

Article

Assessing the Performance of Fuel Cell Electric Vehicles Using Synthetic Hydrogen Fuel

Thomas Bacquart ^{1,*} , Ward Storms ², Niamh Moore ¹, James Olden ², Abigail Siân Olivia Morris ¹, Mathew Hookham ¹ , Arul Murugan ¹  and Vincent Mattelaer ²

¹ Atmospheric Environmental Science Department, National Physical Laboratory, Hampton Road, Teddington TW11 0LW, Middlesex, UK; niamh.moore@npl.co.uk (N.M.); abigail.morris@npl.co.uk (A.S.O.M.); mathew.hookham@npl.co.uk (M.H.); arul.murugan@npl.co.uk (A.M.)

² R&D, Toyota Motor Europe, B-1930 Zaventem, Belgium; ward.storms@toyota-europe.com (W.S.); james.olden@toyota-europe.com (J.O.); vincent.mattelaer@toyota-europe.com (V.M.)

* Correspondence: thomas.bacquart@npl.co.uk

Abstract: The deployment of hydrogen fuel cell electric vehicles (FCEVs) is critical to achieve zero emissions. A key parameter influencing FCEV performance and durability is hydrogen fuel quality. The real impact of contaminants on FCEV performance is not well understood and requires reliable measurements from real-life events (e.g., hydrogen fuel in poor-performing FCEVs) and controlled studies on the impact of synthetic hydrogen fuel on FCEV performance. This paper presents a novel methodology to flow traceable hydrogen synthetic fuel directly into the FCEV tank. Four different synthetic fuels containing N₂ (90–200 µmol/mol), CO (0.14–5 µmol/mol), and H₂S (4–11 nmol/mol) were supplied to an FCEV and subsequently sampled and analyzed. The synthetic fuels containing known contaminants powered the FCEV and provided real-life performance testing of the fuel cell system. The results showed, for the first time, that synthetic hydrogen fuel can be used in FCEVs without the requirement of a large infrastructure. In addition, this study carried out a traceable H₂ contamination impact study with an FCEV. The impact of CO and H₂S at ISO 14687:2019 threshold levels on FCEV performance showed that small exceedances of the threshold levels had a significant impact, even for short exposures. The methodology proposed can be deployed to evaluate the composition of any hydrogen fuel.

Keywords: hydrogen fuel; fuel cell performance; hydrogen quality; gas analysis; hydrogen fuel cell electric vehicles



Citation: Bacquart, T.; Storms, W.; Moore, N.; Olden, J.; Morris, A.S.O.; Hookham, M.; Murugan, A.; Mattelaer, V. Assessing the Performance of Fuel Cell Electric Vehicles Using Synthetic Hydrogen Fuel. *Energies* **2024**, *17*, 1510. <https://doi.org/10.3390/en17071510>

Academic Editors: Kyriakos Panopoulos and Michael Bampaou

Received: 19 February 2024

Revised: 1 March 2024

Accepted: 12 March 2024

Published: 22 March 2024



Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

The deployment of hydrogen fuel cell electric vehicles (FCEVs) powered by hydrogen is critical to achieving zero emissions from the transport sector. Within the European Union, carbon dioxide emissions from all road transportation increased 27.8% from 1990 to 2019 (this represented 71.7% of the EU-27 transport sector emissions) [1]. Similarly, in the US, transportation accounted for 28.5% of greenhouse gas emissions in 2021, making it the largest source of greenhouse gas in the US [2]. Many countries signed the COP26 declaration to accelerate the transition to zero-emission passenger vehicles [3]. The European Union set a new regulation (EU 2023/851 [4]) banning new petrol and diesel cars from 2035 onwards to accelerate the switch to zero-emission vehicles (ZEVs). Typically, electric vehicles, such as battery (BEV) and fuel cell electric vehicles (FCEVs) producing zero tailpipe emissions, are classified as zero-emission vehicles. In 2022, global electric vehicle sales surpassed 10 million vehicles [5]. The share of electric vehicles operating on fuel cell technology was comparatively low, with around 72,000 FCEVs operating in the world at the end of 2022 [5]. However, FCEVs are considered key for reaching zero-tailpipe emissions from transport, especially for long-distance light-duty applications requiring fast refueling (i.e., taxi fleets);

long-distance heavy duty applications, such as in trucks, rail, and maritime; and high-power-input vehicles (i.e., mining vehicles) [6]. For these applications, the performance and durability of FCEVs are critical for ensuring successful deployment.

One key parameter influencing an FCEV's performance and durability is hydrogen fuel quality [7]. International standards, such as ISO 14687 [8], EN 17124 [9], and SAE J2719 [10], define lists of chemical compounds with amount fraction thresholds in hydrogen fuel. These were established in attempt to avoid any issues with reduced performance of FCEVs. Even if the regulatory aspect is clearly established, challenges related to performance, maintenance, and the lifetime of FCEVs still arise for vehicle or fleet operators.

Currently, obtaining a direct determination of proton exchange membrane fuel cell (PEMFC) degradation rates and lifetime is challenging due to a large gap between a single-cell study in the laboratory and a stack in transport applications [11] or even an FCEV in a real-life situation. Solving the challenges around laboratory experimental bench harmonization protocols (i.e., harmonized critical evaluation indicators, fuel cell test bench environment, operation history) is crucial to progress and develop a prognostic method to accurately predict the remaining lifetime of a PEMFC [11]. The development of a test method that can assess real-life FCEV performance would provide a critical tool to advance such prognostic methods and enable proper comparisons of predictions versus real-life performance. This will allow better understanding of reversible and irreversible degradation effects for real-life FCEV cases (i.e., after a poor-quality hydrogen fuel refueling event).

The development of an infrastructure to study the performance of an FCEV powered by different hydrogen fuel qualities would complement the development of knowledge on the single-cell and stack degradation tests in the laboratory. However, there is no study on the actual impact of hydrogen fuel quality on the overall FCEV due to the technical complexity of performing such a study (i.e., how to accurately contaminate an FCEV), cost (i.e., potential irreversible damage caused to the FCEV), and representativity (i.e., there are only a small number of FCEV types available worldwide). Such a study should be carried out in accordance with the FCEV manufacturer and owner. The FCEVs should not be used after such studies (except if approval has been obtained by the manufacturer). There are several approaches that could be taken to deliver hydrogen that simulates poor-quality fuel: (1) creating a complete dedicated hydrogen refueling station (HRS) with controlled levels of contaminants, (2) using gas cylinders with bespoke composition, (3) generating a bespoke hydrogen fuel composition in the FCEV using gas cylinders and a normal HRS.

The first approach, using a dedicated HRS, would allow an FCEV to be refueled with a specific contaminated hydrogen fuel following the normal refueling process. A European project, MetroHyVe 1 [12], investigated the feasibility of such a contamination directly at an HRS. However, the study revealed that this approach was too complex in terms of engineering and with regards to how the HRS would be cleaned up to ensure clean hydrogen could be delivered following the activity. Even if such an experimental HRS was desired, there are currently no known locations where this could be accurately realized. The second approach, using gas cylinders with bespoke composition, may be easier to realize; however, two technical challenges arise: achieving the required pressure from a gas cylinder (not all gas cylinders have a working pressure of 35 or 70 MPa) and achieving the creation of stable gas compositions with a few nmol/mol of reactive gas (i.e., H_2S) in such cylinders. The third approach proposes to perform accurate contamination of an FCEV or mock-up system using direct spiking of the fuel in the FCEV tank with impurities. The approach would follow the guideline for preparing gravimetric gas mixtures (ISO 6142-1 [13]), where the hydrogen in the FCEV tank is considered the final gas mixture. While the mass of the gas added may not be directly recorded, it is possible to determine the mass of the primary reference gas transferred from the gas cylinder into the FCEV tank with a reasonable certainty. Additionally, the mass of gas in the FCEV tank may be determined based on indirect measurement of the temperature, pressure, and volume of the FCEV tank using the non-ideal gas law. Therefore, the third approach was considered realistic and simple to implement. Moreover, it allows any contamination case to be investigated

without the requirement of complex infrastructure, contaminating an operational HRS, or the generation of large volumes of high-pressure contaminated hydrogen fuel in gas cylinders. It also allows the FCEV to be driven in real-life conditions on a safe road (i.e., privately owned racetrack or circuit) to directly understand the performance impact in terms of drivability.

