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Probability of occurrence of ISO 14687-2 contaminants in hydrogen: Principles and examples from steam methane reforming and electrolysis (water and chlor-alkali) production processes model

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ARTICLE INFO

Article history:

Received 9 November 2017

Received in revised form

11 March 2018

Accepted 12 March 2018

Available online 4 April 2018

Keywords:

ISO14687-2

Hydrogen quality

Fuel cell electrical vehicle

Probability of occurrence

Hydrogen production process

ISO 19880-8

ABSTRACT

According to European Directive 2014/94/EU, hydrogen providers have the responsibility to prove that their hydrogen is of suitable quality for fuel cell vehicles. Contaminants may originate from hydrogen production, transportation, refuelling station or maintenance operation. This study investigated the probability of presence of the 13 gaseous contaminants (ISO 14687-2) in hydrogen on 3 production processes: steam methane reforming (SMR) process with pressure swing adsorption (PSA), chlor-alkali membrane electrolysis process and water proton exchange membrane electrolysis process with temperature swing adsorption. The rationale behind the probability of contaminant presence according to process knowledge and existing barriers is highlighted. No contaminant was identified as possible or frequent for the three production processes except oxygen (frequent for chlor-alkali membrane process), carbon monoxide (frequent) and nitrogen (possible) for SMR with PSA. Based on it, a hydrogen quality assurance plan following ISO 19880-8 can be devised to support hydrogen providers in monitoring the relevant contaminants.

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<https://doi.org/10.1016/j.ijhydene.2018.03.084>

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Introduction

Hydrogen production and use as an energy vector is encouraged to accelerate the decarbonisation of the transport sector [1,2]. Hydrogen is a clean and storable solution to the European policy objectives in the transport and energy sectors defined in the Energy roadmap 2050 (COM(2011) 885 final) [3] from the European Parliament. Moreover several countries worldwide have initiated national policies to rapidly introduce hydrogen technologies to the market over the next few decades.

The demand for hydrogen fuel cell electric vehicles (FCEV) is clearly growing across Europe [4]. In the UK it is expected that there could be 1.6 million fuel cell vehicles on the road by 2030 supported by 1100 hydrogen refuelling stations (HRS) [5]. Germany planned 100 HRS by 2018/2019 and 400 HRS by 2023 [6]. One potential barrier that could prevent the uptake of hydrogen technologies in Europe is European Directive 2014/94/EU on the deployment of an alternative fuels infrastructure [7] which sets out that “The hydrogen purity dispensed by hydrogen refuelling points shall comply with the technical specifications included in the ISO 14687-2 standard.” According to this Directive it will be the responsibility of the hydrogen supplier or system integrator to obtain the evidence that proves that their hydrogen is of suitable quality to be provided to a fuel cell vehicle. This involves reliable quality testing and quality assurance to show that none of the threshold limits for the 14 contaminants specified in ISO 14687-2 [8] are exceeded.

The Multi-Annual Work Plan of the fuel cells and hydrogen joint undertaking (FCH-JU) for 2014–2020 [9] points out that Hydrogen gas quality assurance at the nozzle (350 or 700 bar) still constitutes a technical challenge, due to the very stringent requirements (compounds and amount fraction) in hydrogen fuel gas for FCEVs. Currently, no simple methodology or single instrumentation is available for low cost qualifications of hydrogen fuel [9]. A recent literature review by Murugan and Brown [10] compiled all known gas analysis methods for performing quality analysis of fuel cell hydrogen. According to this review, a minimum of eight different instruments are required to determine the 14 contaminants specified in ISO 14687-2. The cost and the current lead time associated with the hydrogen quality analysis are unsustainable to support the emerging hydrogen sector. Today, harmonised RCS (regulations, codes and standards, ed.) and PNR (prenormative research, ed.) to fill RCS knowledge gaps at the European (and world) level are still recognised as a major requirement for the commercialisation of fuel cells and hydrogen products.

Hydrogen contaminant behaviour is clearly an important topic addressed recently due to the lack of analytical methods to measure the 13 gaseous compounds mentioned in ISO 14687-2 [8]. Understanding the complexity and the cost of the complete analysis of 13 gaseous compounds, a quality assurance approaches was considered to mitigate the needs of complete analysis.

Contaminants may originate from various sources; from the hydrogen production process, transportation, refuelling station infrastructure or maintenance operation along the supply chain. In fact, sampling and analysing at the hydrogen refuelling station nozzle will determine if hydrogen is compliant to the ISO 14687-2 requirements however it will not

allow to discriminate or identify the origin of a contamination. Recent European project Hydrogen Contamination Risk Assessment, FCH-JU Grant agreement #621223, 2014–2017 analysed hydrogen from European refuelling stations (at the nozzle) but did not achieve to identify contaminants origin [11].

An hydrogen contaminant risk assessment based on the probability of presence of the contaminant from the hydrogen production to delivery chain and on the severity of the contaminant for the FCEV systems is promoted in both ISO standard (ISO/DIS 19880-8 to be published in 2018 [12] and EN 17124 [13] for hydrogen quality assurance. A report from Pratt et al. [14] proposed a first matrix for hydrogen quality versus station contaminant considering complete systems (feed-stock: alkaline, water, chlor-alkali, natural gas, methanol, ethanol, gasoline; the purification: pressure swing adsorption or drying). However this study did not consider the barrier existing in a real production system. A more systematic approach was needed in order to understand the potential contamination from each part of the system. The first part to investigate was the hydrogen production process.

