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Development of Leakage Detection System for Safe Transportation of Compressed Gaseous Hydrogen Using Tube Trailers

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Reliability, Availability, Maintainability, and Safety (RAMS)
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Preface

The thesis is part of the two-year international master's degree program in Reliability, Availability, Maintainability, and Safety (RAMS) Engineering at the Norwegian University of Science and Engineering (NTNU). The project is carried out in spring semester of 2022 at the Department of Mechanical and Industrial Engineering at NTNU, in Trondheim, Norway. Professor Nicola Paltrinieri from the abovementioned department at NTNU was the main supervisor while Federico Ustolin from the same department at NTNU was the co-supervisor.

Executive Summary

Hydrogen may be a suitable substitute to fossil fuels thanks to its unique properties. After production, hydrogen can be transported and distributed using pipelines, tanker trucks, tube trailers, or ships. Transportation by tube trailers is well established for relatively small quantities over short distances (up to 200 km). Compressed gaseous hydrogen (CGH_2) can be transported by cylinders or bundled tubes on tube trailer on trucks with pressures between 20-70 MPa. However, the safe usage and handling of compressed gaseous hydrogen poses different challenges due to hydrogen's ease of leaking, low ignition energy of 0.017 mJ and wide range of combustible fuel-air mixtures of 4-75%, buoyancy and embrittlement. Therefore, safety is a major concern for the emerging hydrogen infrastructure. An important principle underlying safe hydrogen use is to seek designs and operations that minimize the severity of the consequences of a potential mishap. This can be done by use of alarms and warning devices (including hydrogen and fire detectors), area control around a hydrogen system and use of personal protective equipment. However, there is currently no indication on how to develop a system for detecting leakages for transporting compressed gaseous hydrogen using tube trailers. In this work, a leakage detection system for tube trailers along with automatic safety barriers has been developed. Along with the implementation of a leakage detection system, the assurance of its intended and effective operation is required. Therefore, a modification of 'HSG254: Developing Process Safety Indicators' was adopted, and safety indicators were developed. Through the implementation of the developed leakage detection system and adopting its relevant safety indicators, the risks regarding the transportation of compressed gaseous hydrogen using tube trailers are expected to be controlled to a large extent.

Abbreviations

CGH_2	Compressed Gaseous Hydrogen
CHHP	Combined Heat, Hydrogen, and Power
CNG	Compressed Natural Gas
DOE	US Department of Energy
EC	Electrochemical Sensor
FET	Field Effect Transistor
HDPE	High Density Polyethylene
IET	Institute for Energy and Transport
LEL	Lower Explosive Limit
LFL	Lower Flammability Limit
LH_2	Liquid Hydrogen
LOC	Loss of Containment
MEMS	Micro-Electromechanical Systems
MIMAH	Methodology for the Identification of Major Accident Hazards
MOS	Metal Oxide Semiconductor
MOX	Metal Oxide
NFPA	National Fire Protection Association
NG	Natural Gas
NREL	National Renewable Energy Laboratory
QCM	Quartz Crystal Microbalance
SAW	Surface Acoustic Wave Sensor
TC	Thermal Conductivity Sensor
TIR	Technical Information Report
UAV	Unmanned Aerial Vehicle
UGLD	Ultrasonic Gas Leak Detector
VCE	Vapour Cloud Explosion
YSZ	Yttria Stabilized Cubic Zirconia

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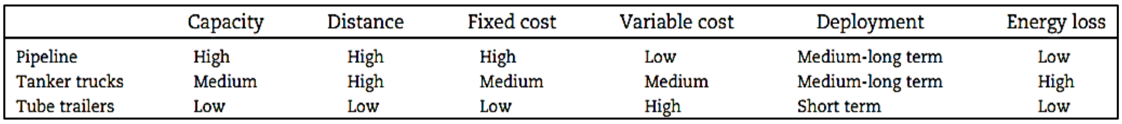
1. Introduction

1.1. Background

Hydrogen may be a suitable substitute to fossil fuels thanks to its unique properties. In fact, it is clean (if employed in fuel cells), abundant, potentially renewable, energy-efficient, and versatile (Najjar, 2013). Hydrogen has been employed in numerous new applications in the last two decades, and some of the most are listed below (Ustolin et al., 2020).

- First combined heat, hydrogen, and power (CHHP) systems plant installed in California in 2016
- First hydrogen-fuelled stove for domestic application developed by Empa company in 2018
- Achievement of the longest duration flight of a multi-rotor unmanned aerial vehicle (UAV) by the hydrogen fuelled drone developed by the EnergyOr company in 2015
- First fuel cell and battery ferry should be operative in 2020 in Norway as result of the HYBRIDship project

Although hydrogen has high gravimetric energy density of 143 MJ/kg (higher heating value), which is three times higher than that of hydrocarbon-based fuels, its low density of 0.084 kg/m^3 at standard conditions requires to compress or liquefy it to achieve large storage capacities (Yoo et al., 2021). The compression of hydrogen is the most popular and common method for hydrogen storage which has great releasing ratio and enables rapid filling (Abohamzeh et al., 2021). Almost 80% of hydrogenation processes all over the world utilize the high-pressure hydrogen storage technologies in both hydrogen storage and transportation fields (Hassan et al., 2021). However, extremely high pressure up to 70 MPa or 100 MPa is required for vehicle application (Hassan et al., 2021). To satisfy industrial requirements such as the volumetric capacity, the internal pressure must be increased up to 70 MPa resulting in 40 g/L hydrogen density and 1.32 kWh/L energy volumetric density (Hassan et al., 2021). Hydrogen can be produced through biological, thermochemical, or electrochemical routes. After production, hydrogen can be transported and distributed using pipelines, tanker trucks, tube trailers, or ships. Several trade-offs (Figure 1) exist among these different methods (Hjeij et al., 2022).

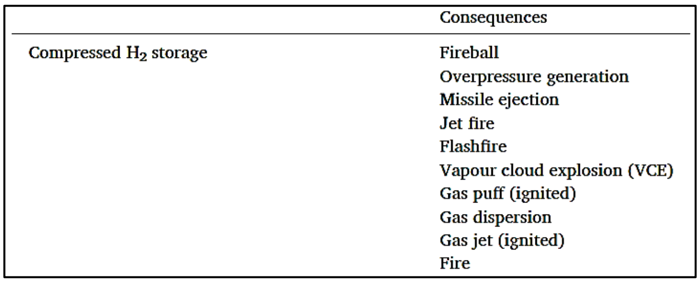


	Capacity	Distance	Fixed cost	Variable cost	Deployment	Energy loss
Pipeline	High	High	High	Low	Medium-long term	Low
Tanker trucks	Medium	High	Medium	Medium	Medium-long term	High
Tube trailers	Low	Low	Low	High	Short term	Low

Figure 1. Trade-off between different methods of hydrogen distribution (Hjeij et al., 2022)

Transportation by tube trailers is well established for relatively small quantities over short distances (up to 200 km) (Hassan et al., 2021). Compressed gaseous hydrogen (CGH$_2$) can be transported by cylinders or bundled tubes on tube trailer on trucks with pressures between 20 -70 MPa (Hassan et al., 2021). Five types of storage tanks with different types of materials and maximum allowable pressures have been developed (Reddi et al., 2018). These will be discussed in detail in latter parts of this report.

The safe usage and handling of compressed gaseous hydrogen poses different challenges due to hydrogen's ease of leaking, low ignition energy of 0.017 mJ and wide range of combustible fuel-air mixtures of 4-75%, buoyancy and embrittlement (Najjar, 2013). The major handling issues of gaseous hydrogen result in loss of containment (LOC). A LOC is an uncontrolled or unplanned release of the substance from primary containment (Abohamzeh et al., 2021). The accident scenarios caused by LOC of gaseous hydrogen are shown in Figure 2 (Abohamzeh et al., 2021).



	Consequences
Compressed H_2 storage	Fireball
	Overpressure generation
	Missile ejection
	Jet fire
	Flashfire
	Vapour cloud explosion (VCE)
	Gas puff (ignited)
	Gas dispersion
	Gas jet (ignited)
	Fire

Figure 2. Consequences of Gaseous Hydrogen LOC (Abohamzeh et al., 2021)

According to Methodology for the Identification of Major Accident Hazards (MIMAH), four types of critical events for gaseous hydrogen either at atmospheric conditions or at storage conditions in high-pressure vessels can occur (Ustolin et al., 2020). They are as following.

- Start of fire
- Breach on the containing shell in vapor phase
- Leak from gas pipe
- Catastrophic rupture

Therefore, safety is a major concern for the emerging hydrogen infrastructure. An important principle underlying safe hydrogen use is to seek designs and operations that minimize the severity of the consequences of a potential mishap (ISO, 2015). This can be done by use of alarms and warning devices (including hydrogen and fire detectors), area control around a hydrogen system and use of personal protective equipment (ISO, 2015). A hydrogen system should include caution and warning devices as necessary to alert personnel in the event of any abnormal condition, malfunction or failure (ISO, 2015). Such devices should provide the personnel with adequate time to respond to the event. The warning system should provide an audible or a visible alarm, or both (ISO, 2015). Some system alarm/warning conditions are as following.

- Pressure (high or low, as appropriate) (ISO, 2015)
- Valve position (open or closed, as appropriate) (ISO, 2015)
- Hydrogen leak (ISO, 2015)
- Fire (ISO, 2015)

As part of a safety system, sensors can perform several important functions, including indication of an unintended hydrogen release, activation of mitigation strategies to preclude the development of dangerous situations (e.g., initiate corrective measures to prevent accumulation or the possibility of delayed ignition), activation of alarm and communication systems, and to initiate system shutdown (Post et al., 2021). The role of hydrogen sensors for assurance of safety is recognized, and accordingly, their use in hydrogen facilities is often mandated by codes like National Fire Protection Association (NFPA) 2, International Fire Code (IFC) 2015.

The term sensor can have different meanings among stakeholders. For example, a sensor has been defined as a small device that transforms chemical information of a quantitative or qualitative type into an analytically useful signal as the result of a chemical interaction or process between the analyte gas and sensor device, (SAE International, 2018). However, there is no universally accepted definition of sensor and the term sensor is often used interchangeably for detection apparatus (e.g., instruments, detectors, analysers) and even sensing elements. By the ISO 26142 definition, the sensing element would be component within a hydrogen sensor or detection apparatus where the presence of hydrogen is transduced into a measurable quantity, usually electrical in nature. Accordingly, the type of sensing element (e.g., the sensor platform) is a controlling factor for the sensor metrological performance including especially cross-sensitivity to other gases. An illustration of the distinction between sensing element, sensor, and detection apparatus is shown in Figure 3.

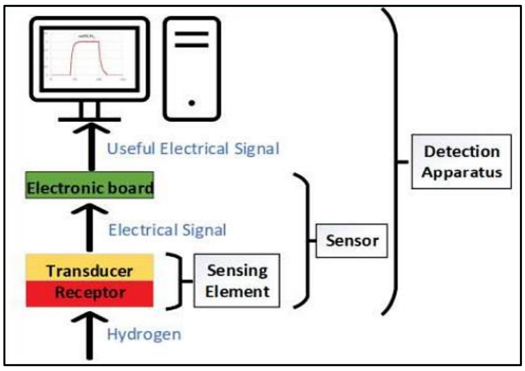


Figure 3. Distinction between Sensing Element, Sensor and Detection Apparatus (SAE International, 2018)

A sensor consists of sufficient support elements (e.g., electronic circuitry) to transform the electrical or physical response of the sensing element into useful information, such as vol% hydrogen or an electrical signal easily converted into hydrogen concentration through a transformation function (or a calibration expression) (SAE International, 2018). Additional elements may be added to the sensor to improve performance of specific metrics (e.g., chemical filters for minimizing cross-sensitivity to other gases). A detection apparatus may include displays, user interfaces, and additional elements for control systems, such as alarm activation and other advanced operations (Post et al., 2021).