The article will detail the methodology used to create four reference contaminated hydrogen fuels in an FCEV using direct spiking of the fuel in the FCEV tank with impurities. The hydrogen fuel compositions in the FCEV involved three types of chemical compounds (inert, low reactivity, and reactive compounds) at a level close to the ISO 14687 threshold. Therefore, the following gases were used: nitrogen (inert gas), a known contaminant of FCEVs; carbon monoxide (low reactivity in gas cylinders); and hydrogen sulfide, a known reactive contaminant of FCEVs. The study will present the contamination apparatus and detailed experimental conditions. The discussion will highlight the determination of the reference amount fraction of each compound in the different composition experiments based on the different measurement methods available (temperature, pressure, mass). The study will present the verification of the actual mixture composition through samplings using the National Physical Laboratory (NPL) sampling tool [14] and gas analysis to assess its representativeness. Then, a discussion on the actual agreement between the theoretical value and the measured value is presented, with a focus on the sampling rig performance. The contaminated vehicle underwent a driving evaluation to assess the impact of the reference contaminated hydrogen fuel on the fuel cell (FC) performance. The results of the driving test will be presented and discussed with potential recommendations for further application.

Finally, insights on future experiments, potential improvement points for the sampling rig and/or the contamination methodology, or future FCEV performance testing will be presented.

2. Materials and Methods

2.1. Reference Gas and Pure Gas Used for the Contamination of the FCEV

2.1.1. Primary Reference Materials for Contamination of Fuel Cell Electrical Vehicles

All primary reference materials (PRMs) presented in Table 1 were prepared gravimetrically by NPL from pure compounds (high-purity nitrogen (>99.9999% BIP+, Air Products, Crewe, UK), high-purity carbon monoxide (>99.998%, Air Products, Crewe, UK), or by dilution of NPL PRM (i.e., dilution of NPL PRM containing 10 $\mu\text{mol/mol}$ of hydrogen sulfide in hydrogen) in high-purity hydrogen (>99.9999%, BIP+, Air Products). The reference materials were prepared according to ISO 6142-1 [13]. The cylinders used in this study were 10-liter aluminum cylinders with an internal SPECTRA-SEAL[®] passivation (BOC, Woking, UK). Details of preparation are provided in Supplementary Materials. The compositions of the mixtures (amount fraction and associated uncertainty) were calculated from the masses of pure compounds, PRM, and hydrogen introduced in each cylinder and using the software package GravCalc version 2 [15]. The amount fractions of each gas mixture are summarized in Table 1.

Table 1. Summary of NPL PRMs used to contaminate the FCEV with gravimetric amount fraction for CO, N₂, and H₂S and respective uncertainty ($k = 1$). When a compound was absent, the limit of detection (LOD) was quoted (<LOD).

	CO [$\mu\text{mol/mol}$]	N ₂ [$\mu\text{mol/mol}$]	H ₂ S [$\mu\text{mol/mol}$]	Matrix Gas
NPL PRM 1 (Cyl ref: NG584R2)	30.04 $\pm$ 0.26	16,871 $\pm$ 18	<0.001	hydrogen
NPL PRM 2 (Cyl ref: NG944)	144.20 $\pm$ 0.26	16,222 $\pm$ 17	<0.001	hydrogen
NPL PRM 3 (Cyl ref: NG659R3)	737.36 $\pm$ 0.33	16,126 $\pm$ 19	<0.001	hydrogen
NPL PRM 4 (Cyl ref: 2744R2)	<0.005	16,717 $\pm$ 19	0.9985 $\pm$ 0.0033	hydrogen

2.1.2. High-Purity Hydrogen for Contamination of Fuel Cell Electric Vehicles

Pure hydrogen was required for purging the contamination system. A high-purity hydrogen cylinder was transferred by NPL from a 50 L high-purity hydrogen cylinder (purity > 99.9999%, BIP+, Air Product, Crewe, UK) into an empty 10 L aluminum cylinder following the methodology presented in the previous section. The 10 L aluminum cylinder of high-purity hydrogen had a final pressure of 120 bar.

2.2. Contamination System for Fuel Cell Electrical Vehicles Experiments

2.2.1. Gas Assembly to Contaminate Fuel Cell Electrical Vehicle

The contamination was realized using an NPL-developed rig, as presented in Figure 1. All parts were made from stainless steel tube of an 1/8-inch or 1/16-inch outer diameter and treated with Sulfinert (Swagelok, London, UK). The pure hydrogen cylinder and contamination cylinder valve included an internal screw thread to minimize dead volume. The contamination cylinder was positioned on a precision balance (Mettler Toledo, Leicester, UK) to determine the mass transferred. The pressure gauge was explosive atmospheres (ATEX)-rated with a range of 0–700 bar (FLUKE, Everett, WA, USA). The gas rig was connected to a 350 bar CHV 08/C nozzle (Staubli, Bayreuth, Germany) to connect to a first-generation Toyota Mirai receptacle.

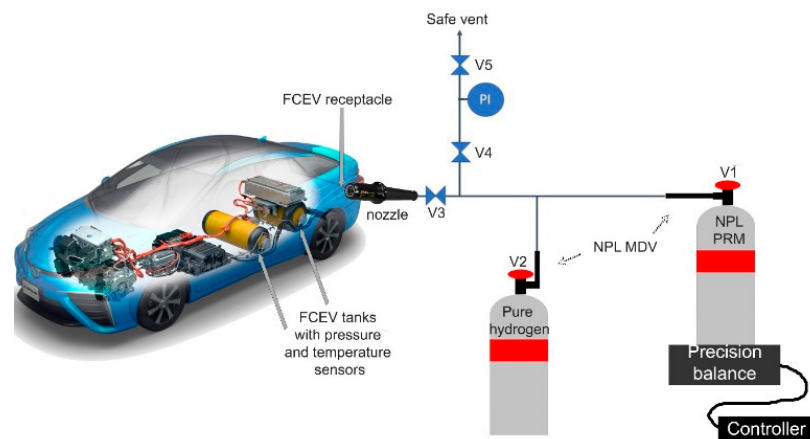


Figure 1. Contamination system in operation.

The first-generation Toyota Mirai was equipped with a temperature sensor in each tank (yellow tank in Figure 1) and a pressure sensor on the intake and outlet manifold of the tanks. No information was available on the sensor accuracy. The tank volume was estimated as 123 L in total, as the study was realized at pressure below 35 MPa. In fact, a slight variation occurs depending on the pressure inside the tank, from 122.4 L (without pressure) up to 124.9 L at 70 MPa. The data from the sensors were obtained after the experiments, once the FCEV was powered again.

2.2.2. Protocol of Fuel Cell Electrical Vehicle Contamination

For safety reasons, the FCEV was first powered off. The cylinders were safely positioned by either attaching them to a solid point or by placing them within a protected area (to prevent them from falling). The NPL PRM was positioned and centered on the precision balance with the protection area around it. The balance was levelled and verified with a 10 g reference mass. All the pressurized gas systems were connected to safety vents to prevent unsafe release of hydrogen.

First, the system was disconnected from the FCEV so that leak checking and verification of system integrity could be performed without connecting the nozzle to the FCEV. Once the system was confirmed to be leak-free, it was purged at least 7–10 times with pure hydrogen through a cycling purge (see Figure 1). The cycling purge was carried out using valve V2 with cycling pressure from 100 bar to 4 bar. The hydrogen was safely vented

through a safe hydrogen release point using valve V5. After the purge with pure hydrogen, the system was considered free of any unexpected contaminants and was then conditioned with the NPL PRM gas. This conditioning was realized through 10 pressure cycles from 100 bar to 4 bar. The conditioning was performed by actuating valve V1 to ensure that only the NPL PRM gas was present in the system. To allow safe connection of the nozzle to the FCEV, the pressure in the assembly was first reduced below 3 bar. Before connecting the NPL contamination rig to the FCEV, valve V3 was closed to isolate the contamination rig and the nozzle–FCEV sections.

Once the contamination assembly was attached to the FCEV, valve V1 was opened to pressurize the system. The cylinder mass was then recorded from the precision balance. At this point, it was possible to open valve V4 to read the pressure of the NPL contamination rig. Valve V3 was opened to transfer 8–20 mg from the NPL PRM into the FCEV tank. At the end of the transfer, valve V3 and V1 were closed. The mass of cylinder 1 was then recorded. After the mass recording, valve V2 was opened to transfer approximately 10 mg of pure hydrogen into the FCEV tank to ensure transfer of any NPL PRM gas remaining in the gas flow path into the FCEV tank. Valve V2 was then closed. The contamination system was depressurized through valve 4 and 5. After depressurizing, the nozzle could be disconnected from the FCEV. The FCEV was powered again to record the temperature and pressure readings from the sensors of the FCEV tanks. The FCEV was then refilled at a nearby HRS with at least 1000 g of hydrogen fuel. If driving was required to reach the nearby HRS, it was carried out in EV driving mode (using only battery power) to avoid any consumption of hydrogen from the FCEV tank. This prevented contamination of the FCEV before the dilution made by the hydrogen from the HRS.