To support the development of hydrogen energy without compromising the requirement on hydrogen quality, the risk assessment approach proposed in ISO 19880-8 is under study in a Joint Research Project (JRP) from the European Metrology Programme for Innovation and Research (EMPIR) *Metrology for sustainable hydrogen energy applications* [15] through a coupled study on the probability of contaminant presence with a contaminant effect on fuel cells. The combination of both studies enables the determination of the most prominent parameters to be monitored based on its probability of presence and on its effect to the fuel cell system [16–19]. A parallel approach on fuel quality assurance was considered to mitigate the needs of complete analysis by developing an hydrogen contaminant risk assessment based on the probability of presence of the contaminant from the hydrogen production to delivery chain and the severity of the contaminant for the FCEV systems in ISO/DIS 19880-8 [12].

The first step assessed the probability of presence of the 13 gaseous impurities listed in ISO 14687-2 [8] in hydrogen from the three hydrogen production processes: steam methane reforming (SMR) process with pressure swing adsorption (PSA) as purification system, electrolysis of aqueous sodium chloride in a membrane cell, referred as Chlor-alkali process in this article and the water proton exchange membrane (PEM) electrolysis process with temperature swing adsorption (TSA) as purification system. The objective was to present the study and the rationale of contaminant presence in the hydrogen at the model production site. Process knowledge and analytical results have been combined. The determination of the probability of contaminant presence at the production was based on the process knowledge. The analytical measurement should be used as evidence to confirm the process assessment.

This study aims at describing the methodology of quality risk assessment used to assess the probability of occurrence of each contaminant identified in the three different production processes. This paper also investigates the various existing barriers in the current process and on line analysis in the process. The paper will provide some comments on other production methods as alkaline electrolysis even if it was outside of the scope of the project.

Materials and methods

Risk assessment

The evaluation of the risk of non-compliance to quality requirements should be based on scientific knowledge and ultimately link to the protection of the FCEV. To clearly define the risk(s) for risk assessment purposes, three fundamental questions were used:

- What might go wrong: which event can cause the impurities to be above the threshold value?
- What is the likelihood (probability of occurrence expressed relative to the number of refueling events) that impurities can be above the threshold value?
- What are the consequences (severity) for the FCEV? [12]

In this study, the risk assessment was limited to three production methods and did not consider the consequence (severity) for the FCEV that requires additional study to understand the severity of compounds like halogenated or ammonia [ref].

Table 1 – Contaminant specification maximum thresholds of the ISO standard 14687-2 [8].

Characteristics (assay)	Type I, Type II Grade D
Hydrogen fuel index (minimum mole fraction) ^a	99.97%
Total non-hydrogen gases	300 $\mu\text{mol mol}^{-1}$
Maximum concentration of individual contaminants	
Water (H ₂ O)	5 $\mu\text{mol mol}^{-1}$
Total hydrocarbons (methane basis) ^b	2 $\mu\text{mol mol}^{-1}$
Oxygen (O ₂)	5 $\mu\text{mol mol}^{-1}$
Helium	300 $\mu\text{mol mol}^{-1}$
Total Nitrogen (N ₂) and Argon	100 $\mu\text{mol mol}^{-1}$
Carbon dioxide (CO ₂)	2 $\mu\text{mol mol}^{-1}$
Carbon monoxide (CO)	0.2 $\mu\text{mol mol}^{-1}$
Total sulphur compounds (H ₂ S basis) ^c	0.004 $\mu\text{mol mol}^{-1}$
Formaldehyde (HCHO)	0.01 $\mu\text{mol mol}^{-1}$
Formic acid (HCOOH)	0.2 $\mu\text{mol mol}^{-1}$
Ammonia (NH ₃)	0.1 $\mu\text{mol mol}^{-1}$
Total halogenated compounds ^d (Halogenate ion basis)	0.05 $\mu\text{mol mol}^{-1}$
Maximum particulates concentration	1 mg kg ⁻¹

For the constituents that are additives, such as total hydrocarbons and total sulphur compounds, the sum of the constituents are to be less than or equal to the acceptable limit.

^a The hydrogen fuel index is determined by subtracting the “total non-hydrogen gases” in this table, expressed in mole percent, from 100 mole percent.

^b Total hydrocarbons include oxygenated organic species. Total hydrocarbons shall be measured on a carbon basis ($\mu\text{mol C/mol}$). Total hydrocarbons may exceed 2 $\mu\text{mol/mol}$ due only to the presence of methane, in which case the summation of methane, nitrogen and argon shall not exceed 100 $\mu\text{mol/mol}$.

^c As a minimum, total sulphur compounds include H₂S, COS, CS₂ and mercaptans, which are typically found in natural gas.

^d Total halogenated compounds include, for example, hydrogen bromide (HBr), hydrogen chloride (HCl), chlorine (Cl₂), and organic halides (R-X).

For each contaminant of the ISO 14687-2 specification and for a given hydrogen source, a dedicated risk assessment was realized. The objective was to identify the probability of occurrence to have each contaminant above the threshold values of specifications given in ISO 14687-2 (see Table 2) in the hydrogen at the end of the production process.

The possible cause of contaminant is established compounds per compounds based on technical knowledge of the process. It also takes into account the following:

- Existing barriers in the current process
- On line analysis in the process (i.e. dew point monitor)

For the probability of occurrence of the event: impurities in hydrogen exceed the threshold value, Table 2 summarizes the five levels of occurrence classes defined in the study.

A link between the occurrence or frequency and the occurrence in hydrogen refueling was done to provide an estimate of the event frequency for FCEV users.