However, there is currently no indication on how to develop a system for detecting leakages while transporting compressed gaseous hydrogen using tube trailers. In this work, a leakage detection system for tube trailers along with automatic safety barriers has been developed. Along with the implementation of a leakage detection system, the assurance of its intended and effective operation is required. Therefore, a modification of 'HSG254: Developing Process Safety Indicators' (Health and Safety Executive., 2006) guideline was applied for the developed system to ensure that the system works adequately. Because carefully chosen indicators through the implementation of this guide can monitor the status of key systems of the leakage detection system. It can also provide an early warning in case of deterioration of components to an unacceptable level. Adoption of the process safety performance indicators may result in.

- An increased assurance on risk management and protected reputation
- Demonstration of the suitability of the risk control systems
- Avoiding discovering weaknesses through costly incidents
- Stopping of collecting and reporting performance irrelevant information resulting in saving costs
- Better use of information collected for other purposes like quality management

Therefore, through the implementation of the developed leakage detection system and adopting its relevant safety indicators, the risks regarding the transportation of compressed gaseous hydrogen using tube trailers are expected to be controlled to a large extent.

1.2. Motivation and Objective

Increasing the safety of road transportation of compressed gaseous hydrogen through tube trailers was the motivation for the work. Several safety concerns arise due to high-pressure leakage while transportation of compressed gaseous hydrogen. This study aimed at avoiding and mitigating consequences due to leakage of hydrogen through developing a generic detection system that can be adopted while transportation of compressed gaseous hydrogen. Therefore, the specific objectives of this thesis are:

- Developing a leakage detection system for CGH_2 tube trailers.
- Develop process safety indicators for the system to ensure proper performance.

1.3. Limitations

This study was concentrated on transportation of compressed gaseous hydrogen through tube trailers. Transportation of liquid was not considered. Moreover, transportation of compressed gaseous hydrogen through pipelines and other means were not studied as well. Although those studies might provide important information regarding safety aspects of hydrogen handling, they were not included in this study.

1.4. Outline

The report contains the following chapters.

- Chapter 1 provides an introduction on necessity of leakage detection system and its safety indicators regarding transportation of compressed gaseous hydrogen.
- Chapter 2 presents the methodology adopted for the thesis work.
- Chapter 3 contains the results regarding the information searched for indications regarding leakage detection system, the development of the leakage detection system. Moreover, application of the detection system and implementation of the modified 'HSG 254: Developing Process Safety Indicators' were presented.

- Chapter 4 focuses on significance of the thesis performed and the limitations and future studies identified.
- Chapter 5 concludes the study giving a brief overview of this thesis.

2. Methodology

The work in this thesis was divided into two parts. In the one part, a leakage detection system containing a sensor system and safety barriers was developed for leakage detection of compressed gaseous hydrogen during transportation. State of the art of the hydrogen sensors and the standards regarding them were studied to gain knowledge about the probable solutions regarding the sensor system. Although the developed system consists of different types of sensors, the other sensors apart from the hydrogen sensors e.g., pressure transducer, flame detectors etc are not discussed here. Because the hydrogen sensors are comparatively newer technologies whereas the other sensors have been used in different fields for a long time and ample knowledge regarding them are available. Furthermore, state of the art of the tube trailers were studied to gain knowledge about the field of application and get indication about sensor numbers and locations. From the analysed information and taking hydrogen properties into account, the sensor system was finally developed. The suggested sensor system was then applied for different types of tube trailers. Combining this sensor system with some safety barriers, a leakage detection system was developed and suggested.

Subsequently in another part of the work, a guideline has been adopted on the basis of the 'HSG254: Developing Process Safety Indicators' issued by Health and Safety Executive to ensure proper performance of the sensor system (Health and Safety Executive., 2006). The guide provided the generic model for establishing a performance measurement system of a risk control system for a system or a procedure. Two indicators named leading and lagging indicators are set in a structured and systematic way for each critical risk control system within the whole process safety management system (Health and Safety Executive., 2006). In tandem they act as system guardians providing dual assurance to confirm that the risk control system is operating as intended or providing a warning that problems are starting to develop (Health and Safety Executive., 2006). However, these performance indicators are not a substitute for an audit programme but a complimentary activity to give more frequent or different information on system performance. Every deviation from the intended outcome or failure of a critical part of a risk control system is followed up.

Lagging indicators reveal weaknesses in the risk control system following reporting and investigation of an incident or adverse event (Health and Safety Executive., 2006). They are a form of reactive monitoring showing whether the desired safety outcome has been achieved (Health and Safety Executive., 2006). For lagging indicators, every accident should be investigated to check whether improvements should be made (Health and Safety Executive., 2006).

Leading indicators identify failings in vital aspects of the risk control system discovered during routine checks on the operation of a critical activity (Health and Safety Executive., 2006). They are a form of active monitoring showing whether the risk control systems are operating as designed (Health and Safety Executive., 2006). For leading indicators, all the deviations from tolerance set for each indicator should be reported and followed up to determine deterioration of components if there is any (Health and Safety Executive., 2006).

Main steps of 'HSG254: Developing Process Safety Indicators' guide are mentioned in the following (Health and Safety Executive., 2006).

Step 1: Establish the organisational arrangements to implement the indicators.

- Appoint a steward or champion.
- Set up an implementation team.
- Senior management should be involved.

Step 2: Decide on the scope of the measurement system. Consider what can go wrong and where.

- Select the organisational level.
- Identify incident scenarios - what can go wrong?
- Identify the immediate causes of hazard scenarios.
- Review performance and non-conformances.

Step 3: Identify the risk control systems in place to prevent major accidents. Decide on the outcomes for each and set a lagging indicator.

- What risk control systems are in place?
- Describe the outcome.
- Set a lagging indicator.
- Follow up deviations from the outcome.

Step 4: Identify the critical elements of each risk control system, (ie those actions or processes which must function correctly to deliver the outcomes) and set leading indicators.

- What are the most important parts of the risk control system?
- Set leading indicators.
- Set tolerances.
- Follow up deviations from tolerances.

Step 5: Establish data collection and reporting system.

- Collect information - ensure information/unit of measurement is available or can be established.
- Decide on presentation format.

It is important to ensure that the relevant information is readily available within the organisation. The data should be presented to clearly show the link between the lagging indicator and the leading indicator(s) of the risk control systems (Health and Safety Executive., 2006). This will clearly highlight the cause-and effect links between them.

Step 6: Review.

- Review performance of process management system.
- Review the scope of the indicators.
- Review the tolerances.

If performance is poor against a group of leading indicators but the associated lagging indicator is satisfactory, it is likely that the leading indicators selected are too far removed from the critical control measure that delivers or maintains the desired outcome (Health and Safety Executive., 2006). If a group of leading indicators are on target and closely linked to the risk control system but the associated lagging indicator shows poor performance, it is likely that risk control system is ineffective in delivering the desired outcome (Health and Safety Executive., 2006).

Every few years, the scope of the full set of indicators needs to be reviewed to ensure indicators still reflect the main process risks (Health and Safety Executive., 2006). It could be that the tolerance has been set at the wrong point, for example set too leniently/stringently, so the information or data does not adequately reflect reality (Health and Safety Executive., 2006). In such cases, the tolerance should be reviewed.

There are several organizational aspects of the guideline which are important for the organizations or enterprises. But they were not included in the scope of the thesis work. In essence, the steps described in the guideline have been modified and adopted to match the scope of the thesis. The following actions were identified from the guideline and performed to provide assurance that the risk control measures are working adequately.

- Identification of the accident scenarios and their causes
- Identification of risk control systems in place and their desired safety outcome
- Identification of important parts and safety indicators of the risk control system

The first two actions have been presented in the first part of the work to highlight the importance of developing a leakage detection system for transporting compressed gaseous hydrogen with tube trailers. However, the third action has been presented after the development of the leakage detection system. Because identification of important parts, lagging indicators and leading indicators require knowledge about the risk control system under study.

3. Results

Implementation of the first two parts of modification of guide 'HSG254: Developing Process Safety Indicators' proposed in methodology have been depicted in Sec 3.1 and 3.2. Then Sec. 3.3 to 3.7 illustrates development of a sensor system for leakage detection in tube trailers. Current hydrogen sensors being used, performance requirements of hydrogen sensors, and hydrogen transportation by tube trailers are investigated for this purpose. Combining the results of the analyses, a sensor system has been developed indicating the required types, numbers and locations of the sensors. Then the developed system has been applied for different configurations of tube trailers (3.8). Lastly, identification of the critical parts of the developed system and their safety indicators are presented in 3.9.

3.1. Identification of Accident Scenarios and Their Causes

The major hazards issues that can happen to CGH_2 tubes during transportation can be related to LOC (Abohamzeh et al., 2021).

Main process safety incident scenario in hydrogen transport: Loss of Containment (LOC)

Causes of LOC: All types of mechanical and physical failures can be considered as LOC causes (Ustolin et al., 2020). These failures include collision during transportation, mechanical failures of hydrogen equipment such as safety devices, storage vessels, vent, exhaust and vaporization systems. Hydrogen embrittlement is a well-known phenomenon responsible of material degradation and thus can be the cause of different types of equipment rupture (Ustolin et al., 2020). For this reason, the materials for hydrogen service must be selected in order to avoid any embrittlement phenomena (Ustolin et al., 2020).

Accident Scenarios of LOC: LOC may lead to hydrogen release which is followed by immediate ignition causing jet fires or a delayed ignition resulting in the explosion (Abohamzeh et al., 2021). The consequences of these events have a direct or indirect impact on humans, the environment, structures, and properties.

A possible scenario that happens after the release of hydrogen is immediate ignition that leads to a jet fire (Abohamzeh et al., 2021). A jet fire is a high-velocity turbulent flame that results from the combustion of fuel releasing in a certain direction with substantial momentum. Another possible scenario after the release of hydrogen is a delay between release and ignition, which may cause an explosion (Abohamzeh et al., 2021). In this case, the hydrogen has sufficient time to be mixed with air before being ignited, and the consequence would be a flash fire or a vapour cloud explosion (VCE) (Abohamzeh et al., 2021). There are risks of damage for equipment and structures exposed to thermal radiation and direct flames (Abohamzeh et al., 2021). A substantial possible risk exists for the explosion of vessels containing hydrogen in the case of their exposure to thermal radiation or high temperature (Abohamzeh et al., 2021). The exposure of hydrogen vessels to high thermal radiation may result in a considerable hazard potential for mechanical explosion. The hydrogen release without fire accidents is still a hazard issue, particularly in confined spaces since it causes asphyxiation. This is because hydrogen, which is a non-toxic gas, replaces oxygen to reduce its concentration below 19.5% by volume (Abohamzeh et al., 2021). The accumulation of hydrogen into closed spaces adjacent to the source then poses an asphyxiation hazard for the people being there.

3.2. Identification of Risk Control Systems in Place and Desired Safety Outcomes

To control and mitigate the consequences of the LOC, there are no risk control systems in place for hydrogen tube trailers. This highlights the importance of developing a leakage detection system to enable swift actions in case of situations when there is a possibility of LOC or even when LOC occurs. This acted as the motivation behind the development of the leakage detection system consisting of sensor system and safety barriers. The development and the application of the leakage detection system will be illustrated in the latter parts.