2.3. Gas Sampling

The gas sampling system was designed following the approach of Bacquart et al. [14] except with several adaptations (schematic presented in Supplementary Materials). The pressure regulator was replaced by a Sulfinert pressure relief valve set at 100 bar. The system had been entirely Sulfinert-treated from C1 to the sampling cylinder. For each experiment, two samplings were carried out consecutively using two different gas cylinders and slightly different FCEV tank pressures. The two samplings were obtained from the same gas sampling line. The sampling protocol applied in this study was described by Bacquart et al. [14].

2.4. Gas Analysis

The National Physical Laboratory (UK) is the UK's National Metrology Institute. The analyses were performed by NPL's Hydrogen Laboratory using NPL internal methods that were ISO 17025 [16] accredited for N₂, O₂, Ar, CO, total sulfur, and H₂O. Analyses were calibrated using NPL PRMs in hydrogen matrix gas. Oxygen and argon were analyzed by gas chromatography (GC) (Agilent, Cheadle, UK) with pulsed discharge helium ionization detector (PDHID) (VICI, CH) using helium as a high-purity carrier gas (Helium BIP®, Air products, Crewe, UK). The GC/PDHID sampling loop was 1 mL. The sample was transferred onto a capillary column molsieve 5A plot (30 m × 0.53 mm × 50 µm) and a second capillary column molsieve 5A plot (50 m × 0.53 mm × 50 µm). The GC oven was set at 30 degrees Celsius. Nitrogen was analyzed by GC with thermal conductivity detector (TCD) (Agilent Technologies, Cheadle, UK). The method used a Haysep Q 80/100 mesh 2 m × 1/8" outer diameter × 2.0 mm inner diameter column and a molsieve 5A 80/100 mesh 9 ft × 1/8" outer diameter × 2 mm inner diameter column with helium carrier. The loop size used for sample injection was 2 mL. Carbon monoxide, carbon dioxide, and non-methane hydrocarbons were measured using a GC (Peak Laboratories, View, CA, USA) coupled with a methanizer and a flame ionization detector (FID). The method used a Haysep D column (186" × 1.5") with nitrogen carrier. The GC column was set at 65 degrees Celsius. The loop size used for sample injection was 5 mL. Sulfur measurement was performed on an Agilent 7890 GC equipped with sulfur chemiluminescence detector

(SCD) (SCD355, Agilent, UK). The GC contained a 5 mL Silconert®-treated sample loop connected to a capillary column HP-1 (30 m × 0.320 mm ID × 0.251 µm film thickness, Agilent, Santa Clara, CA, USA). The GC oven was set at 30 degrees Celsius. Water was measured using a quartz crystal microbalance, QMA401 (Michell, Cambridge, UK). Gases were sampled directly from the gas cylinder to the analyzer, and the pressure was adjusted to 1 bar gauge for the analysis. NPL gravimetric gas standards (NPL, Teddington, UK) and/or dynamic standards were used to generate calibration curves covering both the EN 17124 and ISO 14687 amount fraction thresholds and the measured values from the samples. The dynamic standards were prepared by dilution of NPL PRM (NPL, UK) with high-purity hydrogen (BIP+ quality, Air Products, UK) using calibrated mass flow controller systems (Bronkhorst, Veenendaal, The Netherlands). All of the data were examined so that no results were discarded without a valid technical reason. The calibration curve, results of analysis, and associated uncertainties were determined using NPL software XLGENline version 2 [17]. An expanded uncertainty using a k value of 2 was used for all results. In some cases, a more conservative uncertainty was derived from analytical expert knowledge.

2.5. Gravimetric Determination of Hydrogen Fuel Composition in FCEV

The final composition in the FCEV was calculated using the software package Grav-Calcul2 [17]. The software input required the mass of gas and gas composition for each scenario. Four scenarios were calculated: (1) the original hydrogen fuel in the FCEV tank; (2) the hydrogen fuel added at HRS; (3) the NPL PRM cylinder; and (4) the pure hydrogen cylinder.

The gas compositions were obtained from NPL for the NPL PRM and pure hydrogen cylinder. For the original hydrogen fuel in the FCEV tank and for the hydrogen fuel added at the HRS, they were determined by analytical measurement performed by NPL from samples. For the mass of gases, the values were obtained directly by weighing on mass balances for the NPL PRM and pure hydrogen cylinder and from the determination of mass based on the non-ideal gas law for the original hydrogen fuel in the FCEV tank and for the hydrogen fuel added at the HRS.

The non-ideal gas law equation used to calculate the total mass of hydrogen (m_{H_2}) in grams was as follows:

$$m_{H_2} = \frac{P \times V \times M_{H_2}}{z \times R \times T}$$

where P is the average pressure in both tanks [Pa]; V is the total tank volume [m^3], equal to 0.123 m^3 ; M_{H_2} is the molar mass dihydrogen [g/mol], equal to 2.016 g/mol; R is the universal gas constant [$m^3 \cdot Pa \cdot K^{-1} \cdot mol^{-1}$]; T is the temperature average of both FCEV tanks [K]; and z is the compression factor [-] using the equation in [18].

2.6. Vehicle Evaluation

The contaminated vehicle underwent a driving evaluation to assess the impact on the FC performance. The vehicle was driven in full electric mode to a non-public circuit close to the location where the contamination process took place to avoid any contamination impact on the FC beforehand. Next, the vehicle was driven according to a controlled driving pattern. This driving pattern was designed to fully cover the potential and current output of the FC, so that a full polarization curve could be extracted. The driving pattern consisted of fast accelerations up to 80 km/h. The total driving test took up to 30 min and approximately 0.77 kg of hydrogen from the tank was consumed during each test. It must be noted that the exposure of the contaminated hydrogen fuel to the FCEV during this testing was for very short periods, while the ISO 14687:2019 and EN 17124:2022 [9] amount fraction thresholds were originally set based on extensive use to maintain expected lifetime of the vehicle. Such longer durability testing is outside the scope of this activity; instead, the focus here was to assess impact on immediate FC performance. FC controlled area network (CAN) information was recorded during driving and extracted from the vehicle utilizing a neoVI ION Wireless Data Logger (Intrepid, Dallas, TX, USA). The data sampling

rate was 10 Hz. Post-processing of the CAN data was conducted in order to extract vehicle impact plots such as the FC's polarization curve. The obtained datapoints (current and voltage pairs) were normalized using min-max feature scaling. As a result, all values of FC current and voltage were rescaled to be in the [0, 1] range (dimensionless):

$$X' = \frac{X - X_{min}}{X_{max} - X_{min}}$$

3. Results

A summary schematic of the experiment performed is presented in Figure 2; this schematic highlights all the steps of the experiments (the contamination of the FCEV tank, the addition of hydrogen fuel from the HRS, the sampling from the FCEV, and then the start of the next contamination).

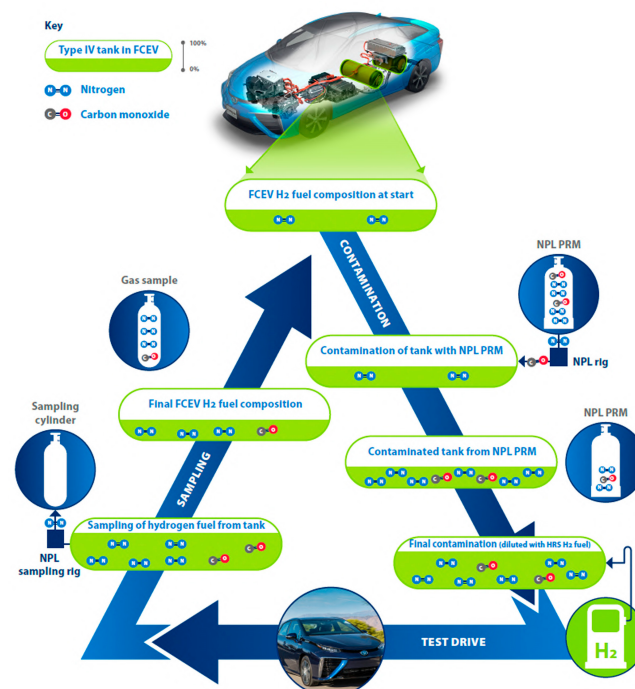


Figure 2. Schematic of the experiment performed following the contamination of the FCEV tank, the addition of hydrogen fuel from the HRS, the sampling from the FCEV, and then the start of the next contamination.

3.1. Accurate Determination of FCEV Tank Composition by Gravimetry (Including Associated Measurement Temperature, Pressure, and Volume)

The gravimetric determination of the hydrogen fuel composition in the FCEV tank was based on four inputs and two parameters: the mass and the chemical composition. The four inputs were (1) the original hydrogen fuel in the FCEV tank; (2) the hydrogen fuel added at the hydrogen refueling station; (3) the NPL PRM cylinder; and (4) the pure hydrogen cylinder. Each input is discussed individually in the following sections.