In doing an effective risk assessment, the robustness of the data set is important because it determines the quality of the output. Revealing assumptions and reasonable sources of uncertainty will enhance confidence in this output and/or help identify its limitations. The output of the contaminant probability of presence is a qualitative description of a range of occurrence.

Steam methane reforming model system

The larger volume of hydrogen produced worldwide is accomplished by steam methane reforming [20]. In this process, methane from natural gas and steam react at high temperature to produce synthesis gas (or syngas). Syngas is a mixture consisting mainly of hydrogen and carbon monoxide [21].

In order to achieve the reaction between natural gas and steam, catalysts and high temperature are required. These catalysts are poisoned by any trace of sulphur compounds. It is then necessary to remove all sulphur components from natural gas before the SMR reaction [22]. The purification system, named hydrodesulfurisation (HDS) is a two steps process: first transformation of all sulphur species in H₂S and then adsorption of H₂S in specific adsorbents. At the outlet of this purification step, the natural gas contains less than 50 nmol mol⁻¹ of H₂S by design and less than 10 nmol mol⁻¹ in reality.

After the reforming reaction, the carbon monoxide is further reacted with steam in a water gas shift reaction. It produces carbon dioxide and hydrogen and it increases the hydrogen yield. Steam methane reforming has an easily manageable feedstock resulting in a product gas with a large hydrogen concentration in comparison with other hydrogen production processes using fossil fuels like coal gasification or O₂-blown autothermal reforming (Table 3) [15–20]. An additional separation step is mandatory to provide H₂ with a purity suitable for FCEV application.

Purification by Pressure Swing Adsorption (PSA)

Pressure swing adsorption (PSA) is a non-cryogenic gas separation process able to provide H₂ with a purity ranging from 99 to 99.999% [23]. PSA uses adsorbent technology to purify H₂ from a gas mixture. Membrane and cryogenic separations are

Table 2 – Definition of occurrence classes as proposed in the ISO/DIS 19880-8 [12].

Occurrence class	Class name	Occurrence or frequency at the production site	Equivalent occurrence or frequency for refueling event
0	Very unlikely (practically impossible)	Contaminant above threshold never observed for this type of source in the industry	Never
1	Very rare	Heard in the industry for this type of source/Supply chain considered	1 per 1000000 refueling
2	Rare	Has happened more than once a year in the industry	1 per 100000 refueling
3	Possible	Has happened repeatedly for this type of source at a specific location	1 per 10000 refueling
4	Frequent	Happens on a regular basis	Often

others techniques capable of purifying H₂ depending on the purity requirement; they can both achieve 98% purity [20]. However, for the purposes of producing H₂ with >99.99% purity for fuel cell applications [8], the best technology is adsorption [24]. In addition to PSA, temperature swing adsorption (TSA) can also be used to achieve even higher purities of 99.9999%, but excessive power is required to perform the cold cycles making it more expensive.

PSA principle is based on preferential adsorption of some gaseous components to others on highly porous materials [25]. These materials adsorb larger amounts of impurities at high partial pressure than at partial low pressure [20,26]. Thus, the column is fed with a high pressure feed gas containing impurities and the pressure is then lowered to regenerate and then to purge the column [20]. To reduce the partial pressure and desorb impurities, the adsorber pressure is swung from the higher feed gas pressure to lower tail gas pressure [20]. At least three columns are operating simultaneously due to the cyclic nature of the process. In fact each column performs the same cycle but the cycles are staggered in time. In practice, many more adsorption columns are used simultaneously to allow a constant hydrogen production.

The PSA ability to trap impurities depends on the affinity between the adsorbent and the gas molecule. Typically a PSA column is typically filled with multiple adsorbents with very high surface area to volume ratios [27]. Typical adsorbents include silica, alumina, molecular sieves and activated carbons, which have different relative strength of adsorption depend on the gaseous compounds (Fig. 1) [20].

A PSA unit will easily retain any of the compounds on the ++++ column in the Fig. 1, including hydrogen sulfide and

ammonia [20]. It will also retain the compounds in column +++ relatively easy, if the adsorbents are correctly selected [20]. The adsorbent selection allows the PSA unit to be adjusted to provide very low levels of CO and CO₂. However, the PSA unit will have difficulties retaining argon, oxygen and nitrogen due to similar affinity for the adsorbent material relative to hydrogen [20]. The concentrations of impurities in the product gas are not independent and depend on various factors such as the feed gas composition, the amount and composition of adsorbent and operation pressure. The optimisation of the hydrogen quality is possible but reducing the contaminant concentration causes a reduction in the H₂ yield and an increase in the adsorbent volume [20].

Water PEM electrolysis model system

Water PEM electrolyser model considered 4 mains parts:

- Raw material: H₂O (water purification unit for tap water purification) and power supply
- Electrolysis cell stack including the cell stack, the water and oxygen management system and the water and hydrogen management system)
- Purification system (drying system and TSA system)
- Power electronics system

Raw material

Tap water, used as raw material, is firstly purified by a reverse osmosis unit and stored in a dedicated tank at atmospheric pressure. This preliminary step is necessary to remove any contaminants and ions from the water before feeding the process with pure water (Conductivity < 0.1 μS/cm). Pure water is fed into the O₂ separator tank using a lift pump. The pump take pure water at atmospheric pressure at the inlet and feed the water to the separator tank at operating pressure (up to 14 bar).