Desired Safety Outcome:

- Proper detection of released CGH_2 and hydrogen flame
- Prevent leakages
- Prevent accumulation of hydrogen in case of leakage

3.3. Hydrogen Sensors

The market acceptance of emerging hydrogen and fuel cell technologies depends directly on their perceived safety. Therefore, hydrogen safety measures have been developed to ensure the safe use of hydrogen, including guidelines for mitigation of fault and accidents. Detection of leaked hydrogen is of utmost importance to avoid accidents. Best practices regarding leakage detection include the following.

- Listen for the sound of high-pressure gas escaping (H2Tools, 2022)
- Listen and watch for alarms (H2Tools, 2022)
- Use portable hydrogen detectors (H2Tools, 2022)
- Check for small leaks by applying a soap bubble solution on the surface of areas where leaks are suspected (H2Tools, 2022)
- Hydrogen (or flammable gas) detectors installed where leaking hydrogen is likely to concentrate (H2Tools, 2022)
- Monitoring piping pressures or flow rates changes (H2Tools, 2022)
- Detection of fire/flame (H2Tools, 2022)

These may be practised by usage of several sensors like hydrogen sensor, pressure transducer, temperature sensor, flame detector and sound detectors simultaneously. Although all the required sensors will be implemented in the sensor system developed in this study, only the hydrogen sensors are discussed in this section due to the technology being comparatively new and still in an improvement phase. There is no perfect hydrogen sensor yet which meets all the performance requirements. Therefore, a comparison between the current hydrogen sensors is presented with their advantages and disadvantages. Knowledge about the hydrogen sensors is necessary as it would provide information about which sensors can be implemented for the desired application. Materials about different types of hydrogen sensors have been studied and their current state of development have been discussed below.

Hydrogen sensors are deployed to increase safety in applications such as hydrogen production, storage, distribution and use. They are typically independent of the main operational features of a system and are required by code for various hydrogen operations (Buttner et al., 2012). Hydrogen safety sensors can trigger an alarm before hydrogen concentrations reach the lower flammability limit (LFL) of 4 vol% in air. This may be followed by additional measures such as closing off the hydrogen supply, increasing ventilation or initiating system shutdowns. This is a significant contribution to the safe use of hydrogen. To facilitate the reliable and proper use of hydrogen sensors, sensor testing facilities were independently established by the European Commission's Joint Research Centre - Institute for Energy and Transport (IET) and by the US Department of Energy (DOE) at the National Renewable Energy Laboratory (NREL) (Palmisano et al., 2015). Several types of hydrogen sensors have been developed. Each of them has their distinct advantages and disadvantages. The result from the studied hydrogen sensors are presented below.

3.3.1. Catalytic Hydrogen Sensors

Catalytic sensors are composed of two thin platinum wires which are embedded in a ceramic bead (pellistor) and connected through a Wheatstone bridge (Foorginezhad et al., 2021). The bead is coated with a catalyst (platinum or noble matals). One pellistor catalyzes hydrogen oxidation selectively. When the pellistor is heated to 500–550° C, hydrogen is oxidized on the active bead surface resulting in temperature and platinum filament resistance increment (Foorginezhad et al., 2021). The generated Wheatstone bridge imbalance can be linearly related to the concentration of hydrogen (Foorginezhad et al., 2021). Catalytic sensors commonly used in the petroleum industry. However, they have some limitations like high power consumption, cross-sensitivity and requirement of presence of oxygenv (Foorginezhad et al., 2021). If these can be solved, catalytic sensors can be used in hydrogen vehicles. Modern Micro-Electromechanical Systems (MEMS) can be utilized to improve the power requirements and response time (Foorginezhad et al., 2021). For example, thermoelectric gas sensor (TGS) based on combination of catalytic combustion and thermoelectric conversion proved to be robust and reliable as well as with wide-range and selective detection (Foorginezhad et al., 2021).

3.3.2. Electrochemical Sensors (EC)

An electrochemical reaction occurs between cathodes and anodes producing a signal (current or voltage) related non-linearly with hydrogen concentration (Foorginezhad et al., 2021). Amperometric and potentiometric sensors are electrochemical sensors producing current or potential respectively proportional to the gas concentration (Foorginezhad et al., 2021). ECs have great sensitivity, a broad linear range, and low energy power consumption. However, there are some disadvantages (Foorginezhad et al., 2021). For example, two-year lifetime, limited temperature range, moderate selectivity, sensitivity to environmental conditions (pressure), and a decrease in sensitivity attributed to deterioration of electrode catalyst (Foorginezhad et al., 2021). In comparison, the response from potentiometric sensors is logarithmic (lower accuracy at higher concentrations), while that of amperometric ones is linear (more sensitive) (Foorginezhad et al., 2021). Recently a cost-effective and wide-detection-range Nafion-based potentiometric hydrogen sensor for fuel-cell vehicles with 20–99.99% detection range and response time was less than 5 s in various environmental conditions has been developed (Foorginezhad et al., 2021). The response changed when the sensor was soaked at 80 °C. Also, yttria-stabilized cubic zirconia (YSZ) based potentiometric sensor coupled with sensing electrode CdWO4 was fabricated having 0.5–3 vol% range of detection at 500 °C (Foorginezhad et al., 2021).

3.3.3. Semi-conductive Metal-oxide Sensors (MOX)

A heater raises temperature of a semiconductive metal-oxide layer to operating temperature (500 °C) (Foorginezhad et al., 2021). The electrical resistance of this layer changes through hydrogen diffusion into the layer and reaction of hydrogen with oxygen (Foorginezhad et al., 2021). They are small, cost-effective, and can conveniently be mass-produced and show sufficient sensitivity for hydrogen applications (Foorginezhad et al., 2021). But they are affected by moisture variation, have long response and recovery time and require oxygen for stable operation (Foorginezhad et al., 2021). Metal-Oxide-Semiconductor (MOS) is a type of MOX sensor that can work properly in absence of oxygen and have simple structure, high sensitivity with a fast response, small size, and low cost (Foorginezhad et al., 2021). But they have limitations like high operating temperature and poor selectivity/sensitivity (Foorginezhad et al., 2021). Further research has been being made to overcome the shortcomings of MOS.

3.3.4. Thermal Conductivity Sensors (TC)

The temperature of an electrically heated sensing element changes after exposure to hydrogen (Foorginezhad et al., 2021). The thermal conductivity of hydrogen is about seven times higher compared with other gases. This makes TC suitable for hydrogen detection. The sensor is heated to the temperature that the sensing element resistance deviates from the limit of Ohm's law (Foorginezhad et al., 2021). Advantages of TC are that oxygen is not required for operation, wide detection range of <1-100% vol. hydrogen, less electric power consumption compared with catalytic sensors and fast operating time (Foorginezhad et al., 2021). Nevertheless, they have poor selectivity and are sensitive to environmental factors (Foorginezhad et al., 2021). But special coatings can improve the selectivity and micro-hotplate technology helped to improve sensitivity to environmental factors. The improvements added with the fact that less maintenance is required make TC the most appropriate ones to be used for automotive applications (Foorginezhad et al., 2021).

3.3.5. Optical Sensors

Optical changes are used to detect hydrogen. Such as, micro-mirror sensors sense reflected light alteration after hydrogen absorption, and optic-fiber hydrogen sensors detect light transmittance change across the fiber (Foorginezhad et al., 2021). Also, optical features of palladium films and chemical mediators are used for optical sensors as well. They possess qualities like proper sensitivity to hydrogen, easy operation, low cost, no requirement of electricity and long-term durability (Foorginezhad et al., 2021). The disadvantages of optical sensors are nonlinear outputs, low qualities of sensitive film, slow response rate, temperature disturbances, and humidity influences (Foorginezhad et al., 2021). It has been suggested that using metals or alloys as sensitive films, the utilization of advanced deposition techniques, optimization of sensing head structures and development of thin-film deposition technology can result into improved sensing performance (Foorginezhad et al., 2021).

3.3.6. Palladium (Alloy) Film Based Sensors

Palladium-film technology works depending on change of film resistance due to hydrogen adsorption. Major categories of Pd-based hydrogen sensors are as following.

- Palladium resistor sensors (Foorginezhad et al., 2021)
- Palladium field-effect transistors (FETs) or capacitor sensors (Foorginezhad et al., 2021)
- Palladium optical sensors (Foorginezhad et al., 2021)
- Palladium meso-wire (nanowire) and nano-particle sensors (Foorginezhad et al., 2021)
- Palladium nanotube and nano-clusters sensors (Foorginezhad et al., 2021)

Palladium films can be used for mechanical device coating like Surface Acoustic Wave Sensors (SAW) or Quartz Crystal Microbalance Sensors (QCM) where Pd mass changes because of hydrogen adsorption and resonant frequency alteration (Foorginezhad et al., 2021). It is also suitable for detecting low level of hydrogen as small changes in the frequency can be accurately measured (Foorginezhad et al., 2021). However, they have several drawbacks. They need oxygen for operation, are dependent on chemical contaminants (mainly sulfide) and have shown slow response time (more than 30s) compared to TC sensors (Foorginezhad et al., 2021). Also, their long-term stability has not been evaluated yet. But it has been reported that chemical poisoning can be reduced using protective layers, while faster response time can be achieved using MEMS platform (Foorginezhad et al., 2021). Hydrogen Sense Technology Co. Ltd developed Pd alloy thin-film solid-state hydrogen specific sensor technology which consists of a hydrogen capacitance and hydrogen resistance sensor, a temperature sensor, a heater, and built-in software (Foorginezhad et al., 2021). They have a fast response (1–60s) with full range hydrogen detection (15 ppm–100%) in different conditions (high humidity, presence of acidic gases, and vacuum or high pressure) and can operate for almost 10 years (Foorginezhad et al., 2021). Also, Argonne National Laboratory has fabricated a tiny sensor (ANL-IN-04-077B) using an ultra-thin layer of palladium beads, leading to more rapid hydrogen adsorption and better sensitivity because of palladium hydride beads mobility (Foorginezhad et al., 2021). This overcomes the limitations of cost, operation speed, susceptibility to other gases, and temperature range. Moreover, they require no warmup, have high selectivity to hydrogen and response time of 75 ms in 2% hydrogen atmosphere, can measure 25 ppm hydrogen and meets response time and concentration criteria set by U.S. Department of Energy (DOE) (Foorginezhad et al., 2021). Overall, use of Pd in different hydrogen gas sensors led to improved performance though it is required to customize the developed sensors for required applications. However, while using Pd in electrical sensing technology, risks of unguaranteed safety and sensor longevity issues persist. But these problems can be overcome using optical sensing technology.