3.1.1. Input for the Pure Hydrogen Cylinder

The gas composition of the pure hydrogen cylinder (hydrogen BIP+, Air Products, UK) was known and is defined in the Materials and Methods section. The mass transferred was small (less than 10 g) and, therefore, was considered a negligible mass compared to the other hydrogen fuel transferred by the HRS (over 1000 g). The input related to the pure hydrogen was considered negligible (less than 1% of the largest mass added) and contained no significant chemical compounds other than hydrogen.

3.1.2. Input for the NPL PRM Cylinder

The NPL PRM cylinder composition was known and is defined in the Materials and Methods section. The NPL PRMs were traceable PRMs with accurate chemical compound amount fractions and small uncertainties. Therefore, the uncertainties related to these inputs were considered low in the determination of the FCEV composition.

The mass transferred was based on the difference in mass of the NPL PRM cylinder before and after transfer into the FCEV tank. The accurate determination of the mass transferred could be realized directly using a precision balance or through indirect measurement of pressure and temperature of the gas (assuming the volume of the container). The mass determination using precision balance was complex due to the actual mass transferred (approximately 10 to 20 g) in comparison with the cylinder mass (17 kg). The experimental environment restricted the use of high-accuracy balance at the gram level. Secondly, the workshop environment was not optimized for accurate weighing (i.e., constant ventilation for safety reasons). The experimental conditions induced a higher uncertainty on the weighing than expected (relative uncertainty above 1%). However, the use of the precision balance did allow onsite calibration, traceability of the measurements, and direct mass determination.

3.1.3. Input for the Hydrogen Fuel Added at the Hydrogen Refueling Station

The chemical composition of the hydrogen fuel from the HRS needed to be determined, as it could not be assumed or extrapolated due to the possible variation in the hydrogen fuel quality delivered by the HRS [19,20]. Any compounds present in the HRS hydrogen fuel would contribute to the final hydrogen fuel composition in the FCEV tank. Only nitrogen and argon were quantified above the limit of detection. The results are presented in Supplementary Materials. Sampling of the hydrogen fuel from the HRS was realized during the second set of experiments, following the methodology described in the Gas Sampling section [21]. The measurements were performed by NPL's hydrogen quality laboratory (Teddington, UK) as described in the Materials and Methods section.

The mass of the hydrogen fuel transferred is critical, as it represents a significant mass in the final determination of the FCEV tank hydrogen fuel composition. The mass was recorded from the HRS flow meter reading and determined by the pressure and temperature sensors from the FCEV tanks. The comparison of the two approaches presented in Supplementary Materials showed that the HRS metering was approximately 6–11% lower than the mass determined by the FCEV sensors. However, the uncertainty of the HRS metering was considered conservative applying the OIML recommendation (maximum permissible error of 5%) [22] and the uncertainties from the FCEV sensors (evaluated to 5%), and, as a result, the values were not significantly different at a confidence level of 95%. The mass determined directly from the FCEV tanks' measurement data was considered more accurate, as it was determined directly from the FCEV tank in which the gas composition was realized.

3.1.4. Input for the Hydrogen Fuel in the FCEV Tank

The hydrogen fuel in the FCEV tank could not be neglected, as the FCEV would always contain a significant amount of hydrogen (between 3 and 5.4 MPa). The starting mass of hydrogen fuel in the tank could only be assessed by indirect measurement of pressure and temperature in the FCEV tanks. The sensors used for this study were the sensors integrated in the FCEV system (part of the commercial FCEV).

The final hydrogen fuel composition would be dependent on the quality of the hydrogen fuel from the previous refueling or experiment within this study (see Figure 2). Before the first experiment, the FCEV was entirely fueled at the HRS. The hydrogen fuel composition in the FCEV tank for the first experiment was assumed to be of similar composition as the hydrogen fuel of the HRS. A more accurate determination would have involved sampling and analysis of the hydrogen fuel from the FCEV tank.

For the subsequent experiments, the hydrogen fuel starting in the FCEV tank was assumed to be the composition of the previous experiment, as presented in Figure 2.

Once all these inputs were determined as described, it was then possible to accurately determine the amount fraction of impurities in the FCEV tanks. A summary of the contamination experiments and determination of the mass of all of the hydrogen gas inputs were provided in Supplementary Materials. The objective of generating a controlled contaminated fuel in the FCEV without complex hardware was achieved; however, to confirm the accuracy of this approach, it was necessary to perform a comparison of the determined amount fractions of impurities between the gravimetric synthetic hydrogen fuel in the FCEV and the gas sampled from the FCEV.

3.2. Experimental Validation of the Synthetic Hydrogen Fuel from FCEV Tank Approach

The FCEV hydrogen fuel was sampled as described in the Materials and Methods section. Two independent samplings were carried out for each experiment. The results of analysis were compared to the expected composition of the hydrogen fuel in the FCEV based on the calculation detailed in section “Gravimetric determination of hydrogen fuel composition in FCEV”. The detailed results of analysis from NPL’s hydrogen quality laboratory (Teddington, UK) are presented in Table 2.

Table 2. Summary of analytical results from the different FCEV contamination experiments. The uncertainty reported corresponds to $k = 1$. Two analyses were repeated and compared to the gravimetric value.

	CO μmol/mol	N ₂ μmol/mol	H ₂ S nmol/mol	H ₂ O μmol/mol	O ₂ μmol/mol	Ar μmol/mol
ISO14687:2019 limit	0.2	300	4	5	5	300
Gravimetric value	0.138 ± 0.008	90.12 ± 3.7	<1	0.91 ± 0.09	<0.10	1.11 ± 0.021
Experiment 1	0.161 ± 0.010	106 ± 1.0	n.a.	9.8 ± 1.0	<0.5	0.89 ± 0.10
Experiment 1	0.159 ± 0.010	105 ± 0.7	n.a.	9.7 ± 1.0	<0.5	0.86 ± 0.10
Gravimetric value	0.536 ± 0.035	91.94 ± 4.5	<1	0.91 ± 0.10	<0.10	1.11 ± 0.022
Experiment 2	0.586 ± 0.010	101.9 ± 1.1	n.a.	9.4 ± 0.9	1.30 ± 0.13	1.03 ± 0.10
Experiment 2	0.565 ± 0.005	99.9 ± 0.4	n.a.	6.8 ± 0.7	<0.5	1.05 ± 0.10
Gravimetric value	4.99 ± 0.31	147 ± 8	<1	0.91 ± 0.09	<0.10	1.10 ± 0.020
Experiment 3	4.737 ± 0.023	146.4 ± 0.9	n.a.	10.4 ± 1.0	0.84 ± 0.10	1.10 ± 0.10
Experiment 3	4.856 ± 0.023	147.8 ± 0.5	n.a.	11.0 ± 1.1	0.82 ± 0.10	1.12 ± 0.10
Gravimetric value	1.08 ± 0.09	231 ± 10	11.05 ± 0.54	0.90 ± 0.10	<0.10	1.10 ± 0.022
Experiment 4	1.104 ± 0.008	216.7 ± 3.7	4.1 ± 0.7	10.3 ± 1.0	0.55 ± 0.10	1.02 ± 0.1
Experiment 4	1.061 ± 0.006	210.0 ± 3.5	4.97 ± 0.49	12.6 ± 1.3	1.85 ± 0.15	1.02 ± 0.1

3.2.1. Inert and Low-Reactivity Compound Behavior

The first three experiments focused on inert and low reactivity gas (nitrogen and carbon monoxide, respectively). Figure 3 presents the results of nitrogen and carbon monoxide amount fraction from direct sampling and analysis compared to the gravimetrically determined amount fractions. Considering the measurement uncertainty, no significant difference at a confidence level of 95% was observed between the gravimetric composition and the measured amount fraction of CO and nitrogen for the experiments except for the nitrogen amount fraction in experiment 1. For experiments 2 and 3, the agreement between the analytical results of the sampling and the synthetic hydrogen fuel used in the FCEV tank demonstrated that it was possible to produce synthetic contaminated hydrogen fuel directly in the FCEV with accurate control of the impurity levels.