Electrolysis cell stack

The electrolyser's power electronics system filters, controls, transforms, and switches the main AC power input to various components throughout the unit. Depending on the size of the generator, the AC input can be single-phase or three-phase power, low voltage (200–240 V) or high voltage (480–500 V), and 50 or 60 Hz. Typically, the input power is divided to provide low voltage 24 DC power for control valves, sensors, etc., and high power for the electrolysis cell stack.

Table 3 – Gas compositions at the outlet of the reactors for the three processes after CO₂ capture [20]

Component	Steam reforming	O ₂ -Blown ATR (autothermal reforming)	Coal gasification
H ₂	94.3%	93.2%	87.8%
CO	0.1%	1.4%	2.6%
CO ₂	2.5%	1.7%	3.9%
N ₂	0.2%	0.7%	5.0%
Ar	0	0.6%	0.9%
CH ₄	2.9%	2.4%	0.01%
T (°C)	33.3	35.0	30.0
P (bar a)	26.3	25.0	28.0
Q (Nm ³ ·h ⁻¹)	17318	17631	19402

RELATIVE STRENGTH OF ADSORPTION


+	++	+++	++++
<i>He</i>	<i>Ar</i>	<i>CO</i>	<i>C₃H₆</i>
<i>H₂</i>	<i>O₂</i>	<i>CH₄</i>	<i>C₄H₈</i>
	<i>N₂</i>	<i>CO₂</i>	<i>C₅+ H₂S</i>
<div style="display: flex; align-items: center;"> <div style="writing-mode: vertical-rl; transform: rotate(180deg); margin-right: 5px;">- Strength +</div> <div style="text-align: center;">  </div> <div style="margin-left: 10px;"> <p style="color: green;"><i>Alumina</i></p> <p style="color: purple;"><i>Carbon Prefilter</i></p> <p style="color: grey;"><i>Activated Carbon</i></p> <p style="color: purple;"><i>Molecular Sieve</i></p> </div> </div>		<i>C₂H₆</i>	<i>NH₃</i>
		<i>C₂H₄</i>	<i>H₂O</i>
		<i>C₃H₈</i>	

Fig. 1 – The relative strength of adsorption for gases onto the indicated adsorbents [20].

The electrolysis cell stack power supply converts incoming AC power to DC power at the proper voltage and current range to match the load characteristics of the electrolysis stack. The power supply subsystem provides monitoring of the stack voltage and current and variably controls stack hydrogen output via varying the current supply to the stack. The amount of amps applied to the stack directly corresponds to the flow rate of hydrogen (and oxygen) produced.

Water and Oxygen Management System (W&OMS) circulates water through the cell stack using a circulating pump. It also removes heat from the cell stack and maintains the stack temperature while regulating system pressure. Part of the pure water is oxidized into gaseous O₂ at the anodic side of the cell stack resulting in a gas/liquid mixture (O₂ + H₂O) at the outlet of stack. Liquid water phase is separated from the gaseous O₂ phase into the “O₂ separator tank”. The oxygen gas that is generated is cooled before venting outside of the electrolyser enclosure. Resultant condensed water from this gas stream is drained or returned to the water reservoir for reuse. Other functions of the W&OMS are to monitor water purity, minimum stack water flow, water quantity/level, stack exit water/oxygen temperature, pressure, and level of H₂ content in oxygen gas production. Generally, a deionization water conditioner is present in the closed water loop to remove ionic contaminants coming from construction materials.

Water and Hydrogen Management System (W&HMS) circulates water through cathodic side of the cell stack using a circulating pump. Due to the effect of the electric field, protons generated at the anodic side of the cell stack migrate across the polymer membrane to the cathodic side where they are reduced to produce gaseous hydrogen (H₂). Due to the water electro-osmotic flow, water from the anodic side of the cell stack pass through the membrane to the cathodic side resulting in a gas/liquid mixture (H₂ + H₂O) at the outlet of the cathodic side of stack. Typically, nominal operating temperature of PEM electrolysers is between 50 °C and 60 °C so gaseous hydrogen produced is water saturated. Liquid water phase is separated from the gaseous H₂ phase into the “H₂ separator tank”. In order to minimize the amount of water consumption in the electrolyser, a pipe (with a normally

closed valve) connects separator tanks in order to balance water levels in both separator tank. When water level reached the high limit threshold in the H₂ separator tank, valve opens and part of the water of H₂ separator tank is transferred to O₂ separator tank thanks to difference of pressure between O₂ side (up to 14 bar) and H₂ side (up to 35 bar). Then, valve closes when the water level reached the low limit threshold. As this water transfer is a critical step of the process, water reintroduction is typically done in two steps: i) Step 1 allows for the removal of dissolved hydrogen gas in the pressurized water, which effervesces out of solution at O₂ operating pressure (up to 14 bar) then first drained from the system and ii) Step 2, once the dissolved hydrogen has been allowed to release from solution, the remaining water can be introduced safely to the O₂ separator tank.

Typically, a deionization water conditioner is also present in the cathodic closed water loop to remove ionic contaminants coming from construction materials and the membrane (fluoride release). Hydrogen management system is responsible for creating and regulating system back pressure on the hydrogen side of the cell stack and monitoring system pressures, temperatures and water level [28].

Purification system

The purification system is modelled into two parts: drying system followed by TSA purification unit. The hydrogen gas that is generated is preliminary cooled before going to the TSA purification unit. Resultant condensed water from this gas stream is generally drained out of the enclosure of the electrolyser.

After preliminary cooling, the hydrogen produced is then purified using temperature swing adsorption unit. This purifier removes i) oxygen traces coming from gas cross over through the membrane using catalytic deoxidizer and ii) water content using two dryer column.