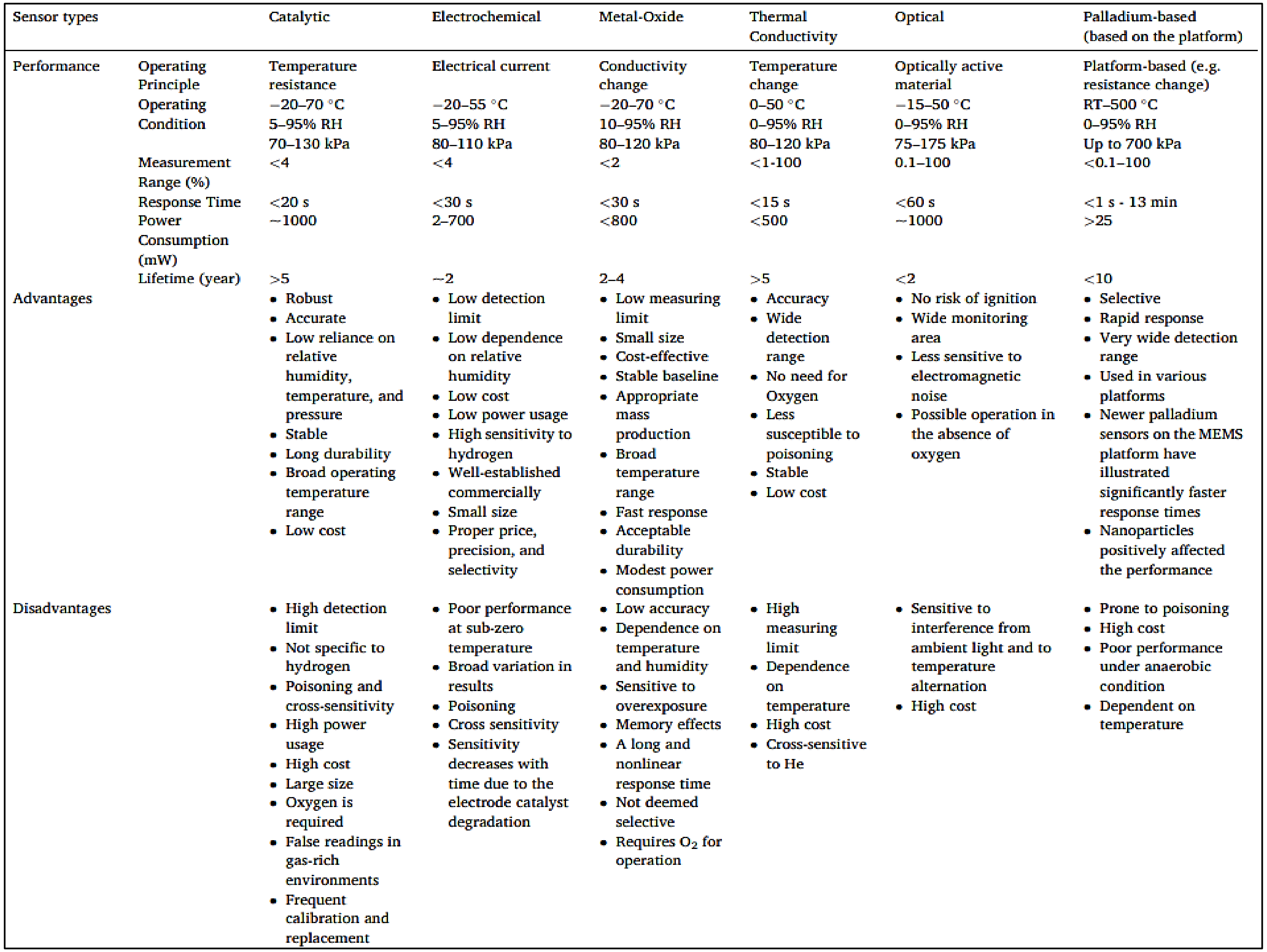
3.3.7. Combined Technology Sensors

Various sensing technologies have been combined for improved performance. For example, a small-sized H_2-Semicon®-Detector has been developed combining selective MOX gas sensor and thermal conductivity detector (Foorginezhad et al., 2021). The combination resulted in highly selective measurement of hydrogen in the 0–10% range (optional up to 100%), high sensitivity, stability, and safety, with ≤1 s response time at 5 ppm (Foorginezhad et al., 2021). This can be used for leakage monitoring either in fuel cell systems or by mobile and stationary devices, in addition to chemical processes and equipment monitoring in the industry. Furthermore, Cyber Genius (Sensitron S.r.L.) has been developed through a combination of an electrochemical cell and a pellistor sensing platform (Foorginezhad et al., 2021). Additionally, HLS-440 sensor (Applied Sensor GmbH) is fabricated based on a field-effect sensing element combined with a thermal conductivity sensing element, a temperature sensor, and a heater (Foorginezhad et al., 2021). Measurement of 100% hydrogen, operation in harsh environment, and response and recovery time below 5 s and 10 s respectively are properties of this sensor (Foorginezhad et al., 2021). Also, XEN-5310 device (Xensor Integration BV), a thermal conductivity-based sensor combined with temperature and humidity sensors, can measure 0–4% or 0–100% of hydrogen with a response and recovery time of 1 s (Foorginezhad et al., 2021).

The abovementioned sensors require direct contact of hydrogen with the sensors. They are called point sensors. A comparison between the abovementioned sensors is illustrated in Figure 4. However, while usage in outdoor, hydrogen may easily dilute and move away from the sensor. This problem may be solved by using Ultrasonic Gas Leak Detector (UGLD).

3.3.8. Ultrasonic Gas Leak Detector (UGLD)

UGLDs detect the airborne acoustic ultrasound generated when CGH_2 escapes from a leak (Fecarotta & Janowski, 2021). They are non-concentration-based sensors and send signal to the control system as soon as the onset of a leak (Fecarotta & Janowski, 2021). UGLDs can be considered as a first layer of protection in pressurized gas installations and used together with conventional gas detection methods to secure optimal protection in outdoor usages. The main advantage of this type of sensor is that it does not need to wait for a gas concentration to accumulate and form a potentially explosive cloud. Their detection coverage depends on surrounding noise level (Fecarotta & Janowski, 2021). For high, low and very low noise areas, their detection coverages are respectively 5-8 m, 9-12 m and 13-20 m (Fecarotta & Janowski, 2021). However, they cannot be used to detect liquid leaks or in places with extreme levels of ultrasonic background noise (>95 dB) (Fecarotta & Janowski, 2021).



Sensor types		Catalytic	Electrochemical	Metal-Oxide	Thermal Conductivity	Optical	Palladium-based (based on the platform)
Performance	Operating Principle	Temperature resistance	Electrical current	Conductivity change	Temperature change	Optically active material	Platform-based (e.g. resistance change)
	Operating Condition	−20–70 °C 5–95% RH 70–130 kPa	−20–55 °C 5–95% RH 80–110 kPa	−20–70 °C 10–95% RH 80–120 kPa	0–50 °C 0–95% RH 80–120 kPa	−15–50 °C 0–95% RH 75–175 kPa	RT–500 °C 0–95% RH Up to 700 kPa
	Measurement Range (%)	<4	<4	<2	<1-100	0.1–100	<0.1–100
	Response Time	<20 s	<30 s	<30 s	<15 s	<60 s	<1 s - 13 min
	Power Consumption (mW)	~1000	2–700	<800	<500	~1000	>25
	Lifetime (year)	>5	~2	2–4	>5	<2	<10
Advantages		• Robust • Accurate • Low reliance on relative humidity, temperature, and pressure • Stable • Long durability • Broad operating temperature range • Low cost	• Low detection limit • Low dependence on relative humidity • Low cost • Low power usage • High sensitivity to hydrogen • Well-established commercially • Small size • Proper price, precision, and selectivity	• Low measuring limit • Small size • Cost-effective • Stable baseline • Appropriate mass production • Broad temperature range • Fast response • Acceptable durability • Modest power consumption	• Accuracy • Wide detection range • No need for Oxygen • Less susceptible to poisoning • Stable • Low cost	• No risk of ignition • Wide monitoring area • Less sensitive to electromagnetic noise • Possible operation in the absence of oxygen	• Selective • Rapid response • Very wide detection range • Used in various platforms • Newer palladium sensors on the MEMS platform have illustrated significantly faster response times • Nanoparticles positively affected the performance
Disadvantages		• High detection limit • Not specific to hydrogen • Poisoning and cross-sensitivity • High power usage • High cost • Large size • Oxygen is required • False readings in gas-rich environments • Frequent calibration and replacement	• Poor performance at sub-zero temperature • Broad variation in results • Poisoning • Cross sensitivity • Sensitivity decreases with time due to the electrode catalyst degradation	• Low accuracy • Dependence on temperature and humidity • Sensitive to overexposure • Memory effects • A long and nonlinear response time • Not deemed selective • Requires O_2 for operation	• High measuring limit • Dependence on temperature • High cost • Cross-sensitive to He	• Sensitive to interference from ambient light and to temperature alternation • High cost	• Prone to poisoning • High cost • Poor performance under anaerobic condition • Dependent on temperature

Figure 4. Comparison between various hydrogen detection technologies (Foorginezhad et al., 2021)

3.3.9. Existing Detection System

Exponential Power, Inc. has developed a hydrogen detector named SBS-H2-DoD (Figure 5) which meets US Department of Defence (DoD)'s rigorous UFC 3-520-05 requirements (SBS, 2019). It has a main unit which can be connected with up to three hydrogen sensors using CAT cable connections and with safety machineries like blower or fan with relays (SBS, 2019). It provides visual and audible alarms at detected 0.5% and 1% hydrogen concentration along with relays enabling triggering of necessary safety precautions (SBS, 2019).

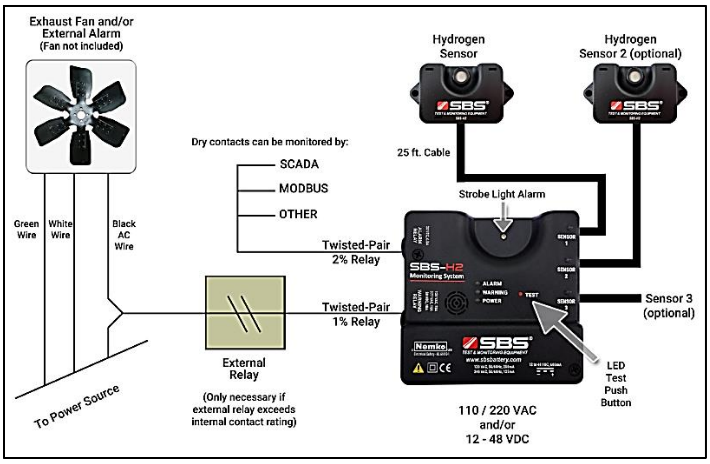


Figure 5. Schematic of SBS-H2-DoD hydrogen detector (SBS, 2019)

When the concentration of hydrogen reaches 1% by volume, the "1% Caution" yellow LED will light and the 1% internal relay will close (SBS, 2019). If the hydrogen gas concentration reaches 2% by volume, the "2% Warning" red LED will flash and an 80 dB alarm will sound (SBS, 2019).

3.4. Other Sensors

The sensors stated so far are hydrogen sensors. Other type of sensors and detectors such as pressure gauges, flame detectors and temperature sensors can be used as added layer of detection system to increase the system's capability to detect hydrogen. This will be discussed in latter parts of the report.

3.5. Hydrogen Sensors' Performance Requirements

Knowledge about performance requirements of the hydrogen sensors is important as it would provide concrete selection basis for implementation in different applications. Moreover, testing requirements can be excerpted from this knowledge as well. Therefore, standards regarding hydrogen sensors and materials have been studied to collect information about relevant requirements.

3.5.1. Standards and Technical Reports for Hydrogen Sensors or Detection Apparatus

Different standards and technical reports have been studied and presented below to learn about the requirements regarding the usage of compressed gaseous hydrogen.

3.5.1.1. ISO 26142: Hydrogen Detection Apparatus: Stationary Applications

The standard provides requirements regarding mainly sensor performance and their assessment methods and conditions for hydrogen detection apparatus in stationary applications. It was primarily intended for hydrogen detection apparatus at vehicle refuelling stations, but this standard can also be applied to other stationary installations where the detection of hydrogen is required. Hydrogen-related facilities might be required to have the ability to detect hydrogen leak concentrations before a specified concentration of hydrogen or fraction of flammable limit is reached to allow for single and/or multilevel safety operations (ISO, 2010). The requirements applicable to the overall safety system, as well as the installation requirements of such apparatus, are excluded. This international standard sets out only the requirements applicable to a product standard for hydrogen detection apparatus, such as precision, response time, stability, measuring range, selectivity and poisoning (ISO, 2010).

