H2 Reference Fuel and Reference Gases

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Introduction

This document is a scoping paper on *the need and feasibility for a hydrogen reference fuel and for other reference gases* in the context of the establishment of a GTR for H2-powered vehicles. As agreed at the 3rd meeting of the Subworking Group Environment of the Informal UN-ECE Informal GRPE Working Group on Hydrogen Fuel Cell Vehicles (H2SGE-03-WP-02), JRC would initiate such a scoping paper.

The discussions during the meeting identified that reference gases could potentially be relevant and useful in three areas covered by the GTR: fuel quality, emissions and fuel consumption. The question was raised to what extent these different applications resulted in different requirements for a reference gas.

The purpose of the present document is **not** to provide an overview of ongoing standardisation and regulatory activities worldwide on these three topics or of the supporting R&D activities.

Purpose and resulting requirements for reference gases in support of GTR for H2 vehicles

1. Checking purity (quality) of hydrogen for use as a propulsion fuel
   - 1.1. H2 purity (quality) is an issue in FC vehicles, much less in H2-ICE vehicles. At present, only PEM type FC are considered for propulsion applications in passenger cars and light vehicles. Impurities in the reactants (both in hydrogen at the anode, but also in air/oxygen at the cathode) affect the catalyst performance and hence the electrochemical reactions as well as the membrane properties.
   - 1.2. Impurities in the H2 fuel depend primarily on the H2 production method, although the storage method and medium may also play a part. The most important contaminants in H2 produced by natural gas reforming (the most commonly used production method) are CO, sulphur containing compounds (H2S in particular), nitrogen containing compounds (NH3), and unsaturated and aromatic hydrocarbons.
   - 1.3. To enable calibration of equipment for impurity analysis, a reference gas containing specified impurities/contaminants in an otherwise pure H2 matrix (i.e. carrier gas) is needed
   - 1.4. According to consulted metrology institutes the production of such a reference gas (reference fuel) is technically feasible, but stability and conservation/storage period require attention and investigations.
   - 1.5. If H2 is not only used for propulsion but also for APU (for which a different type of fuel cell may be used), other purity requirements will prevail. However, because only one kind of CH2 is expected to be stored on board, the most stringent requirements (corresponding to PEMFC for propulsion) will apply.
   - 1.6. In the context of GTR such a reference fuel is not needed for vehicle certification *per se*, but for reducing the impact of fuel-dependent factors on vehicle performance aspects covered by the GTR, such as emissions and fuel consumption. When the H2 used during the vehicle certification test complies with the fuel quality specifications the variability in test results is considerable decreased, allowing more accurate verification of true performance.
   - 1.7. In the context of GTR a H2 reference fuel should be globally recognised (which would be achieved by a certification according to international standards, such as ISO Guide 34 and corresponding equivalence studies)

2. Monitoring of vehicle emissions
   2.1. Vehicle certification requires measurement of tail-pipe emissions during a test cycle. This necessitates the use of calibrated emission monitoring equipment.
   2.2. Measurement of tail-pipe emissions from fossil fuelled ICE vehicles uses a constant volume sampling (CVS) system that compensates for load variations during the test cycle by maintaining a constant total flow rate of vehicle exhaust plus dilution air. The diluted exhaust is collected and the concentrations of emission products CO, CO2, HC and NOx are determined (g/km).
   2.3. When using H2 as a fuel, either in FC or ICE, the emissions are much lower than those from conventional ICE vehicles or simply there are not present (i.e. FC except H2O). This requires additional measures to ensure accuracy of emission measurement and calibration of the emission monitors.
   2.4. The major emission product from H2 powered vehicles is water vapour. The associated humidity may affect the accuracy of measurement of pollutants (e.g. non-dispersive infra-red analysers for CO and CO2 are known to be sensitive to H2O).
   2.5. Emissions from H2-vehicles powered by FC are different from those with ICE, because different purity requirements and hence impurity contents of the fuel. For ICE additional emissions are NOX from the combustion of the air-fuel mixture as well as carbon compounds from engine lubrication (UHC, CO, CO2).
   2.6. For the calibration of the different types of emission monitors reference gases are needed. Because of the lower emission levels to be measured these must have a higher purity than those for calibration of emission measurement equipment for conventional ICE vehicles. This also applies for the gases used as "zero-gas" in analysers based on differential measurement (e.g. NDIR).
   2.7. When instead of total emissions over the test cycle ("bag" measurement) emissions are measured instantaneously on a second-by-second basis (“modal” measurement) (e.g. to assess performance of catalyst and engine system at a particular time in the cycle), the instantaneous exhaust mass flow rate must be determined. This represents a serious technical challenge. In this case also more stringent requirements apply for the emission monitors (e.g. response time).