Dryer column is regenerated at low pressure under temperature (around 200–250 °C) when adsorbents are fully saturated of water. Heating the column and flushing with pure hydrogen allow to remove all the water catches by the dryer pellets. Regenerated dryer column is fully operational when

temperature is back to room temperature. Pure hydrogen used as regeneration gas may be provided by external H₂ tank or taken directly from the outlet hydrogen production of the electrolyser.

Since, O₂ and H₂O are the main impurities; the design of this TSA purification unit is performed to have specific level at the outlet (typically <5 ppm of O₂ and <5 ppm of H₂O) [24].

Chlor-alkali membrane electrolysis model system

Chlor-alkali model considered 3 mains parts:

- Raw material: H₂O (with water purification unit) and brine (saltwater);
- Membrane electrolyser;
- Purification system.

Raw materials

Tap water, used as raw material, is firstly purified by a reverse osmosis unit. This preliminary step is necessary to remove any contaminants and ions from the water before feeding the process with pure water (Conductivity < 0.1 μS/cm). The brine as raw material requires purification before entering a membrane cell. The brine must be of a very high purity for membrane electrolysis [29].

Membrane electrolyser

Saturated brine is passed into the first chamber of the cell where the chloride ions are oxidised at the anode, losing electrons to become chlorine gas. At the cathode, positive hydrogen ions pulled from water molecules are reduced by the electrons provided by the electrolytic current, to hydrogen gas, releasing hydroxide ions into the solution. The ion-permeable ion exchange membrane at the centre of the cell allows the sodium ions (Na⁺) to pass to the second chamber where they react with the hydroxide ions to produce caustic soda (NaOH) [30]. The overall reaction for the electrolysis of sodium chloride is thus: $2\text{NaCl} + 2\text{H}_2\text{O} \rightarrow \text{Cl}_2 + \text{H}_2 + 2\text{NaOH}$.

A membrane cell is used to prevent the reaction between the chlorine and hydroxide ions. Moreover it avoids contact between Cl₂ and H₂ gases which can react violently. For safety reason, sensor were considered present to monitor any transfer of chlorine through the membrane. The process takes place in the liquid phase. Hydrogen is recovered from the reaction cell through gas pipe. Water can contaminate the hydrogen by-product, for this reason a drying step is included in the process.

Nitrogen is often use in maintenance operation and for purging the process. It can contaminate the hydrogen stream.

Purification system

The hydrogen gas that is generated is preliminary cooled [25]. Resultant condensed water from this gas stream is generally drained out of the enclosure of the electrolyser.

After preliminary cooling, the hydrogen produced is then purified using temperature swing adsorption unit. This purifier removes i) oxygen traces coming from gas cross over through the membrane using catalytic deoxidizer and ii) water content using two dryer column. Since, O₂ and H₂O

are the main impurities; the design of this TSA purification unit is performed to have specific level at the outlet (typically < 5 μmol mol⁻¹ of O₂ and <5 μmol mol⁻¹ of H₂O).

Results and discussion

Steam methane reforming (SMR) process

Based on model considered, the probability of occurrence for SMR model system is presented in Table 4.

Nitrogen

The possible source of nitrogen is the raw material (natural gas) and PSA malfunction. The PSA was considered as the main barrier to nitrogen in the hydrogen product. An additional nitrogen on line monitor set to stop the system if the nitrogen exceed 100 μmol mol⁻¹ was a second barrier. Based on it, nitrogen probability of occurrence was classified as possible with probability class of 3.

Argon

The possible source of argon is the raw material. In the natural gas, argon is present at a level lower than the nitrogen. The PSA was considered as a barrier however depending on the raw material the PSA sizing may not be sufficient. Due to the raw material variation, argon probability of occurrence was classified as rare with probability class of 2.

Oxygen

Oxygen is not present in the raw material and it is not stable in the reforming conditions. Moreover, the PSA cannot be used with oxygen content for safety reason. Based on that, oxygen probability of occurrence is unlikely with probability class set to 0.

Helium

Helium is not present in the raw material (He < 10 μmol mol⁻¹ in Europe). Helium has no barrier in the process. Helium probability of occurrence is unlikely with probability class set to 0. This probability may be modified for other sources of natural gas.

Carbon dioxide

The source of carbon dioxide is the reforming reaction and the gas shift reaction which are producing CO₂. The PSA is considered as a barrier considering the adsorption of the different absorbents: molecular sieve, silicagel and activated carbon (see Table 1). In normal operations carbon dioxide is removed by the PSA. Carbon dioxide probability of occurrence was classified as unlikely with probability class of 0.

Methane

The source of methane is the raw material (natural gas or syngas) and PSA malfunction. The PSA was considered as the main barrier to methane in the hydrogen product. The design and operating parameters of PSA determined the output amount fraction of methane. If CO was used to design the PSA, therefore the amount fraction of methane will be below the limit of 100 μmol mol⁻¹.

Table 4 – Probability of occurrence of impurities (P) in SMR process with the different barrier existing in the process.