3.5.1.2. ISO 23273: Fuel cell road vehicles — Safety specifications — Protection against hydrogen hazards for vehicles fuelled with CGH_2

The standard deals with important requirements and specifications of different components of a vehicle fuelled with CGH_2 for both normal operating (fault-free) and single fault conditions (ISO, 2013). It does not apply to manufacturing, maintenance, and repair.

3.5.1.3. ISO 15916: Basic considerations for the safety of hydrogen systems

Requirement of hydrogen detection components and other safety systems are provided. Moreover, locations for hydrogen sensor placement are mentioned in this standard. They are stated below (ISO, 2015).

- Locations where hydrogen leaks or spills are possible (ISO, 2015)
- At hydrogen connections that are routinely separated (for example, hydrogen refuelling ports) (ISO, 2015)
- Locations where hydrogen could accumulate (ISO, 2015)

Besides, important factors to consider while selecting hydrogen sensor according to the standard are mentioned below.

- Accuracy (ISO, 2015)
- Reliability (ISO, 2015)
- Cross sensitivity (ISO, 2015)
- Maintainability (ISO, 2015)
- Calibration (ISO, 2015)
- Zero drift (ISO, 2015)
- Detection limits (high and low) (ISO, 2015)
- Response time (ISO, 2015)
- Recovering or non-recovering in time (ISO, 2015)
- Active or passive techniques with and without energy supply (ISO, 2015)
- Compatibility with the system (ISO, 2015)

It is also stated in the standard that apart from the stationary detection system, a portable hydrogen detector should be used by hydrogen system users in and around a hydrogen system (ISO, 2015). Concentration level for main alarm is specified to be 1% volume in air, which is equivalent to approximately 25 % of the LFL (ISO, 2015). This level normally should provide adequate time to respond in an appropriate manner, such as system shutdown, evacuation of personnel, or other measures as necessary (ISO, 2015).

Furthermore, invisible hydrogen air fire detectors are suggested to be used in this standard (ISO, 2015). A hydrogen/air flame is almost invisible to the human eye during daylight and the radiation of a hydrogen flame is low (ISO, 2015). Due to these two characteristics of a hydrogen flame, means to detect the presence of a hydrogen flame are instructed to be used in all areas in which leaks, spills, or hazardous accumulations of hydrogen may occur (ISO, 2015).

Important factors for consideration while selecting a hydrogen flame detector are as following.

- Detection distance and area covered (ISO, 2015)
- Susceptibility to false alarms from sources such as the sun, lightning, welding, lighting sources and background flare stacks (ISO, 2015)
- Response time (ISO, 2015)
- Sensitivity to appropriate radiation spectrum (ISO, 2015)

Apart from the above-mentioned standards, other standards also provide instructions regarding hydrogen sensor usage for addressing essential health issues and safety requirements. According to other standards, the application of sensors in atmosphere with explosion possibility requires the consideration of the three important aspects (Foorginezhad et al., 2021).

- The sensor needs to be protected against explosion in accordance with the series of standards IEC60079.
- The sensor must meet performance requirements to monitor hydrogen as flammable gas in accordance with IEC 60079 and ISO 26142.
- The sensor should follow performance safety in accordance with the series of standards IEC 61508 for electrical, electronic, and programmable electronic safety-related systems.

3.5.1.4. SAE J3089 Report: Characterization of On-Board Vehicular Hydrogen Sensors

SAE Technical Information Report (TIR) provides test methods for evaluating hydrogen sensors when the hydrogen system integrator and/or vehicle manufacturer use such devices on board hydrogen vehicles (SAE International, 2018). These tests are derived from methods developed by researchers at the NREL Hydrogen Safety Sensor Test Laboratory. Many are similar to the test procedures presented in ISO 26142 (SAE International, 2018). The original NREL test methods were modified to be more compatible with the on-board vehicular environmental and operating conditions and accordingly are consistent with SAE component standards. The tests are divided in two categories. One is performance-based and were developed to assess hydrogen sensor metrological parameters; the others were supplemental electrical safety and physical stress tests (SAE International, 2018).

The following tests have been designed to quantify metrological performance of on-board hydrogen sensors (SAE International, 2018).

- Sensor Kinetics and Validation Test (Initial Validation) (SAE International, 2018)
- Measuring Range/Accuracy of Response (SAE International, 2018)
- Short-Term Stability/Repeatability Test (SAE International, 2018)
- Pressure Dependence Test (SAE International, 2018)
- Temperature Dependence Test (SAE International, 2018)
- Humidity Dependence Test (SAE International, 2018)
- Operation Above the Measuring Range Test (SAE International, 2018)
- Orientation Effect Test (SAE International, 2018)
- Sensor Power Tests (SAE International, 2018)
- Cold Power-Up (SAE International, 2018)
- Over- and Under-Power Test (SAE International, 2018)
- Sensor Kinetics and Validation Test (Final Validation) (SAE International, 2018)
- Chemical Stress Test - Impact of Chemical Interferents and Chemical Poisons (SAE International, 2018)
- Impact of Chemical Interferents (Cross Sensitivity) (SAE International, 2018)
- Impact of Chemical Poisons (SAE International, 2018)

The following tests have been designed to characterize the robustness of on-board hydrogen sensors against various physical stressors and to provide assurance that the hydrogen sensor will not cause an ignition when exposed to a combustible atmosphere (SAE International, 2018).

- Thermal Shock Test (SAE International, 2018)
- High Humidity/High-Temperature Storage Test (SAE International, 2018)
- Vibration Test (SAE International, 2018)
- High-Temperature Operation Test (SAE International, 2018)
- Ignition Test (SAE International, 2018)

The sensor test protocols were adapted to allow test procedures that can be performed within a more practical time period than that afforded by the methods specified in either ISO 26142 or the standard NREL sensor test protocols while maintaining rigorous data quality (SAE International, 2018). However, no specific application, performance specification or pass/fail criteria were defined. Therefore, the hydrogen system integrator needs to determine which tests and associated test conditions are relevant for their applications. Thus, it is up to the hydrogen system integrator to set specific test acceptance criteria necessary to achieve the required performance of their process control and protective systems.

Table 1. Different standards and report studied and their relevance

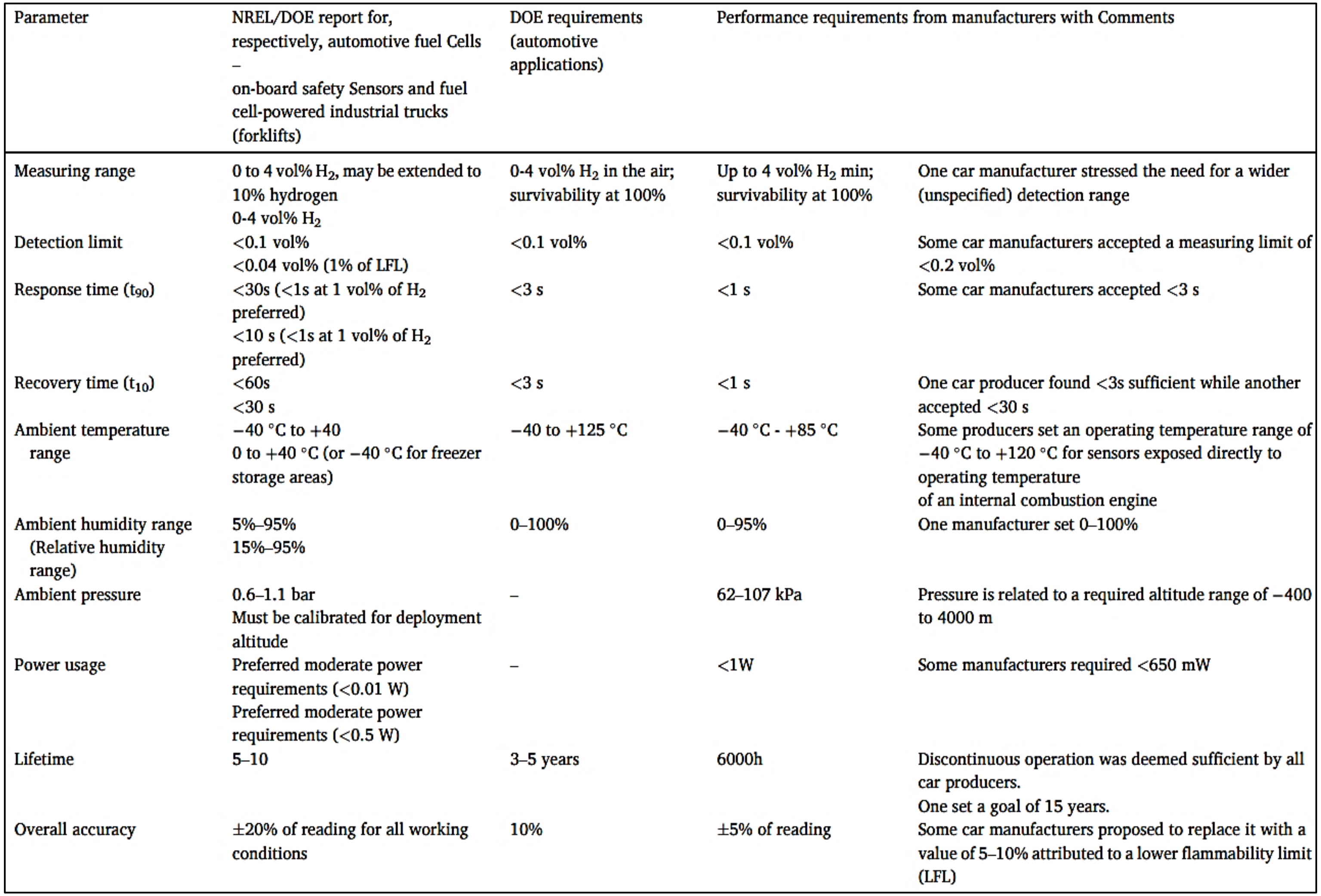
Standard/Report	Main Focus	Relevance
ISO 26142: Hydrogen Detection Apparatus: Stationary Applications	Performance requirements and assessment methods of sensors	Provides knowledge and benchmark for testing sensors to ensure proper working
ISO 23273: Fuel cell road vehicles — Safety specifications — Protection against hydrogen hazards for vehicles fuelled with CGH_2	Provides requirements and specifications of different components of a vehicle fuelled with CGH_2	Provides acceptance criteria of components used in storage system
ISO 15916: Basic considerations for the safety of hydrogen systems	Provides overview of safety issues related to hydrogen technology	Provides indications about types of sensors required, important factors regarding the sensors and probable locations where sensors are required
SAE J3089 Report: Characterization of On-Board Vehicular Hydrogen Sensors	Metrological performance and robustness tests of on-board hydrogen sensors	Provides indications about tests required for the sensors to determine performance and robustness

3.5.2. Performance Metrics and Requirements

Four essential sensor performance factors are described below.

- **Performance:** Sensors must function properly in different environments including air, nitrogen, and inert atmospheres, and should be sensitive under the lower explosive limit (LEL) (Foorginezhad et al., 2021). Both characteristics related to the chemical parts and physical features have to be also noted. Therefore, time of response is a variable to be considered along with temperature, pressure, and the ambient humidity range based on the application of sensors (Foorginezhad et al., 2021).
- **Lifetime:** Life is identified based on applications of the sensor. Generally, the range of hydrogen sensors lifetime can be established as less than 10 years for stationary power systems, 3–5 years in industrial processes, and more than 10 years for transportation purposes (Foorginezhad et al., 2021). Sensors should be operational without the need for replacement, cleaning, or persistent calibration within lifetime (Foorginezhad et al., 2021).
- **Reliability:** False alarms should be prevented to ensure the stability and safety of hydrogen usage (Foorginezhad et al., 2021). Also, responses must be accurate and sensitive (Foorginezhad et al., 2021). Sensors should be functional in high hydrogen concentrations regardless of their integrity, and their evaluations must be approved and certified based on international standards (Foorginezhad et al., 2021).
- **Cost:** Sensors need to be cost-efficient in terms of purchase, installation, and maintenance (Foorginezhad et al., 2021). Costs are related to applied technologies and expectedly, the ratio of cost/technology changes constantly with new developments (Foorginezhad et al., 2021).