3. Measurement of fuel consumption
   3.1. Vehicle certification requires measurement of fuel consumption during a test cycle. For H2-powered vehicles a number of methods have been and are under investigation (see Annex I). Each of these methods has disadvantages.
   3.2. The need for harmonisation of fuel consumption measurement procedures in the context of the GTR is addressed in the previous technical report to SGE (Technical Report I). The present document therefore focuses on the potential role that reference gases could play in this respect.
   3.3. In the context of a GTR, the use of a single universally accepted method definitely provides added value. Moreover, for reasons of economy, efficiency and comparability with certification of non-H2 powered vehicles, the use of an "elemental balance" method as for conventional ICE vehicles (carbon-balance) that does not require vehicle modifications, presents huge advantages.
   3.4. This is achieved by using the so-called Hydrogen-Balance method which measures the hydrogen-containing compounds H2O (non-dispersive infra-red analyser) and unburned

H2 (sector field mass spectroscopy) from the FC or ICE exhaust. The method requires some modifications to the testing procedures and system that are used for conventional ICE vehicles (as explained in Annex I).

3.5. Because they are based on the same measurement principle, fuel consumption and emission monitoring have similar requirements for equipment calibration. Hence, the above considerations on the use of reference gases for calibration of low-level emission monitors also apply for H2 fuel consumption measurement. Extra requirements originate from the use of additional H2O and H2 analysers. Because the expected concentration of H2 in the exhaust is very low, the H2 sensor can be calibrated using a readily available appropriate reference gas. However calibration of the H2O NDIR analyser calibration requires a dedicated humidification system.

3.6. For FC vehicles an Oxygen-Balance method based on measurement of the oxygen concentration in the exhaust has also been proposed. The method is not directly based on mass conservation, but on the measurement of a relatively small decrease in oxygen concentration between the inlet and outlet of the fuel cell stack, which requires a high accuracy of the oxygen analyser.

3.7. For FC vehicles, measurement of electrical current generated by the FC can also be translated into H2 consumption. However internal losses from hydrogen leaks and cross-over, while definitely contributing to consumption, are not captured by such a measurement.

Requirements

The implementation in the context of the GTR of harmonised fuel quality, emission and fuel consumption measurements translates into the following needs for further R&D and technical requirements for reference gases:

1. Fuel quality for FC vehicles

Issue: tolerance level of impurities to limit degradation of performance to acceptable level

Need: pre-normative research into

- test methods for quantifying performance under dynamic conditions reproducing typical driving cycles
- reliable detection and quantification of impurities in gaseous H2, including validation of high-pressure sampling methods
- understanding and quantifying the effect of impurities (single as well as multi-constituent) on FC performance and determination of acceptable level of impurities

Implications for feasibility of reference fuel:

- identification of type and amount of impurities in hydrogen carrier gas
- investigate stability and conservation/storage period

2. Emission monitoring for FC and ICE vehicles

Issue: measurement challenges related to high water vapour and low emissions of other pollutants

Need: pre-normative research into

- low-cost and reliable analysis methods and emission monitors
- calibration of sensors

Implications for feasibility of reference gases:

- calibration of emission analysers at low detection ranges requires higher purity levels in the reference gas than for conventional ICE vehicles

3. Fuel consumption (FC and ICE)

Issue: same as for emission monitoring

Need: additional pre-normative research into

- Further validation of hydrogen-balance approach and equations for different vehicle types
- Cost-effective calibration method for H_2O detectors

Implications for feasibility of reference gases:

- need for dedicated reference gas(es?) allowing calibration of H_2O detectors over wide H_2O range
- for calibration of H_2 detectors: none (e.g. 0.5% H_2 in N_2).

Conclusion and Recommendation

International collaboration in pre-normative research as well as close interaction with international standardisation bodies aimed at harmonisation of test requirements, of test methods and of test equipment performance for fuel quality, emission and fuel consumption measurement can greatly contribute to the establishment of the GTR. This should also include the development and certification of a reference fuel and gases for these three applications according to ISO standards which would ensure their global recognition.

Annex I: Measurement of Fuel Consumption

Fuel consumption is defined as the mass amount of fuel used by a vehicle in a prescribed test cycle, expressed in g/km. In principle, three methods exist for experimentally determining gaseous H2 consumption in FC or ICE-vehicles: (i) determination of fuel mass change in the container before and after test, (ii) determination of H2 flow rate and (iii) measuring the concentration of relevant species in the exhaust with subsequent back-calculation to fuel consumption. A compilation of available methods is given in table 3.