Contaminant	Threshold [$\mu\text{mol}/\text{mol}$]	Cause possible for the source studied	Existing barrier	P
Inert gas: N_2	100	Raw material PSA malfunction	PSA	3
Inert gas: Ar		Raw material		2
Oxygen	5	Not expected to be present.		0
Carbon dioxide	2	Raw material	PSA	0
Carbon monoxide	0.2	Raw material	PSA CO sensor on line	4
Methane (CH_4)	100	Raw material	PSA methane sensor on line	2
Water	5	Raw material	PSA	2
Total sulphur components	0.004	Raw material	Desulfuration unit Sulphur trap in reforming system (poisoning/process operation) PSA Stainless steel pipe and vessel	0
Ammonia	0.1	Raw material	PSA	0
Total hydrocarbons	2	Trace of hydrocarbons after reforming process	PSA	0
Formaldehyde	0.01	Raw material	PSA	1
Formic acid	0.2	Raw material	PSA	0
Helium	100	Not expected to be present.		0
Halogenated compounds	0.05	Raw material	Desulfuration unit Chlorinated trap in reforming system (poisoning/process operation) PSA Stainless steel pipe and vessel	0

Based on the PSA design for CO purification, methane probability of occurrence was classified as rare with probability class of 2.

Carbon monoxide

The source of carbon monoxide is the syngas and PSA malfunction. The PSA was considered as the main barrier to carbon monoxide in the hydrogen product. The design and operating parameters of PSA determined the output amount fraction of carbon monoxide and it was not expected to be over the ISO 14687-2 threshold in normal operation. It is considered possible to have CO peak at $\mu\text{mol mol}^{-1}$ level during the switch of PSA columns. As CO was used to design the PSA, therefore the amount fraction of CO should be below $0.2 \mu\text{mol mol}^{-1}$ level in normal operation. Additionally, on line monitor were considered to control the PSA operation, emergency stop was implemented if CO amount fraction is over process defined amount fraction (range of $\mu\text{mol mol}^{-1}$ level).

Carbon monoxide probability of occurrence was classified as frequent with probability class of 4.

Formic acid, ammonia and formaldehyde

The source of ammonia, formaldehyde and formic acid is the reforming reaction itself. Depending on the operating conditions, they could be present at various concentrations (rang of mmol mol^{-1} level). The PSA was considered as the main barrier considering the alumina adsorption for these three components (see Table 1). A PSA designed to deliver CO amount fraction below $10 \mu\text{mol mol}^{-1}$ was considered to deliver hydrogen on the output with less than $0.1 \mu\text{mol mol}^{-1}$ of

ammonia, $0.2 \mu\text{mol mol}^{-1}$ of formic acid and less than $0.1 \mu\text{mol mol}^{-1}$ of formaldehyde. Based on the low concentration in syngas and the PSA as an effective barrier, ammonia and formic acid probability of occurrence was classified as unlikely with probability class of 0. Considering the extremely low amount fraction specified in ISO 14687-2, the probability of occurrence of formaldehyde was considered very rare and set to 1 as a conservative approach based on the lack of analytical evidences.

Water

Water was considered present in the syngas (Syngas saturated in water). The PSA was considered as a barrier considering the adsorption of molecular sieve and alumina (see Table 1). A PSA designed to deliver CO amount fraction below $10 \mu\text{mol mol}^{-1}$ was considered to deliver hydrogen on the output with less than $5 \mu\text{mol mol}^{-1}$ of water. Water probability of occurrence was classified as unlikely with probability class of 0.

Sulphur compounds

Sulphur compounds were considered present in the raw material (Natural gas). Several barriers were considered in place: desulfuration unit upstream the reformer system with outlet amount fraction below 50nmol mol^{-1} . Secondly, the catalysts used for the reforming processes are irreversibly poisoned by sulphur compounds. Then, the prereformer, the reformer and the shift unit could be poisoned and were considered highly sensitive to sulphur compounds. In case of malfunction or breakthrough, the process conditions could not be achieved and the system should be stopped. In addition to that, the PSA was considered as the final barrier considering sulphur

compounds will adsorb before CO and CO₂ species. The last barrier considered was pipe and vessels itself as sulphur compounds has strong affinity with steel. Considering all these barriers, sulphur compounds probability of occurrence was classified as unlikely with probability class of 0.

Halogenated compounds

Halogenated compounds were considered possibly present in the raw material (Natural gas). Several barrier were considered in place for chlorinated compounds: hydrodesulphurization (HDS) unit upstream the reformer which should stop any chlorinated compounds present in the raw material. Secondly, the process of reforming is poisoned by chlorinated compounds. The pre-reformer, the reformer and the shift unit could be poisoned and were considered highly sensitive to chlorinated compounds. The catalyst is irreversibly poisoned by chlorinated. In case of malfunction or breakthrough, the process conditions could not be achieved and the system should stopped. The PSA was considered as the main barrier considering chlorinated compounds will adsorb before CO, CO₂ species. Considering all these barriers, halogenated compounds (especially chlorinated) probability of occurrence was classified as unlikely with probability class of 0.

Hydrocarbons

Hydrocarbons (number of carbon above 2) were considered present as trace amount fraction after the reforming reaction. The PSA was considered as a barrier considering the adsorption of molecular sieve and alumina. A PSA designed to deliver CO amount fraction below 10 μmol mol⁻¹ was considered to deliver hydrogen on the output with less than 2 μmol mol⁻¹ of total volatile hydrocarbons (excluding methane). Total volatile hydrocarbons probability of occurrence was classified as unlikely with probability class of 0.

Water PEM electrolysis

Based on the model considered, the probability of occurrence for electrolysis model system is presented in Table 5. Several parameters (argon, helium, carbon monoxide, formaldehyde, formic acid, hydrocarbons) are not expected to be present as they are completely absent from the production process.

Nitrogen

Two sources of nitrogen were investigated: air intake and pure nitrogen used as purging/inerting/actuating gas.