Additionally, there are other sensor performance metrics which have different requirements. For hydrogen safety sensors NREL, DOE and automobile manufacturers envisioned several technical performance requirements which are presented in Figure 6.



Parameter	NREL/DOE report for, respectively, automotive fuel Cells – on-board safety Sensors and fuel cell-powered industrial trucks (forklifts)	DOE requirements (automotive applications)	Performance requirements from manufacturers with Comments	
Measuring range	0 to 4 vol% H_2, may be extended to 10% hydrogen 0-4 vol% H_2	0-4 vol% H_2 in the air; survivability at 100%	Up to 4 vol% H_2 min; survivability at 100%	One car manufacturer stressed the need for a wider (unspecified) detection range
Detection limit	<0.1 vol% <0.04 vol% (1% of LFL)	<0.1 vol%	<0.1 vol%	Some car manufacturers accepted a measuring limit of <0.2 vol%
Response time (t_{90})	<30s (<1s at 1 vol% of H_2 preferred) <10 s (<1s at 1 vol% of H_2 preferred)	<3 s	<1 s	Some car manufacturers accepted <3 s
Recovery time (t_{10})	<60s <30 s	<3 s	<1 s	One car producer found <3s sufficient while another accepted <30 s
Ambient temperature range	−40 °C to +40 0 to +40 °C (or −40 °C for freezer storage areas)	−40 to +125 °C	−40 °C - +85 °C	Some producers set an operating temperature range of −40 °C to +120 °C for sensors exposed directly to operating temperature of an internal combustion engine
Ambient humidity range (Relative humidity range)	5%–95% 15%–95%	0–100%	0–95%	One manufacturer set 0–100%
Ambient pressure	0.6–1.1 bar Must be calibrated for deployment altitude	–	62–107 kPa	Pressure is related to a required altitude range of −400 to 4000 m
Power usage	Preferred moderate power requirements (<0.01 W) Preferred moderate power requirements (<0.5 W)	–	<1W	Some manufacturers required <650 mW
Lifetime	5–10	3–5 years	6000h	Discontinuous operation was deemed sufficient by all car producers. One set a goal of 15 years.
Overall accuracy	±20% of reading for all working conditions	10%	±5% of reading	Some car manufacturers proposed to replace it with a value of 5–10% attributed to a lower flammability limit (LFL)

Figure 6. Technical performance requirements envisioned by NREL, DOE, and automobile manufacturers for hydrogen safety sensors (data is received from automotive manufacturers as part of the EU's Sixth Framework Program Integrated Project – StorHy) (Foorginezhad et al., 2021)

3.6. Hydrogen Delivery by Tube Trailers

Hydrogen gas produced at a central plant is transmitted to a distribution terminal through a pipeline. Then it is compressed and transported on tube trailers to fuelling stations from distribution terminals. Tube trailers are trucks used to transport compressed gaseous hydrogen (20-50 MPa) stored in tubes bundled together in 20 or 40-feet long containers on top of it (Hjeij et al., 2022). The weight of the tubes limits the amount of hydrogen that can be transported via truck. Therefore, lighter composite materials have been developed to carry more hydrogen at higher pressures (Hjeij et al., 2022). Five types of pressure vessels can be used for storing hydrogen (Reddi et al., 2018).

- **Type I:** Fully metallic pressure vessels (Moradi & Groth, 2019). This type is the most conventional, least expensive, and also heaviest with approximately 3.0 lb/L (Moradi & Groth, 2019). They are normally made from aluminium or steel and can contain pressures up to 50 MPa (Moradi & Groth, 2019).
- **Type II:** Steel pressure vessel with a glass fiber composite overwrap (Moradi & Groth, 2019). The steel and composite material share about the same amount of structural load. Manufacturing Type II vessels costs about 50% more than Type I, but they offer 30-40% less weight (Moradi & Groth, 2019).
- **Type III:** Full composite wrap with metal liner (Moradi & Groth, 2019). The structural load is mainly carried by the composite structure (carbon fiber composite) and the liner (aluminium) is for sealing purposes (Moradi & Groth, 2019). In this type of pressure vessel, the metal liner shares about 5% of mechanical load (Moradi & Groth, 2019). This type of pressure vessel has proven to be reliable for 45 MPa working pressure but still has problems with passing the aging tests at 70 MPa (Moradi & Groth, 2019). Type III provides 0.75-1 lb/L weight, which is about half of the type II, but their cost would be twice the cost of Type II (Moradi & Groth, 2019).
- **Type IV:** Fully composite (Moradi & Groth, 2019). Commonly a polymer like High Density Polyethylene (HDPE) is used as liner and carbon fiber or carbon-glass composites are used for carrying the structural load (Moradi & Groth, 2019). This type of pressure vessel is very light yet again the price is still relatively very high. Type IV pressure vessels can withstand pressures up to 100 MPa (Moradi & Groth, 2019).
- **Type V:** Linerless fully composite pressure vessel (Reddi et al., 2018). This is the lightest but most expensive one currently developed. It is relatively a new technology which is approved for storage of gases, but it has not been tested for hydrogen storage (Reddi et al., 2018).

Tube trailers can carry 720-1350 kg of hydrogen in a single trip depending on the pressure inside the tube (25-54 MPa) (Hjeij et al., 2022). Usage of tube trailers is of interest because it is the simplest method in terms of infrastructure requirements. Moreover, the history behind gaseous storage and transportation of other gases provides ample knowledge about the physics involved. Another advantage is that hydrogen loss is minor and compression cost at fuelling stations is low and can be further reduced by 60% in comparison with liquid hydrogen (LH_2) transportation (Moradi & Groth, 2019). Transportation of hydrogen gas via tube trailers has been under attention by DOE and its collaborators from several years ago. HEXAGON Lincoln has developed a high-pressure tube trailer, called TITAN (Hexagon Composites, 2015). The operating pressure of these tanks is 25 MPa and the total capacity of a TITAN is 616 kg of hydrogen considering the mass of hydrogen stored in large tanks is approximately 7% of the tank weight (Hexagon Composites, 2015). Figure 7 shows an evolution of tube trailers from steel bottles to composite tubes leading to higher amount of compressed gas transportation in less costs (Hexagon Composites, 2015). Although, the figure is representative of storage of compressed natural gas (CNG), they can be applied in case of CGH_2 as well. For example, the TITAN4 can be used to store either 364000 cubic feet (scf) of CNG or 617 kg of compressed gaseous hydrogen (Hexagon Composites, 2015). Moreover, TITANT5M can contain 802 kg of CGH_2 (Hexagon Composites, 2015).

Structurally, four types of tube trailers are used for transportation of CGH_2.

- Tubes without roofs and with no box with ports and connections
- Tubes without roofs and with box with ports and connections
- Tubes inside containment with roofs and with no box with ports and connections
- Tubes inside containment with roofs and with box with ports and connections

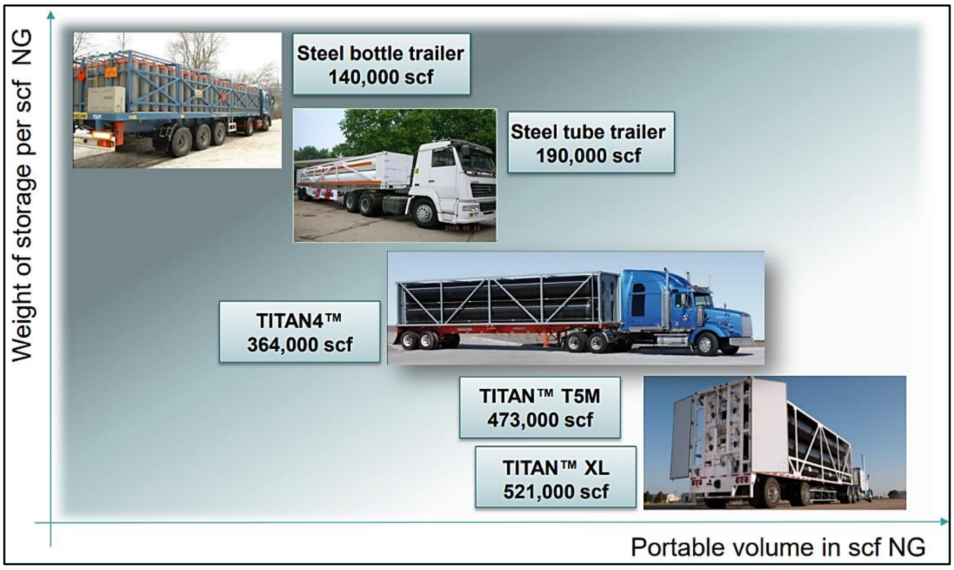


Figure 7. Evolution of tube trailers regarding usage of pressure vessels (abbreviations: scf: cubic feet, NG: Natural Gas) (Hexagon Composites, 2015)

3.7. Recommended Sensor System

From the information presented in the previous sections, a sensor system has been attempted to be developed suitable for using in different configurations of the tube trailers. Indication of the sensors and their locations and numbers have been selected keeping their usage in tube trailers in mind. However, they are subject to change when being used in different applications.

3.7.1. Sensors

From the above stated hydrogen sensors, TC sensors are most widely used and most appropriate for automotive purposes (Foorginezhad et al., 2021). However, due to the application being outdoors, it is highly probable that the leaked hydrogen may not come in contact with the sensor due to the outdoor air flow and dissipate in the wind (Fecarotta & Janowski, 2021). Therefore, for extra layers of protection consisting of UGLD sensors, pressure gauges and flame sensors are recommended. UGLD sensors can sense the ultrasonic sound generated due to leakage of compressed gaseous hydrogen. But there might be different noise levels in different areas outdoor which may hinder the detection of ultrasonic sound as well. So, pressure gauges can provide added layer of monitoring. Because in case of a leakage, the pressure inside the hydrogen storage will decrease whether the sensors can detect it or not. So, reduction in pressure shown by pressure gauges can provide indication of probable leakage.

Moreover, hydrogen flames caused by leaked hydrogen are scarcely visible. Thus, flame detectors around the storage system are also needed in case of flames according to ISO 15916. Temperature around the storage system could be increased for other reasons than the hydron flames as well. This may lead to the increase in pressure of the compressed gaseous hydrogen which can potentially result in accidents like rupture of tank or other hazardous events. Therefore, temperature sensor can be used for providing necessary safety information instead of detecting leakage.

Although, there was a mention of portable hydrogen detectors around the storage system in ISO 15916, this can be omitted in the sensor system due to the nature of application being in outdoors. But the truck driver or the workers that will be dealing with the hydrogen storage and the truck can use the portable detectors while working because it can notify them in case of leakage of hydrogen from the truck or accumulation of hydrogen due to other reasons. It would enable them to take necessary safety precautions and avoid any unwanted consequences.

3.7.2. Number of Sensors

The number of hydrogen sensors depend on the area of hydrogen storage needed to be monitored and the coverage distance of each sensor. So, increased area of coverage would require increased number of sensors. Also, if there are more joints like welding, ports or bends in piping, more sensors may be required. Moreover, more sensors are required if a redundant sensor system is aimed to build.