Methods (i) and (ii) require a test vehicle to be supplied with hydrogen from an external, rather than the onboard tank. This requires dedicated live hydrogen feeds during testing and adjustment of various components in the test vehicle (with associated safety implications). These methods are also not suitable for vehicles with liquid hydrogen storage.

(i) Determination of mass change

Mass change is measured statically before and after the test, either by weighing the fuel tank with its H2 contents, or by determining the equilibrium temperature and pressure before and after testing in a storage tank of known volume (PVT). The former method suffers from the disadvantage that the weight of H2 is very small compared to that of the tank, resulting in low measurement accuracy. PVT measurement needs also less instrumentation and test personnel, and hence potentially offers higher repeatability and lab-to-lab reproducibility. It requires use of a standardised equation for hydrogen density as a function of temperature and pressure.

(ii) Flow rate measurement

This type of measurement allows determining the instantaneous flow rate of hydrogen. Different measurement principles exist: mechanical, optical, thermal, ultrasonic, Coriolis, etc. They all require an intervention to the fuel supply line which can introduce inaccuracies. Also dedicated signal treatment and analysis equipment is needed for all but the simplest flow meters.

(iii) Method based on emission measurement

At present, US-EPA only accepts weighing, PVT and Coriolis mass flow according to SAE J2572 for the determination of H2 gas fuel consumption in fuel cell and fuel cell hybrid vehicles. Adaptation of a method based on emission measurements to determine fuel consumption in conventional ICE vehicles receives increasing attention. It uses identical equipment as for emission measurement and works on the conservation of mass principle: what goes into the engine or fuel cell must come out as exhaust components. This measurement of fuel consumption does not require direct contact with the fuel, which contributes to enhanced accuracy and simplicity.

For gasoline- and diesel-fuelled ICE vehicles the Carbon-Balance method is used. The total amount of carbon in the exhaust must have gone in as fuel (C-containing species in the exhaust from non-fuel sources are much lower than from the fuel and are hence neglected). The measured concentrations of C-containing species (CO, CO2, THC) in the diluted exhaust volume that is collected during the test cycle are calculated into a fuel consumption for the particular test cycle.

For H2-powered vehicles, a Hydrogen-Balance method (sometimes also called water-balance) is applied, which measures the hydrogen-containing compounds H_2O (non-dispersive infra-red analyser) and unburned H_2 (sector field mass spectroscopy) from the exhaust. This method requires some modifications to the testing procedures and CVS system that are used for conventional ICE vehicles. These arise mainly from two factors:

- It must be ensured that all H_2O present in the exhaust is effectively measured. This requires avoiding condensation on cold parts (e.g. vehicle exhaust pipe at cold starts, exhaust gas lines to monitor) and diffusion of H_2O from the exhaust into the dilution pipe.
- H_2O in the exhaust may originate from other sources than oxidation of the H_2 fuel, namely the humidity in the environment (motor intake) and in the dilution air, which must not only be known but also remain constant during the test cycle

When the above points are appropriately addressed, the fuel consumption determined by the Hydrogen-Balance method agrees with the results obtained by the three EPA-recognised methods (SAE Paper 2008-01-1038). The method works for FC as well as for ICE vehicles.

Table 1 Adapted from HarmonHy WP4 Deliverable D4.1 – Industrial and societal needs – Sept. 2006

Test method	Description	Advantages	Disadvantages and issues
Carbon balance method	Derived from exhaust gas, carbon content in fuel and exhaust gas are the same	Simultaneous measurement during exhaust gas test Vehicle remodelling unnecessary	N/A for direct hydrogen FCV
Flow method	Direct measurement using flow meter	Field-proven for Internal combustion engine vehicles	Vehicle remodelling needed Verification of flow meters
Electrical current method	Calculated from electrical current generated in the fuel cell	Current easily measured from output wiring of the fuel cell	Gas crossover and leak Measurement of H2 purge
Hydrogen balance method	Derived from exhaust gas, H2content in fuel and exhaust gas are the same	Simultaneous measurement during exhaust gas test Vehicle remodelling unnecessary	H2 balance complicated Difficult to measure
Oxygen balance method	Measuring the decline in O2 concentration in exhaust gas	Simultaneous measurement during exhaust gas test Vehicle remodelling unnecessary	Decline in O2 is low Accuracy of oxygen analyzer
Pressure method	Calculated from press. / temp. change of fuel container	Easily to measurement Support H2 purge	Limited to a high pressure container to store fuel
Weight method	Calculated from weight change of fuel container	Direct and simple Support H2 purge	N/A for onboard measurement Connecting to gas line