The sole source of air was an intake into pure water tank at anodic side during normal operation. However the operating conditions applied in anodic separator tank and the PEM membrane (low cross-over through the membrane) were considered as two existing barriers leading the probability of occurrence of air leak to zero.

Nitrogen is commonly used to actuate pneumatic valves, a leakage of nitrogen from the valve may be a source of nitrogen in the hydrogen gas. However it was considered very rare as it will be an issue for the process and detected on the pneumatic valves monitoring.

A leakage of hydrogen inerting valve was considered as another source of nitrogen contaminant, however the hydrogen operating pressure is superior to the nitrogen

pressure supply which will not allow a leakage of nitrogen into hydrogen. For this reason, the probability of this hydrogen source was considered very rare.

The highest probability of occurrence of nitrogen was considered from the nitrogen use for venting during emergency shut down and/or maintenance action. However the procedure requires that the gas production should be temporary vented after restart for a certain period of time (depending on the installation). For this reason, the probability of risk was considered rare as it will require an issue with the restart procedure.

From the different possible sources of nitrogen contamination, the nitrogen contamination was considered rare and the main source may be an issue in the venting procedure and restart.

Ammonia

The possible source of ammonia in hydrogen was the tap water used at the anodic side. However the reverse osmosis purification unit is considered a sufficient barrier to consider the probability of occurrence to zero. Additionally, the PEM membrane was considered as a second barrier because no transfer through the membrane was foreseen.

Halogenated compounds

The possible source of halogenated compounds in hydrogen was the tap water used at the anodic side. However the reverse osmosis purification unit is considered a sufficient barrier to consider the probability of occurrence to zero.

Sulphur compounds

The possible source of sulphur compounds was the potential release of nmol mol⁻¹ level of sulphur from material gaskets or valve seats. To avoid it, the selection of the material should consider if it contains sulphur. If sulphur containing material is excluded, the probability of sulphur compound presence is considered unlikely and set to zero.

Water

Water is a reactant in the electrolysis process. Three possible source of water in the hydrogen produced were considered: permeation through the PEM membrane due to electro-osmosis, hydrogen water saturated at 60 °C and TSA malfunction. Several barrier were considered in the model like the TSA dryer that should remove most of the water. If failure occurred, the system had dew point analyser with a trip at the TSA outlet set below 5 μmol mol⁻¹ in order to avoid any water coming out of the TSA. Finally, the procedure may ensure venting of gas production after restart (time of venting depend on the installation). Following the reason, water was considered as rare and set to class 2.

Carbon dioxide

Carbon dioxide sources might be the tap water at anodic side or the air into the pure water tank at the anodic side. The first barrier was CO₂ filter on the pure water tank air intake which should avoid CO₂ and air contamination. For the tap water, the reverse osmosis purification unit is considered an efficient barrier to avoid CO₂. Additionally, an anodic separator tank, ion exchange resin in closed water loop were additional

Table 5 – Probability of occurrence of impurities (P) in Water PEM electrolysis process with the different barrier existing in the process.

Contaminant	Threshold [$\mu\text{mol/mol}$]	Cause possible for the source studied	Existing barrier	P
Inert gas: N_2	100	Maintenance/start-up phase Leakage	Operating procedure (maintenance/restart) PEM membrane cross over	2
Inert gas: Ar		Air intake into water tank	H_2 operating procedure > N_2 pressure supply	0
Oxygen	5	Not expected to be present.		
		Generation at the anodic side of cell stack	TSA operating condition	2
		Membrane cross over	Oxygen sensor	
		TSA malfunction	Operating procedure (maintenance/restart)	
Carbon dioxide	2	Water at anodic side	CO_2 filter	1
		Air into the pure water tank	reverse osmosis purification unit for water Anodic separator tank Ion exchange resin in closed water loop PEM membrane	
Carbon monoxide	0.2	Not expected to be present.		0
Methane (CH_4)	100	Not expected to be present.		0
Water	5	Reactant	TSA dryer	2
		Through PEM membrane	Dew point monitor	
		Hydrogen water saturated	Operating procedure	
		TSA malfunction		
Total sulphur components	0.004	Not expected to be present.		0
Ammonia	0.1	Water at anodic side	Reverse osmosis purification unit PEM membrane	0
Total hydrocarbons	2	Not expected to be present.		0
Formaldehyde	0.01	Not expected to be present.		0
Formic acid	0.2	Not expected to be present.		0
Helium	100	Not expected to be present.		0
Halogenated compounds (organo-halogenated)	0.05	Water at anodic side	Reverse osmosis purification unit	0
Halogenated compounds (Cl_2)	0.05	From process/higher pressure of hydrogen rather than Cl_2	Safety monitoring at the anode/Separation of Cl_2 and H_2 by the process/detected due to faster diffusivity of hydrogen gas/process would be shut down in time to avoid contamination of hydrogen with $\text{Cl}_2(\text{g})$.	0
Halogenated compounds (HCl)	0.05	Conversion of $\text{Cl}_2(\text{g})$ would be likely to convert into $\text{HCl}(\text{g})$ at the catalyst surfaces.	Chemical reaction (negative Gibbs free energy)/ Cl_2 is unlikely to be present	0

barrier. Finally, carbon dioxide has a low cross over through the PEM membrane. The probability of carbon dioxide was considered very rare and set to class 1.