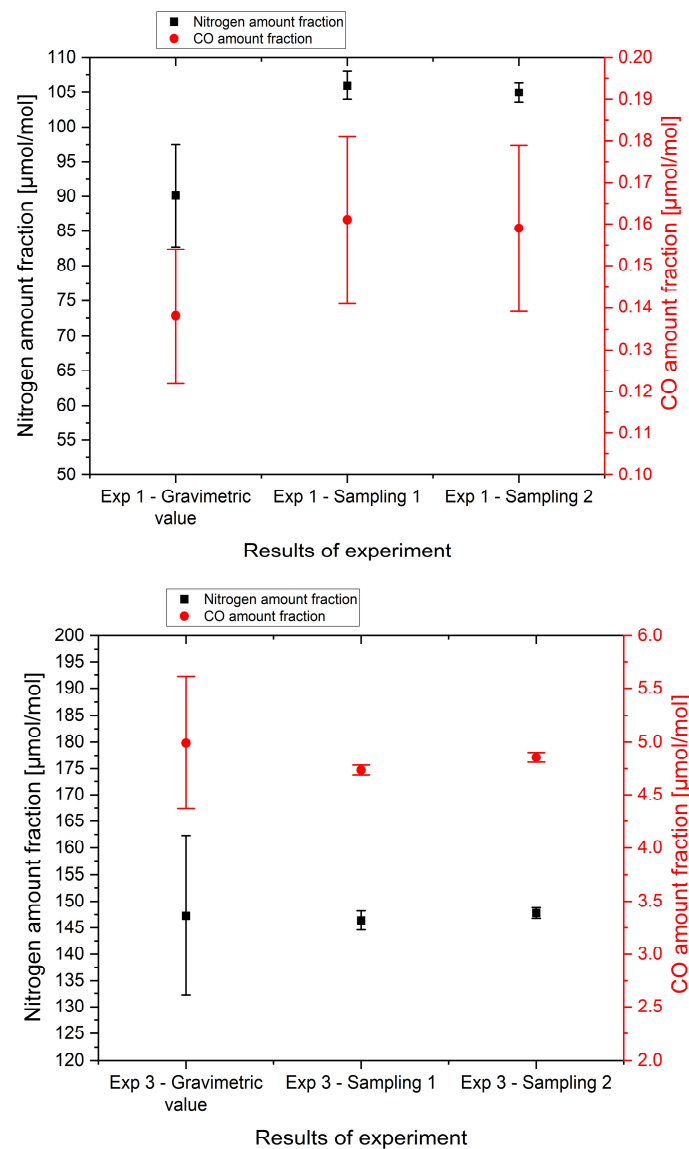


Figure 3. Results of 1st and 3rd synthetic hydrogen fuel made in the tank of the FCEV. The figure presents the gravimetric determination of CO and nitrogen in the hydrogen fuel in the FCEV tank compared to the results of analysis from the two samples taken from the FCEV. The expanded uncertainties are presented ($k = 2$).

Experiment 1 indicated a relative difference of approximately 15% between the expected composition of the hydrogen fuel in the FCEV and the measured composition (from both samples) for N_2 and CO. Even if the difference was not statistically significant for CO due to a larger measurement uncertainty, the trend observed (15% increase) was coherent with the nitrogen observation. As the bias seemed consistent, a few potential explanations for the differences due to inaccuracies were considered: (1) determination of the mass transferred from NPL PRM into the FCEV, (2) the actual hydrogen fuel composition in the FCEV, (3) the hydrogen fuel composition from the HRS.

As detailed in the previous section, the actual mass of NPL PRM transferred from the gas cylinder may not have been accurately determined due to a lack of shielding from environmental disturbances or a slight bias in the weighing related to the pressure measurement which was performed in parallel. The gas composition could be another potential reason for the discrepancies. The composition of the hydrogen in the FCEV tank was not sampled before the first contamination; it was assumed to be similar to the hydrogen fuel composition from the HRS (measured during the second experiment). This

highlights how critical it is to determine the hydrogen fuel composition of the four inputs (hydrogen fuel in the FCEV, hydrogen fuel from HRS, NPL PRM, and pure hydrogen) before starting any contamination experiments. Even if the results of experiment 1 showed a small difference, the sample results were not far from those of the synthetic hydrogen fuel realized in the FCEV tank. This provides confidence in the methodologies used for this study and highlights the importance of each step.

The results from the two samplings obtained during each experiment agree within a 95% confidence level for carbon monoxide and nitrogen. This demonstrates that the sampling procedure was repeatable and that the gas mixture in the FCEV tank was homogenous.

In Figure 4, the CO and N₂ results from all of the experiments are presented together. This graph highlights the coherence of the results between the expected fuel composition and the measured values. All results except nitrogen in experiment 1 overlapped with the 0 line of the figure, which meant there was no significant difference between the measured values and the gravimetric amount fraction of the synthetic hydrogen fuel in the FCEV tank. This validates the approach proposed here to accurately contaminate the FCEV and to reliably sample hydrogen from the FCEV tank using the NPL sampling tool for inert and low-reactivity compounds.

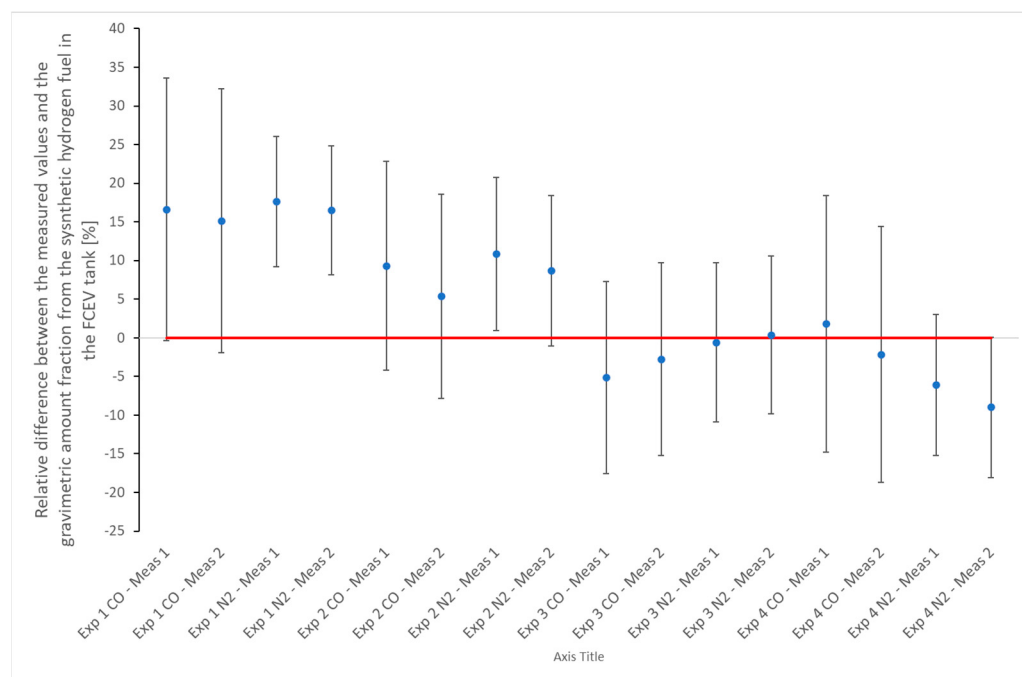


Figure 4. Relative difference between the CO and N₂ amount fraction in the hydrogen fuel sampled by analysis and the expected composition from the synthetic hydrogen fuel in the FCEV tank. The uncertainties reported correspond to the uncertainties of the differences combining the gravimetric uncertainties of the synthetic fuel and the measurement uncertainties with confidence levels of 95% ($k = 2$). The red line represents no difference between the measured value and the gravimetric value.

3.2.2. Reactive Compound Behavior

In the fourth experiment, the FCEV tanks were contaminated with a reactive compound (H₂S) in addition to CO and N₂ (results are shown in Figure 5).

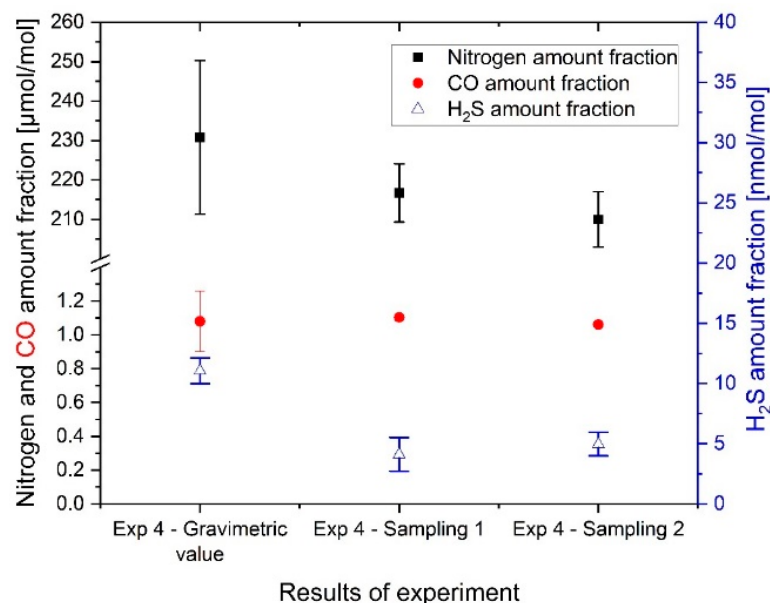


Figure 5. Results of 4th synthetic hydrogen fuel made in the tank of the FCEV. The figure presents the gravimetric determination of CO, H₂S, and nitrogen in the hydrogen fuel in the FCEV tank compared to the results of analysis from the two samples taken from the FCEV. The expanded uncertainties are presented ($k = 2$).