3.7.3. Locations of Sensors

The position of the hydrogen sensors can be determined by taking in consideration the structure and the type of storage used in trucks along with the properties of hydrogen. Hydrogen tends to move upwards due to being light, so point sensors should be located in the upper locations of the truck. Therefore, if there is a roof over the hydrogen cylinders, the hydrogen sensors can be located beneath the roof over the cylinders where hydrogen is likely to accumulate. Also, sensors should be positioned in weak locations like joints like welding, ports or bends in piping according to ISO 15916. Thus, hydrogen sensors should be positioned in the top of the box with connections and ports to detect leaked hydrogen.

UGLD sensors can be positioned at the middle point of each side of the tubes as it would provide sufficient coverage of leak detection. Pressure transducers can be positioned at any point inside in each of the cylinders and provide measure of pressure reduction in case of leakages. Flame sensors can be located keeping their field of view and coverage capacity in mind. Flame sensors can be located at two extremities of the tube on each side to cover the whole area of the cylinders. Temperature sensors can be positioned in different points in optimal distances so that they can provide full coverage of the area and the temperature can be sensed instantaneously from any point.

3.7.4. Detection Device

Adequate alarm systems are necessary along with proper detection for providing understandable indications of leakage or accumulation of hydrogen. Furthermore, a display containing all the necessary information about the sensors together would allow the system user to learn about the condition of the system easily and take actions immediately as per the displayed information. Also, inherent safety barriers in hydrogen systems can be useful in avoiding dangerous accidents as well. Taking all these into account and inspiration from SBS-H2-DoD detector presented before, a detection device using the sensors mentioned in the previous section has been suggested.

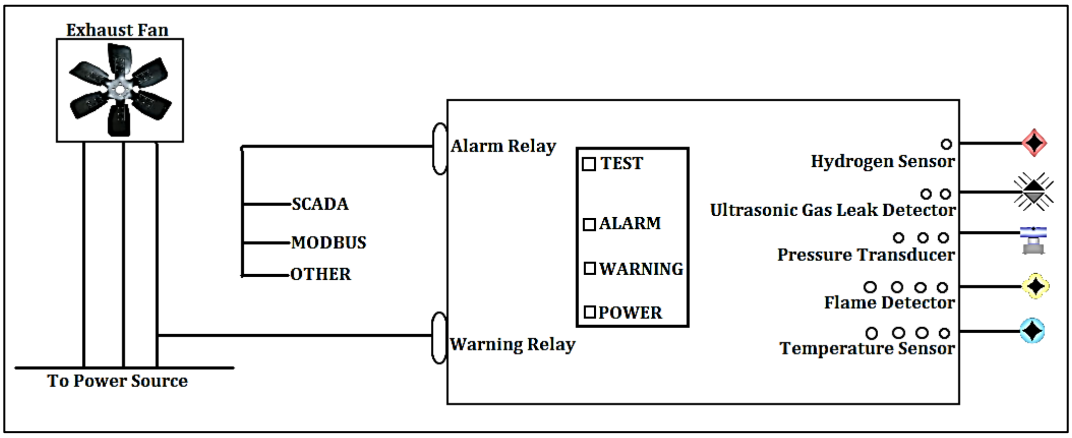


Figure 8. Suggested hydrogen detection device

Different sensors have been suggested to provide a robust detection system in case of leakage. All of the sensors can be connected together in the device showed in Figure 8. In place of 3 hydrogen sensors shown in SBS-H2-DoD detector, hydrogen sensors, UGLD, pressure transducers, flame detectors and temperature sensors can be added with adequate alarm system for each. Visual alarm with LED light for each of the sensors implemented in the system has been depicted in the device. Audible alarms should be implemented along with the visual alarms.

Hydrogen sensors would provide a warning before the hydrogen concentration reaches 4.1% vol in air. Different coloured LED lights can be allocated to be lit up at certain concentration of hydrogen to differentiate type of accumulation of hydrogen and take necessary steps accordingly. For example, yellow 'WARNING' and red 'ALARM' LED can be assigned respectively along with the LED assigned for each hydrogen sensors for 1 % vol and 2 % vol detected hydrogen concentration level.

UGLD sensors would provide indication of leakage in any part of the tubes. The red 'ALARM' LED along with the LED assigned for UGLD would indicate detection of leakage in the tubes. Pressure transducers would depict either the rise in pressure due to temperature increase or decrease in pressure due to leakages in the tubes. The red 'ALARM' LED along with LED for pressure transducers could be activated due to the reduction in the pressure from a predefined value. The yellow 'WARNING' LED along with LED for pressure transducers would be activated due to the increase in the pressure from a predefined value. Flame detectors would demonstrate presence of flames. The red 'ALARM' LED along with LED for flame detectors could be activated in presence of flames. Temperature sensors would reveal external source of heat which might result in excessive pressure in the tubes or act as ignition source as well. The red 'ALARM' LED along with the LED for temperature sensors could be activated in a predetermined temperature.

The solid green LED for 'POWER' on the main control indicates that the detector is powered on. The 'TEST' button can be used to test the electronic circuitry of the device. Sensors can be tested using test kits provided with the device as suggested in SBS-H2-DoD detector (SBS, 2019). However, the electric circuitry testing and the sensor testing are not described in this study. For all the sensors, audible alarms can be triggered along with visual alarms. The predefined values of pressure and temperature mentioned above depend on the type of application. They depend on the structural parameters of the tubes used in the trailer and the desired safety from the hydrogen system integrators.

In addition to the alarm system, safety barriers triggered automatically upon the hydrogen concentration reaching dangerous levels have been included. When 'WARNING' LED is turned on, the Warning Relay is powered, and exhaust fans can be powered on automatically due to this. Several exhaust fans placed strategically in different parts of the tube trailer can help to avoid accumulation of hydrogen. When the 'ALARM' LED is turned on, the Alarm Relay is activated along with the Warning Relay and emergency responders are automatically contacted. Additional relays could be added as per requirements to increase the safety of the system. For example, in case of activation of LED of flame detectors, a relay connected to water sprinklers or other adequate flame extinguishing systems could be activated to put out the flame.

Through a combination of the sensor systems, proper alarms and automatically triggered safety barriers in required situations, the device can successfully avoid dangerous consequences while transferring compressed gaseous hydrogen using tube trailers.

3.8. Application of Recommended Sensor System for Tube Trailers

The sensor system developed in the previous sections has been applied for the four different structures of tube trailers and the applications have been depicted below. Specific suggestions for sensor types selected and their specifications are presented in Table 2. However, different sensors apart from the suggested ones can be selected as well. Tube trailers manufactured by City Machine and Welding Inc containing 12.192 m long tubes have been adapted for this application (City Machine and Welding, 2022).

- Hydrogen sensors are required for detecting accumulated hydrogen in case of leakages. In case of containments, hydrogen generally gather in the topmost places . Hydrogen accumulation can occur in two places in the trailer, on top the box containing connections and ports, and under the roof containing the tubes. Therefore, hydrogen sensors are required on top of the boxes with connections and ports, and beneath the roof of where the congestion due to structure of the roof is high.
- UGLD sensors are required to detect ultrasonic sound generated due to the leakage of pressurized tubes. In each side of the tubes, one UGLD sensor positioned centrally is required so that the ultrasonic sound can be detected from any part of the tubes.
- Pressure inside the tubes can be sufficiently detected with one pressure transducer in each of the tubes.

- Although the measurement range of flame detector selected is high, there might be blind spots if only one flame detector is used. So, two flame detectors at the extremities of each side of the tubes enable detection of flames generated in each side.
- Temperatures sensors should be positioned in such a way that they provide instantaneous coverage of the whole area in case of temperature fluctuation. Two sensors at an optimal distance in each side of the tubes are required for this.

Table 2. Suggestions regarding sensor selections and their specifications

Sensor Type	Sensors	Specifications
TC Sensor	XEN-5320 (Xensor NL, 2020)	Measurement Range: 100 pp to 100% (Xensor NL, 2020), Response Time: <1s (Xensor NL, 2020), Temperature Range: -40 to 85 °C (Xensor NL, 2020)
UGLD	Incus Ultrasonic Gas Leak Detector (Emerson, 2022)	Instantaneous response to all gas leaks, Measurement Range: 2 to 40 m (Emerson, 2022), Temperature Range: -40 to 85 °C (Emerson, 2022)
Pressure Transducer	WIKA manufactured transducer (Ortiz Cebolla et al., 2019)	Measurement Range: 0 to 100 MPa (Ortiz Cebolla et al., 2019)
Flame Detector	SMC 3600-M (Sierra Monitor Corporation, 2010)	Measurement Range: 30m (Sierra Monitor Corporation, 2010), Response Time: 5s (Sierra Monitor Corporation, 2010)
Temperature Sensor	Thermocouples type K (Class 1) (Ortiz Cebolla et al., 2019)	Measurement Range: -270 to 1300 °C (Ortiz Cebolla et al., 2019)

3.8.1. Roofless Truck with Box Containing Connections and Ports

The numbers and the locations of the sensors are as following.

- Hydrogen Sensor: 1, Top of the box with connections and ports
- UGLD: 2, Each in Middle point of each side of cylinders
- Pressure Transducer: 3, Each inside one cylinder
- Flame Detector: 4, 2 in two extremities of each side of cylinders
- Temperature Sensor: 4, 2 in each side of cylinders at optimal distance

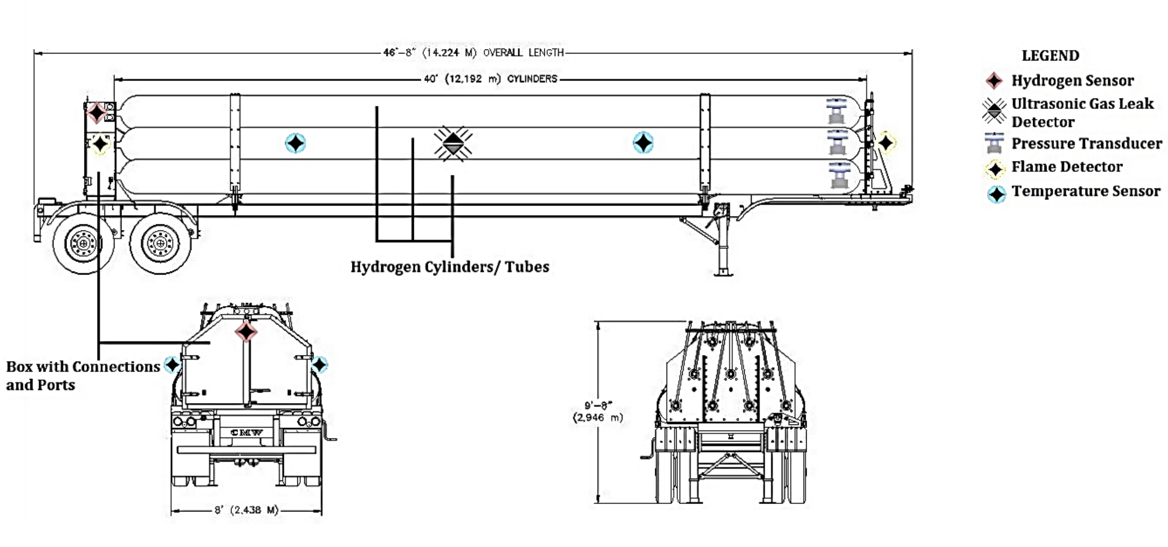


Figure 9. Schematic of the recommended sensor system in a roofless tube trailer [adapted from (City Machine and Welding, 2022)**]**

3.8.2. Truck with Roof and Box Containing Connections and Ports

The number and the locations of the sensors are as following.

