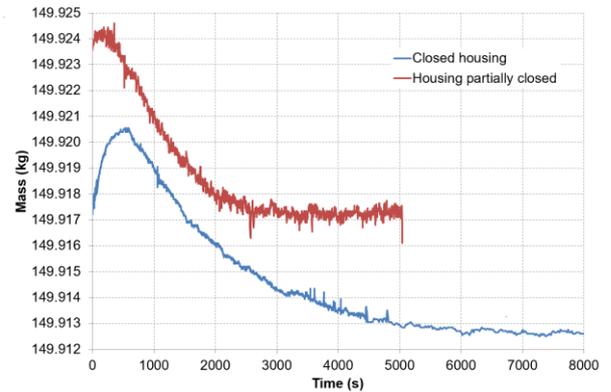
Figure 8: Scale reading profile as a function of time after the fill with a closed and partially closed housing. .

This is by far not negligible. On a brighter side, closing the housing partially accelerates the loss of mass and leads to a stable scale value with less than 1 g of spread after approximately 30 minutes.
We also performed similar measurements with a closed housing flooded with nitrogen or argon gas. We chose argon because it is heavier than air and filled the housing up to the top and remained in the housing, contrary to nitrogen. With both gases, we still observed icing on the pipes but less than with air. Flooding with an inert gas brings more unknowns in the buoyancy correction because the flooding level is not well defined. In addition, the amount of gas needed is quite large.
It looks like the best solution is to measure with a partially closed housing, cleaning/drying the pipes after the fill and wait for the scale to show a stable value.

5. Uncertainty budget

The various contributions to the uncertainty budget are addressed in this section. The uncertainty of the gravimetric measurement can be calculated by

$$\left[\frac{u(m_{H2})}{m_{H2}}\right]^2 = \sum_i S_{x_i}^2 \cdot \left(\frac{u(x_i)}{x_i}\right)^2 \tag{4}$$

where x_i are the measurands from Equation (3) and S_{x_i} are the normalised sensitivity coefficients for each variable and can be calculated by

$$S_{x_i} = \frac{\partial m_{H2}}{\partial x_i} \cdot \frac{x_i}{m_{H2}}. \tag{5}$$

The scale has a resolution of 0.1 g and was calibrated against METAS standard masses with the frame on the scale. We observed a maximum deviation of 0.5 g. The calibration was checked over time and the scale showed a drift of 0.4 g over

90 minutes. The scale will always be calibrated on site in the field during measurements. As the effect of field use could not be determined yet, we consider a conservative uncertainty value of 0.7 g (k=1) for the scale.

Air density depends mainly on the parameters pressure, temperature and humidity. These quantities are continuously logged with uncertainties of 50 Pa, 0.3 °C and 5%, respectively and allow the determination of air density with an uncertainty of 0.15% (k=1) during measurements.

The digital pressure transducers have a resolution 2 kPa and a long-term stability of 100 kPa according to the specifications. The sensors have been calibrated and shown to lie within the specifications of 200 kPa.

Table 1 presents a summary of the uncertainty components and their magnitude for the gravimetric method for a test collection of 1 kg of gas. As expected, the scale is the largest contributor to the uncertainty, followed by the air density. The values presented here yield an expanded uncertainty of 0.22% or 2.2 g for 1 kg of gas collected.

Table 1: Summary of uncertainty components and their magnitude for the gravimetric method for 1 kg of gas collected.

Uncertainty component	Nominal value	$u(x_i)$ %	Contribution %
Initial mass	150.0000 kg	$4.7 \cdot 10^{-4}$	40.5
Final mass	151.0000 kg	$4.7 \cdot 10^{-4}$	40.5
Tank volume	0.120 m^3	4.17	0.16
Frame volume	0.070 m^3	7.14	< 0.1
Initial air density	1.1500 kg/m^3	0.15	8.9
Final air density	1.1500 kg/m^3	0.15	9.0
Initial tank pressure	0.10 MPa	20	< 0.1
Final tank pressure	35.00 MPa	0.057	< 0.1
Pressure coefficient	$2.2 \cdot 10^{-10}$ Pa^{-1}	10	0.93

The uncertainty contribution due to condensation and icing of the pipes can be minimised if we wait at least 20 minutes or clean the pipes before reading the value of the scale. A 1 g spread as considered in section 4 under the assumption of a rectangular distribution yields an uncertainty contribution of 0.58 g (k=1) and leads to an expanded uncertainty of 2.5 g (0.25%) for 1 kg of gas collected. The required expanded uncertainty of 0.3% or less is therefore achieved. Futher measurements in the field under real conditions are planned to validate some of the assumptions taken in the present uncertainty budget.

7. Conclusion

The design of the HFTS and its associated gravimetric measuring method have been presented in this paper. Experimental results obtained with te HFTS under laboratory conditions with cold nitrogen gas at -40°C to mimic real fill conditions in a hydrogen refuelling station showed varying temperature and scale reading profiles. Condensation on the HFTS leads to a change in mass over time of up to 7 g, which is not negligible compared to the minimum quantity of 1 kg to be weighed during field-testing. A complete uncertainty budget has been presented and we obtained an expanded measuring uncertainty for the HFTS of 0.25%. The results presented in this paper can serve as a guide for future designs of similar field testing instruments based on the gravimetric principle.

References

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