As detailed in the previous section, the amount fraction of CO and N₂ agreed well between the synthetic hydrogen fuel realized in FCEV tank and the results of analysis (no significant difference at a confidence level of 95%). It validated that the preparation of the synthetic hydrogen fuel in the FCEV tanks was determined accurately.

The results of H₂S amount fraction showed that the measured values from the two samplings agreed within a confidence level of 95% at 4–5 nmol/mol. This demonstrated that the nmol/mol level of H₂S in hydrogen fuel will remain in the gas phase of an FCEV tank (i.e., it does not adsorb on walls) and may reach and degrade the fuel cell. This is a significant finding, as previously, it was hypothesized that H₂S may be adsorbed out and, therefore, never reach the fuel cell.

However, the composition in the FCEV tank was expected to be around 11 nmol/mol; a difference of almost 6–7 nmol/mol of H₂S was observed between the gravimetric FCEV composition and the analysis of the samples. Several factors were identified as potential reasons for the loss of H₂S experienced: absorption along the FCEV gas flow path, time of contact (i.e., sampling realized after test drive), sampling representativity (i.e., delay between sampling and analysis of few weeks), or reactions between impurities within the FCEV tanks. The results obtained for CO and N₂ demonstrate that the preparation was realized correctly, which excludes this as a possible cause of the discrepancies. H₂S is known to react and adsorb onto metallic materials [23]. The FCEV system was not built with passivated materials; therefore, adsorption of reactive impurities along the FCEV gas flow path (i.e., valve, pipes, regulators) could have reduced the amount of sulfur in the samples obtained.

Further experiments are needed to better understand how the FCEV gas flow path interacts with reactive compounds such as H₂S (i.e., proportional loss or saturation amount fraction at which the tank material cannot adsorb additional H₂S). The behavior of H₂S on type IV FCEV tanks with a liner was previously unknown. This study shows that this nmol/mol level of H₂S does not completely adsorb onto this type of liner; therefore, it essentially does not act as a defensive measure. To improve the accuracy of this experiment, one would need to investigate the behavior of H₂S with type IV FCEV tanks; the experiments would be significantly improved if adsorption in this part of the FCEV system could be reduced.

The sampling representativity was important to exclude as a potential explanation of the discrepancy observed. Similar to the FCEV gas path, adsorption of the H_2S could have occurred when hydrogen was in contact with the sampling rig, on the contamination rig, or within the sampling vessels. As the analysis from the two samplings agreed, it could be assumed that the sampling vessel type was not the main source of H_2S loss. Even if the sampling hardware was fully Sulfinert-treated and provided representative samples for contaminants such as CO and nitrogen, it may be possible that it is not sufficient to ensure 100% of H_2S recovery in the gas phase at low nmol/mol.

The contamination hardware that was used to transfer gas from the NPL PRM into the FCEV tank could have been an adsorption source, as it is mainly made of metallic materials. For this part of the system, all of the pipes, connections, and valves, except for the nozzle, were Sulfinert-treated, which would limit any potential H_2S adsorption. As it was therefore possible that H_2S adsorption may have happened between the nozzle and the receptacle, the use of a dedicated Sulfinert nozzle in the future would help to further reduce potential adsorption within this section of the system.

Even after recognizing these challenges in the experimental approach, the study demonstrated that it is possible to contaminate hydrogen fuel in the FCEV with 5–10 nmol/mol of H_2S . It is important to continue the study with a simpler system to determine at which step of the process the loss of H_2S occurred.

3.2.3. Identification of Other Compounds

During the experiments, no oxygen, argon, or water were deliberately added into the hydrogen fuel. Therefore, these compounds should only have been present due to their original presence in the hydrogen fuel in the FCEV or from the hydrogen fuel at the HRS. These compounds were measured to evaluate whether air contamination had occurred during the experiments. The oxygen amount fraction was always below the limit of detection or close to 1 $\mu\text{mol/mol}$. Even if the oxygen amount fraction values were very low, the fluctuation in this value between samples suggested the potential presence of a very small air leak during sampling. As the air leak was considered very small, it would not significantly affect the overall sample representativity even if a better control on this connection would be recommended in the future. Argon may be a good tracer to confirm whether an air leak was identified using oxygen. However, the argon/oxygen ratio was close to one, which was different from the argon/oxygen ratio expected to be found in air (approximately 1/22). The argon amount fraction remained constant over the different experiments, indicating that the hydrogen fuel from the HRS was the only source of argon. This hypothesis was confirmed by the good agreement between the measured argon fraction and the actual gravimetric hydrogen fuel composition determined using the same composition of the hydrogen fuel from the HRS.

A significant amount fraction of water was observed at similar levels across all experiments. This presence was surprising, as no water was introduced by any of the inputs; this was confirmed by the results of purity analysis. Therefore, the water amount fraction was expected to have decreased during the experiment through dilution with purer hydrogen. There were two potential sources of water: (1) labile water present in the FCEV tank adsorbed on the cylinder wall (equilibrating with the gas phase) or (2) measurement issues (sampling bias or measurement bias). An implementation of online water analysis during the sampling may provide new evidence to support this investigation. This unexpected outcome highlighted the need for further investigation into the potential source of water amount fraction in the hydrogen fuel sampled from the FCEV.

3.3. Impact of Contamination on FCEV Performance

In Figure 6, the polarization curves obtained from the FCEV test drive following the controlled contamination are summarized. On each plot, the trend line of the original polarization curve without contamination is visualized. It is clear that the current–voltage data-points are not shifted significantly to lower voltage levels compared to the original trend

line during experiment 1 and 2 (measured CO contamination of 0.161 and 0.586 $\mu\text{mol/mol}$, respectively; ISO14687:2019 CO limit is 0.2 $\mu\text{mol/mol}$). In experiment 3 and 4, however, the shift of the datapoints to lower voltages becomes clearly visible (experiment 3: measured CO contamination of 4.7 $\mu\text{mol/mol}$; experiment 4: 1.1 $\mu\text{mol/mol}$ CO and 4.1 nmol/mol H_2S). ISO 14687:2019 has a CO limit of 0.2 $\mu\text{mol/mol}$ and a H_2S limit of 4 nmol/mol.