- Hydrogen Sensor: 4, 1 on top of the box with connections and ports, 3 below the roof with highest congestions
- UGLD: 2, Each in Middle point of each side of cylinders
- Pressure Transducer: 3, Each inside one cylinder
- Flame Detector: 4, 2 in two extremities of each side of cylinders
- Temperature Sensor: 4, 2 in each side of cylinders at optimal distance

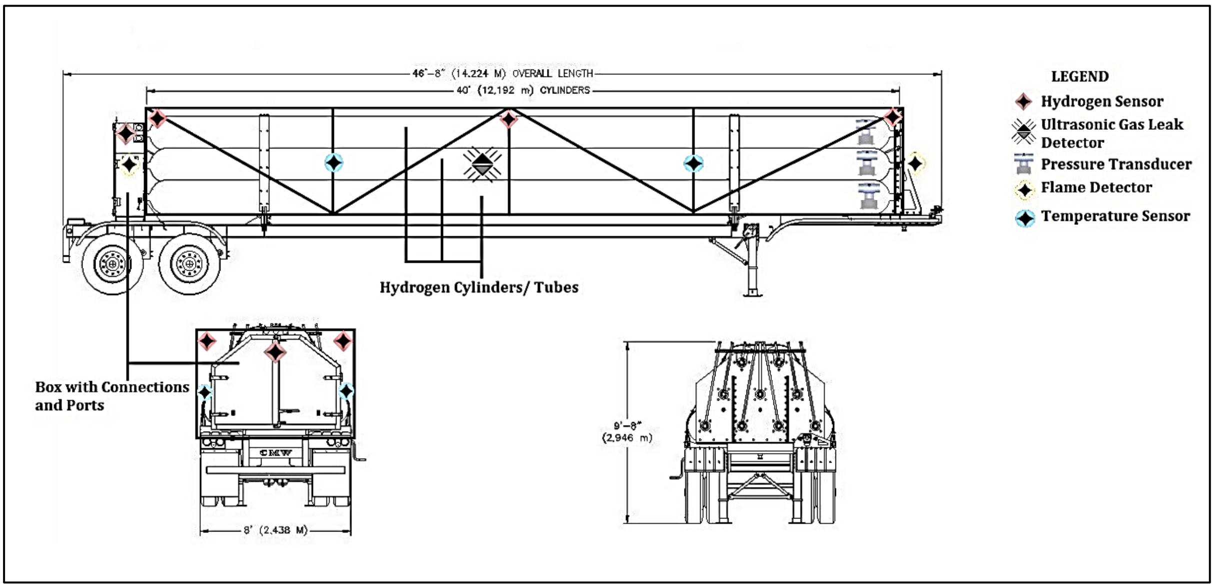


Figure 10. Schematic of the recommended sensor system in a tube trailer with roof [adapted from (City Machine and Welding, 2022)**]**

3.8.3. Trucks Without Box Containing Connections and Ports

For the trucks without the box with connections and ports, only the one hydrogen sensor inside the box can be omitted. Rest of the designs are applicable for those two types of trucks.

3.9. Identification of important parts and safety indicators of the risk control system

Critical elements of RCS: Fault free sensors.

Lagging Indicators:

- Number of accidents caused by leakage of hydrogen due to failure of detection of hydrogen.
- Number of false positive alarms by sensor system

Leading Indicators:

- Periodic inspection for ensuring proper functioning of sensors comparing with performance requirements. Maintenance staff should review the inspection list and make sure the entire inspection plan is completed and documented as scheduled (H2Tools, 2022).
- Maintenance and recalibration of leak and flame detectors should be performed periodically following a well-planned preventive maintenance schedule (typically every 3-6 months) or as recommended by the manufacturer (H2Tools, 2022).

- Documentation of implementation of sensors and equipment with proper acceptance criteria (ISO, 2013).
- Proper inspection plan containing suitable tests mentioned in Sec 3.5.1.4. of this report against requirements set by the organization.

4. Discussion

A leakage detection device containing sensor system, alarms and safety barriers was developed and suggested in this study for proper detection of leakage of CGH_2 while transportation with tube trailers. As per methodology, performance requirements from the standards and different materials, state of the art of the hydrogen sensors in practice and state of the art of the tube trailers were studied. The information gathered acted as input for the development of the sensor system. However, although the scope of the study concerns with the storage during mobile applications of hydrogen, no standards were found regarding mobile applications of hydrogen. Therefore, indications are taken from the standards developed for stationary applications and modified accordingly. Moreover, the locations of the sensors mentioned in ISO 15916 are provided for the fuel cell equipment, not for the bulk storage applications; still these indications were adapted for this study.

Regarding the hydrogen sensors, it was found that no perfect hydrogen sensor exists which can fulfil all the requirements simultaneously for all the applications. Therefore, from the several discussed hydrogen sensors, the most suitable one for this study was selected to be TC sensors. However, for different applications, different hydrogen sensors can be selected and applied as per requirements. As the application in this study is outdoor, it is highly probable for the leaked hydrogen not to come in contact with point hydrogen sensors. Therefore, additional sensors like UGLD, pressure transducers, flame detectors and temperature sensors are included in the sensor system to increase the likelihood of leakage detection and prevent loss of containment of hydrogen. But pressure transducers, flame detectors and temperature sensors were not discussed in this study as they are well developed technologies and have been in practice for long time. Only indications regarding them which are appropriate for this study were adapted for this application. These indications might be different if the application is different to fulfil the requirements. Also, portable hydrogen sensors were not focused on this study although they are suggested in ISO15916. But they can be included to increase the safety of the system users. For example, the system users may be instructed to leave a location if the hydrogen concentration indicated by the portable hydrogen sensors reaches a pre-defined limit.

Hydrogen tube trailers were studied and categorized in this study as well. State of the art of the hydrogen tube trailers were presented to learn about the application field of the sensors. Depending on the structure used for containing the hydrogen storage tanks, the tube trailers were divided in four categories. This categorization helped to show difference between the sensor system required for different structures through application of the developed sensor system in each of them. The main reason between this difference is that different structures lead to different types of accumulation of hydrogen which require different detection strategies. All the strategies have been depicted in this report. Required strategy as per requirements of the tube trailers used can be adopted from them.

Although this study mainly aims at leakage detection of hydrogen, some safety barriers were suggested as well. A detection device was developed taking inspiration from SBS-H2-DoD detector. The detection device enables easy notification and swift actions in case of a leakage. Two safety barriers are included in the detection device with two relays. One relay turns on the exhaust fans automatically and prevents accumulation of hydrogen. The other one enables the automatic contact in case of emergency. Design of additional safety barriers for each type of sensors can provide future scope of work and be included in the tube trailers for increasing safety. Apart from these safety barriers enabled by the detection device, an inherent safety barrier can be provided by modifying the roof of the hydrogen tube trailers. Instead of placing the roofs horizontally, if the roofs are tilted, it can facilitate accumulation of hydrogen in one location instead of several places in tube trailer. This can lead to decrease of required number of hydrogen sensors placed. Also, the exhaust fans switched on by the tube trailers can be placed strategically so that the accumulated hydrogen in one location can be easily blow away and dispersed in the wind without any serious consequences. However, this needs to be done in such a way that it does not modify the truck aerodynamics and hinders its movement.

A modification of the guideline suggested by ‘HSG254: Developing Process Safety Indicators’ guide was adopted for the sensor system to ensure its proper functioning. Lagging and leading indicators were selected such that they provide dual assurance regarding the performance of the sensor system. One of the leading factors include periodic inspection of leak and fire detectors. Different testing method of sensors include chamber tests and flow tests. In these tests, the sensors are individually placed in the experimental setups. However, in the application of this study, sensors are required to be tested on board where the tests suggested in SAE J3089 report could be adopted. But the testing method using the test kits of SBS-H2-DoD detector can be useful in this regard as well. Moreover, future studies can be directed in this case to find out optimum and easy testing of on-board sensors without needing to take them off the tube trailer.

This study is concerned with the transportation of CGH_2 with tube trailers. Apart from tube trailers, CGH_2 can be transported using pipelines which have not been studied in this report. Moreover, transportation of LH_2 has not been studied in this report. None of the sensors studied in this report mentions the use in LH_2 detection. Sensor system for detection of release of LH_2 can be an important future study to solve complex issues related with LH_2 transportation. Furthermore, development of sensor system for transportation of compressed gaseous hydrogen using pipelines can be investigated in the future as well.

While developing the sensor system, it was not considered whether the vehicle was fuelled by hydrogen or other fuels like gasoline. The difference between a vehicle fuelled by hydrogen and gasoline is the state of the fuel. Gasoline is a liquid and does not require compressed storage system like hydrogen. Therefore, the hazards related to a compressed fuel storage system does not comply with gasoline. Furthermore, the fuel system of the hydrogen fuelled cars need to be modified to avoid leakage of hydrogen. Therefore, sensors are required in the fuel system if hydrogen fuelled cars along with the tubes transported. Moreover, difference in global warming emissions between the two fuels can be noted as well (UCSUSA, 2014). Emissions are reduced compared to gasoline while using hydrogen (UCSUSA, 2014). Emissions depend on how the hydrogen fuel is made and delivered. Currently, most hydrogen is made by converting natural gas into hydrogen gas and carbon dioxide (UCSUSA, 2014). However, hydrogen can also be produced from sources of energy that are lower in carbon than natural gas. For example, by splitting water using electrolysis using electricity from solar or wind power. Even while using hydrogen produced from natural gas which contains higher amount of carbon, the emission is reduced by 30% (UCSUSA, 2014). The cleaner the hydrogen, the higher the reduction in emissions (UCSUSA, 2014). If there is more emission, the difference in the sensor system could be caused due to requirement of higher selectivity of the sensors to hydrogen in presence of other gases. Therefore, for vehicles fuelled by other fuels with higher emissions, sensors with high selectivity are required. Whereas for vehicles powered by hydrogen, sensors would be less affected by other gases and requirement regarding selectivity is less stringent. More research can be directed in the future between different fuels to get more information which would help to develop the sensor systems.

Using leakage detection system and implementation of indicators to ensure its performance represent a commitment to the safe use and handling of hydrogen. But no information resource can provide 100% assurance of safety. Engineers with applicable expertise should always be consulted in designing and implementing any system carrying a potential safety risk.

5. Conclusion

This study was aimed at developing a leakage detection system in case of transportation of compressed gaseous hydrogen with tube trailers. Through analysed information from current standards in place for hydrogen applications, state of the art of hydrogen sensors and tube trailers, a sensor system was developed indicating the type of sensors applicable, their required numbers and strategic positions. This sensor system was applied for different structures of tube trailers which indicates adaptability of the system as per requirements. However, lack of relevant standards for transportation of hydrogen in mobile applications were noticed. Therefore, indications were taken from current standards applicable for hydrogen equipment and stationary applications. The sensor system consisted of different sensors like hydrogen sensors, UGLD, pressure transducers, flame detectors and temperature sensors. This provided multiple layers of detection and avoiding loss. However, no perfect hydrogen sensor was found. Sensors were selected due to their suitability in the studied application. of containment scenarios. Combining alarm system and safety barriers with the developed sensor system, a detection device was suggested taking inspiration from SBS-H2-DoD detector. The alarm system enables swift action from system users in case of leakages or other potential unwanted scenarios. Furthermore, two safety barriers were provided which were connected to the device with two relays and are automatically enabled in case of emergency. Moreover, a modification of 'HSG254: Developing Process Safety Indicators' guide was implemented to ensure adequate performance of the sensor system. Lagging and leading indicators were developed and expected to provide dual assurance regarding the functioning of the sensor system. However, these indicators may be subject to change in the future through proper review if they do not provide satisfactory performance. The developed leakage device and the application of guideline is expected to provide safety assurances by proper detection of leaked hydrogen and avoiding loss of containment. But 100% safety cannot be assured through information resources. Consultation of expert personnel during the design and implementation of systems and utmost carefulness regarding the applications can reduce the risks to acceptable levels.

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