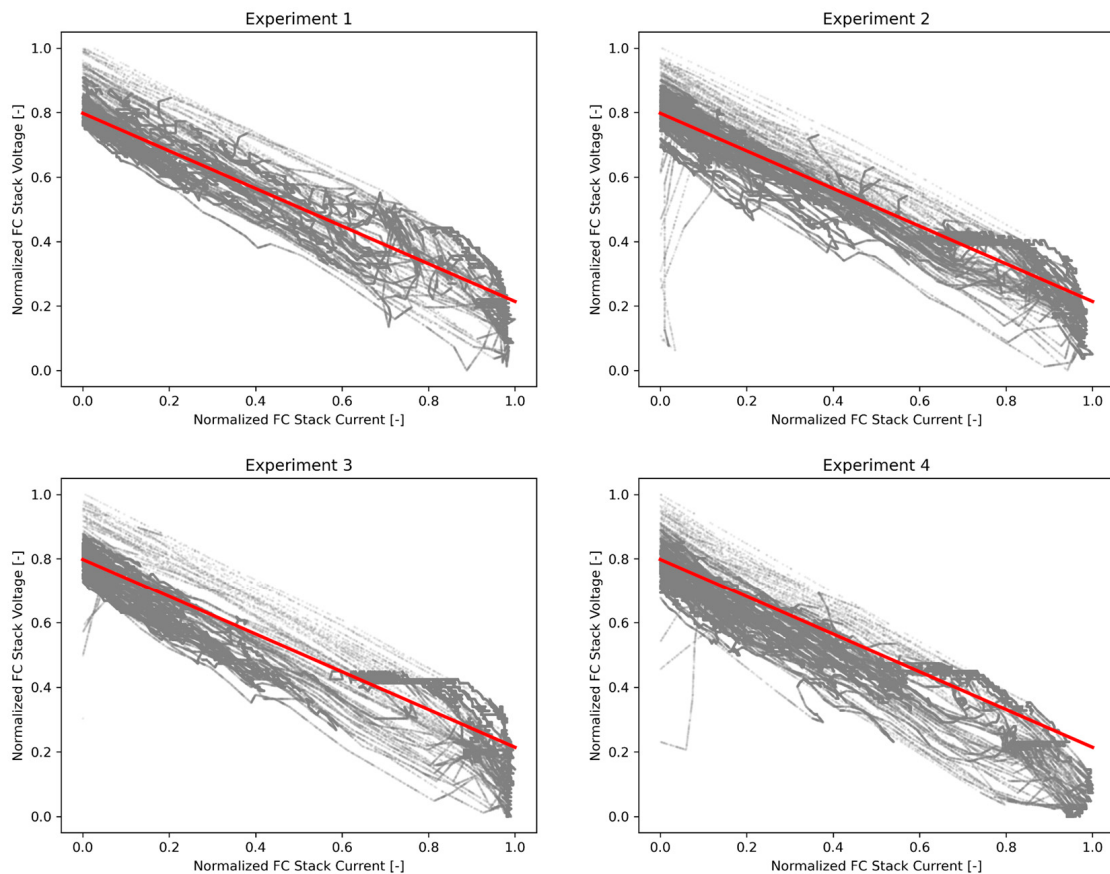


Figure 6. Impact of FCEV contamination on the polarization curve of the FC. Current and voltage have been normalized by min-max feature scaling. The trend line is obtained from the current–voltage plot of the FCEV’s test drive before contamination ($R^2 = 0.92$). Next, the trend line is plotted on all 4 current–voltage plots obtained from test drives after the contamination took place. Relative comparison of the datapoints to the original trend line allows the visualization of the immediate impact on the FC’s performance.

In order to quantify these findings, a data slice ranging from 0.4 to 0.6 normalized FC current (centre of the FCEV’s current range) was obtained, and the histogram of the voltages in this current range was obtained (see Figure 7). Separate figures were made for experiment 1 to 3 (CO contamination) and 1 and 4 (CO and H_2S contamination). The mean normalized voltages in this range were 0.50, 0.48, 0.43, and 0.45 for experiments 1 to 4, respectively. The data obtained after experiment 1 were taken as a reference for visualization purposes, in order not to overload the graph, as the FC voltages did not drop during this experiment. Although not apparent in the polarization curve, a drop in potential could also be observed for experiment 2 in the histograms.

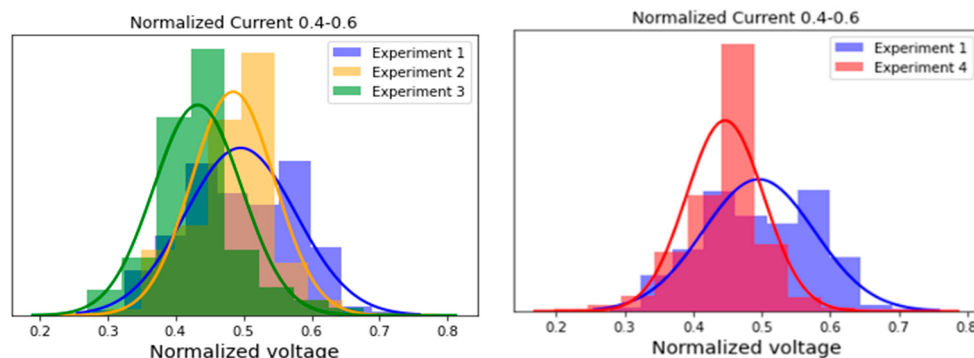


Figure 7. Histogram of the normalized FC voltages in the normalized current range 0.4–0.6: **(left)** CO contamination experiments 1 to 3 and **(right)** H₂S contamination experiment 4. For visibility reasons, experiment 1 is used as a reference, as the polarization curve was not impacted by the contamination of experiment 1.

This shows that even a slight exceedance of the ISO 14687:2019 CO threshold has an immediate effect on the FC performance, even from a fairly low exposure to contaminants during the short test drive. The voltage drop trend continues to experiment 3, where a CO contamination level of 4.7 $\mu\text{mol/mol}$ was achieved.

For H₂S contamination in experiment 4, the interpretation of the vehicle test data is more difficult because the tests were performed back to back. No cleaning of the FC stack and/or tanks was conducted, and the stack did not undergo any recovery procedure in between the experiments. Therefore, the effect of catalyst poisoning contaminants (such as CO) of previous experiments is very likely to be observed in the following contamination experiment. For this reason, the concentration of CO was increased stepwise from experiments 1 to 3. For experiment 4, however, the CO in the tank was diluted from 4.7 to 1.1 $\mu\text{mol/mol}$ while 4.1 nmol/mol H₂S contamination was achieved, which resulted in the partial recovery of the normalized voltage from 0.43 to 0.45 going from experiment 3 to 4. Although the adsorption intensity and the poisoning effect of H₂S is higher than that of CO, the higher concentration of CO inhibited the adsorption of H₂S (competitive adsorption) [24]. As a result, the CO and H₂S influence cannot be decoupled, as no specific H₂S poisoning experiment was performed.

The compounds' amount fraction thresholds specified in ISO 14687:2019 and EN 17124:2022 have been defined by taking the useful vehicle life into account: the durability of FCEVs should not be affected by exposure to contamination in line with the standards. Although the discussed experiments do not contain evaluation of the durability of the FC, they clearly demonstrate that contamination of CO at ISO 14687 threshold levels does not appear to have an immediate effect on FCEV performance, while even slight exceedances of these thresholds immediately cause a voltage drop in the FC, even for short exposure times.

3.4. Perspective for FCEVs

The studies presented in this paper provide several interesting results for the future of FCEVs. Firstly, the studies verify that the novel sampling system for FCEV sampling proposed in this paper has been metrologically validated when targeting compounds of interest. The methodology applied did not require extensive infrastructure and is expected to have limited or controlled impact on the actual FCEV compared to other possible approaches. After contamination of the FCEV, it was possible to vent the FCEV tank without using the fuel cell. A proper FCEV clean-up process may, however, need to be adapted based on the selected compound and amount fraction level.

Secondly, the study allows direct testing of FCEV performance using simulated hydrogen fuel. This is an important validation step for the study of contaminants' impact at the FCEV level for both instant impact as well as long-term durability effects. Compared to state-of-the-art single-cell or stack level testing, FCEV testing provides a dataset for

real-life conditions adding the complexity of the FCEV's gas flow path and a more direct experience of the impact from contaminants, including how the end user or customer would experience this. For future evaluation of FCEVs, special attention should be paid to avoiding cross-effects of different contaminations, such as in experiment 4 in this study. Countermeasures could consist of flushing of the tanks and fuel lines when switching to other species or less concentrated contamination. Additionally, an FC regeneration protocol should also be considered to revert the state of the FC as close as possible to the original condition. The effect of exposure time should be considered as well, as longer test drives could provide more insight into contaminant behavior over time.

The results of the contamination studies from this paper using low level H_2S are extremely interesting for industry. This study demonstrates, for the first time, that sulfur compounds present in hydrogen fuel may be transferred into an FCEV's tank at nmol/mol (without, for example, being lost through adsorption upstream of the fuel cell). Despite the loss observed between the expected amount fraction and the measured amount fraction, low nmol/mol sulfur may still reach the fuel cell system. This result emphasizes the importance of monitoring sulfur compounds, as FCEV's gas flow path may not act as a significant barrier for removing impurities from low-quality hydrogen fuel.

4. Conclusions

The study presented in this paper has demonstrated that it is possible to produce a synthetic hydrogen fuel in an FCEV tank without complicated hardware (e.g., an HRS with contaminated fuel) or without involving an extensive number of gas cylinders. The experiment was repeated four times with three contaminants (CO , N_2 , and H_2S); each repeat showed good agreement between the gravimetric amount fraction of the synthetic hydrogen fuel realized in the FCEV tank and the measured value (within a confidence level of 95%). One study disagreed slightly; however, the bias in that case was small. The results of this study validated the methodology used to accurately contaminate an FCEV with inert and low-reactivity compounds. The study investigated the different inputs for the process and highlighted several possibilities to determine the mass and the importance of the different gas composition inputs.

The experiment involving a reactive compound (H_2S) presented several challenges but a positive outcome. Firstly, it demonstrated that it is possible to contaminate an FCEV with low nmol/mol sulfur within a complex system. However, it highlighted the complexity related to reactive compounds, as the measured value in the two samples differed significantly from the expected composition. Due to the complexity of the system, it is difficult to determine if this complexity relates to the contamination (adsorption of the reactive compounds on the nozzle/receptacles or in the FCEV tanks) or to adsorption in other parts of system. This investigation could be performed using a simpler system, involving only a tank, and then progressively adding in the other components of the FCEV gradually until the loss of impurities is achieved.

For the first time, a traceable H_2 contamination impact study on an FCEV was performed. The impact of CO and H_2S around ISO 14687:2019 threshold amount fraction levels on the FCEV's performance were demonstrated. Small exceedances above the threshold limits show a significant impact, even with relatively short exposure times.

The methodology proposed in this article can then be deployed to evaluate a large variety of hydrogen fuel compositions. It would allow performance testing of sampling methodologies and/or FCEVs using accurate and traceable hydrogen fuel composition and under real conditions. This will allow new studies to be carried out which closely simulate real-life conditions, thereby permitting new challenges to be identified (e.g., the impact of reactive compounds) and roadblocks (e.g., the validation of the sampling protocol) to be removed. When applying this approach to FCEV durability studies, key input could be provided to H_2 standardization bodies such as ISO technical committee 197.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/en17071510/s1>, Table S1. Hydrogen fuel quality from the hydrogen refuelling station used to fill the FCEV. The analysis focussed only on the compounds that influenced the final composition (carbon monoxide, nitrogen, and sulphur). Additional compounds were measured (water, oxygen, and argon) to evaluate potential air contamination in the experimental setup. Figure S1. NPL sampling rig modified from Bacquart et al. [14] including sulfinert passivated flow path from C1 to the cylinder. Table S2. Evaluation of hydrogen mass in the FCEV based on pressure measurement from the venting tool and from the FCEV system including pressure and temperature sensors. Table S3. Summary of the masses of hydrogen fuel inputs in the FCEV with the associated uncertainties. The masses were determined for each contamination experiments. Figure S2. Results of 2nd synthetic hydrogen fuel made in the tank of the FCEV. The figure presents the gravimetric determination of CO and nitrogen in the hydrogen fuel in the FCEV tank compared to the results of analysis from the two samples taken from the FCEV. The expanded uncertainties are presented ($k = 2$).

Author Contributions: Conceptualization, T.B., W.S. and V.M.; methodology, T.B., W.S., N.M. and V.M.; formal analysis, T.B., N.M., A.S.O.M., M.H., W.S., J.O. and V.M.; investigation, T.B., W.S., J.O. and V.M.; data curation, T.B., W.S., N.M., J.O., A.S.O.M., A.M. and V.M.; writing—original draft preparation, T.B.; writing—review and editing, T.B., W.S., N.M., J.O., A.S.O.M., M.H., A.M. and V.M.; supervision, T.B., W.S., A.M. and V.M.; project administration, T.B.; funding acquisition, T.B. and W.S. All authors have read and agreed to the published version of the manuscript.

Funding: This research was co-funded by the European Union’s Horizon 2020 research and innovation program and the European Metrology Programme for Innovation and Research (EMPIR) Participating States, grant number «19ENG04—Metrology for hydrogen vehicles 2».

Data Availability Statement: The original contributions presented in the study are included in the article/Supplementary Materials; further inquiries can be directed to the corresponding author/s.

Conflicts of Interest: Authors Ward Storms, James Olden and Vincent Mattelaer were employed by the company Toyota Motor Europe. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

References

1. European Environment Agency. *Transport and Environment Report 2021—Decarbonising Road Transport—The Role of Vehicles, Fuels and Transport Demand*; Publications Office of the European Union: Luxembourg, 2022. [CrossRef]
2. EPA. Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2021. U.S. Environmental Protection Agency, EPA 430-R-23-002. Available online: <https://www.epa.gov/ghgemissions/inventory-us-greenhouse-gas-emissions-and-sinks-1990-2021> (accessed on 20 September 2023).
3. Available online: <https://acceleratingtozero.org/the-declaration/> (accessed on 20 September 2023).
4. Council of the European Union. European Parliament, Regulation (EU) 2023/851 of the European Parliament and of the Council of 19 April 2023 amending Regulation (EU) 2019/631 as regards strengthening the CO₂ emission performance standards for new passenger cars and new light commercial vehicles in line with the Union’s increased climate ambition (Text with EEA relevance), PE/66/2022/REV/1. *Off. J. Eur. Union* **2023**, *110*, 5–20.
5. IEA. Global EV Outlook 2023, IEA, Paris. 2023. Available online: <https://www.iea.org/reports/global-ev-outlook-2023> (accessed on 20 September 2023).
6. Samsun, R.C.; Rex, M.; Antoni, L.; Stolten, D. Deployment of Fuel Cell Vehicles and Hydrogen Refueling Station Infrastructure: A Global Overview and Perspectives. *Energies* **2022**, *15*, 4975. [CrossRef]
7. Nguyen, H.L.; Han, J.; Nguyen, X.L.; Yu, S.; Goo, Y.-M.; Le, D.D. Review of the Durability of Polymer Electrolyte Membrane Fuel Cell in Long-Term Operation: Main influencing Parameters and Testing Protocols. *Energies* **2021**, *14*, 4048. [CrossRef]
8. ISO 14687; Hydrogen Fuel Quality—Product Specification. International Organization for Standardization: Geneva, Switzerland, 2019.
9. EN 17124:2022; Hydrogen Fuel. Product Specification and Quality Assurance. Proton Exchange Membrane (PEM) Fuel Cell Applications for Road Vehicles. European Committee on Standardisation: Bruxelles, Belgium, 2022.
10. J2719_202003; Hydrogen Fuel Quality for Fuel Cell Vehicles. SAE International: Warrendale, PA, USA, 2020.
11. Zhang, Q.; Harms, C.; Mitzel, J.; Gazdzicki, P.; Friedrich, K.A. The challenges in reliable determination of degradation rates and lifetime in polymer electrolyte membrane fuel cells. *Curr. Opin. Electrochem.* **2022**, *31*, 100863. [CrossRef]
12. Murugan, A.; de Huu, M.; Bacquart, T.; van Wijk, J.; Arrhenius, K.; te Ronde, I.; Hemfrey, D. Measurement challenges for hydrogen vehicles. *Int. J. Hydrogen Energy* **2019**, *44*, 19326–19333. [CrossRef]

13. ISO 6142-1; Gas analysis—Preparation of Calibration Gas Mixtures—Gravimetric Method for Class I Mixtures. International Organization for Standardization: Geneva, Switzerland, 2015.
14. Bacquart, T.; Moore, N.; Mattelaer, V.; Olden, J.; Morris, A.S.O.; Storms, W.; Murugan, A. First Hydrogen Fuel Sampling from a Fuel Cell Hydrogen Electrical Vehicle—Validation of Hydrogen Fuel Sampling System to Investigate FCEV Performance. *Processes* **2022**, *10*, 1709. [[CrossRef](#)]
15. Brown, A.S. *GravCalc2 (Version 2.3.0) User Guide*, 2009. NPL Report AS 42; National Physical Laboratory: Teddington, UK, 2009.
16. ISO 17025; General Requirements for the Competence of Testing and Calibration Laboratories. International Organization for Standardization: Geneva, Switzerland, 2017.
17. Smith, I.M.; Onakunle, F.O. SSfM-3 1.6.1—XLGENLINE, *Software for Generalised Least-Squares Fitting, Developed by the (NPL), Teddington, UK, NPL Document Reference: CMSC/M/06/657*; National Physical Laboratory: Teddington, UK, 2007.
18. J2601_202003; Fueling Protocols for Light Duty Gaseous Hydrogen Surface Vehicles. SAE International: Warrendale, PA, USA, 2020.
19. Aarhaug, T.A.; Kjos, O.; Bacquart, T.; Valter, V.; Optenhostert, T. Assessment of hydrogen quality dispensed for hydrogen refuelling stations in Europe. *Int. J. Hydrogen Energy* **2020**, *46*, 29501–29511. [[CrossRef](#)]
20. Aarhaug, T.A.; Kjos, O.S.; Ferber, A.; Hsu, J.P.; Bacquart, T. Mapping of hydrogen fuel quality in Europe. *Front. Energy Res.* **2020**, *8*, 585334. [[CrossRef](#)]
21. Bacquart, T.; Moore, N.; Hart, N.; Morris, A.; Aarhaug, T.A.; Kjos, O.; Aupretre, F.; Colas, T.; Haloua, F.; Gozlan, B.; et al. Hydrogen quality sampling at the hydrogen refuelling station—Lessons learnt on sampling at the production and at the nozzle. *Int. J. Hydrogen Energy* **2020**, *45*, 5565–5576. [[CrossRef](#)]
22. OIML R 139-1 2014 (E), *International Recommendation: Compressed Gaseous Fuel Measuring Systems for Vehicles*; International Organization of Legal Metrology: Paris, France, 2014.
23. Arrhenius, K.; Francini, L.; Bükér, O. Sampling methods for renewable gases and related gases: Challenges and current limitations. *Anal. Bioanal. Chem.* **2022**, *414*, 6285–6294. [[CrossRef](#)] [[PubMed](#)]
24. Weiyu, S.; Baolian, Y.; Ming, H.; Zhigang, S. The effects of H₂S and CO mixtures on PEMFC performance. *Int. J. Hydrogen Energy* **2007**, *32*, 4412–4417. [[CrossRef](#)]

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.