Oxygen

Oxygen source was generation at the anodic side of the cell stack, cross over through the PEM membrane and TSA malfunction. However the TSA system has temperature sensitivity if oxygen content is too high (overheat). The TSA has a temperature monitoring with trip at temperature above 50°C . Secondly an oxygen sensor was considered to monitor on line oxygen content with a trip at the TSA outlet set below $5 \mu\text{mol mol}^{-1}$ in order to avoid any oxygen coming out of the TSA. Finally, the procedure may ensure venting of gas production after restart (time of venting depend on the installation). Following the reason, oxygen was considered as rare and set to class 2.

Chlor-alkali membrane electrolysis

Based on model considered, the probability of occurrence for chlor-alkali model system is presented in Table 6. Several parameters (argon, helium, carbon monoxide, formaldehyde,

sulfur compounds, ammonia, hydrocarbons, formic acid) are not expected to be present as they are completely absent of the production process. The process use demineralized water which eliminates the chances to find significant amount of ammonia, formic acid or any water soluble impurities. Then the probability of occurrence was set to zero.

Halogenated compounds

For chlor-alkali process, the presence of chlorine and hydrochloric acid in hydrogen gas required particular focus. Several barriers influencing their presence in the gas phase and in the final hydrogen gas product are present: NaOCl and HOCl equilibrate in liquid phase. Their expected concentration at the cathode is 10 mg kg^{-1} . Theoretically, there is an equilibrium between $\text{HOCl}(\text{l})$ with $\text{Cl}_2(\text{g})$ at the cathode, but this is probably too low to be significant. Both $\text{HCl}(\text{g})$ and $\text{Cl}_2(\text{g})$ has extremely high water solubility and would effectively leave with water in the drying clean up step of the process. In the process, Cl_2 and H_2 are separated from each other and not expected to have Cl_2 in the hydrogen stream after the membrane separation. The transfer of $\text{H}_2(\text{g})$ and $\text{Cl}_2(\text{g})$ through the membrane could occur in the event of failure of liquid levels. A continuous gas phase through the membrane would be

Table 6 – Probability of occurrence of impurities (P) in Chlor-alkali process with the different barrier existing in the process.

Contaminant	Threshold [$\mu\text{mol/mol}$]	Cause possible for the source studied	Existing barrier	P
Inert gas: N_2	100	Maintenance/start-up phase		2
Inert gas: Ar		Not expected to be present.		0
Oxygen	5	can be present		4
Carbon dioxide	2	Oxidation of organic matter in the brine. Degradation of the membrane on the cathode side made of conducting by carboxylic acid end groups	Chemical reaction (CO_2 is expected to remains in the caustic soda lye that is produced)	1
Carbon monoxide	0.2	Not expected to be present.		0
Methane (CH_4)	100	Not expected to be present.		0
Water	5	can be present/Process parameter	Drying system to a dew point below -20°C (dew point: $\sim 40\text{--}60^\circ\text{C}$)	2
Total sulphur components	0.004	Not expected to be present.		0
Ammonia	0.1	Not expected to be present.	Use of pure water (demineralised water)	0
Total hydrocarbons	2	Not expected to be present.		0
Formaldehyde	0.01	Not expected to be present.		0
Formic acid	0.2	Not expected to be present.	Use of pure water (demineralised water)	0
Helium	100	Not expected to be present.		0
Halogenated compounds (organo-halogenated)	0.05	Not expected to be present.		0
Halogenated compounds (Cl_2)	0.05	From process/higher pressure of hydrogen rather than Cl_2	Safety monitoring at the anode/ Separation of Cl_2 and H_2 by the process/detected due to faster diffusivity of hydrogen gas/process would be shut down in time to avoid contamination of hydrogen with $\text{Cl}_2(\text{g})$.	0
Halogenated compounds (HCl)	0.05	Conversion of $\text{Cl}_2(\text{g})$ would be likely to convert into $\text{HCl}(\text{g})$ at the catalyst surfaces.	Chemical reaction (negative Gibbs free energy)/ Cl_2 is unlikely to be present	0

detected due to faster diffusivity of hydrogen gas. As hydrogen at the anode is monitored for safety reasons (H_2 and Cl_2 can give an explosive mixture), the process would be shut down in time to avoid contamination of hydrogen with $\text{Cl}_2(\text{g})$. Any possible presence of $\text{Cl}_2(\text{g})$ would be likely to convert to $\text{HCl}(\text{g})$ at the catalyst surfaces. Although the conversion has negative Gibbs free energy, an energy barrier is expected to slow down direct conversion. For the above mentioned reason and considering the barrier and online monitoring, the Cl_2 probability of occurrence was set to zero.

Others organo-halogenated compounds are not expected to be present as no organic compounds are expected to be present in the process.

Water

The water is present in the process and it can end up in the hydrogen gas stream. The process contain a dryer that ensure dew point of -20°C . It is clearly higher than the ISO 14687-2 specification. However depending on the process requirement, the hydrogen gas stream can be set for dew point between -40°C and -60°C . Water was then considered with a probability of occurrence of 2 considering the risk of variation in the drying process.

Nitrogen

Nitrogen is used for safety reason in process. It is mainly to avoid the presence of air in the system prior to hydrogen

generation and transportation, so it concerns only start-up and maintenance phase. During these two phases, a large amount of nitrogen is used so if there is problem in seal from purging line or in the purging process nitrogen can be present in hydrogen.

Carbon dioxide

Carbon dioxide is not likely to be in the product as is remains in the caustic soda lye that is produced. $\text{CO}_2(\text{g})$ could also be formed from oxidation of organic matter in the brine. The membrane on the cathode side is made conducting by carboxylic acid end groups. It has been speculated that $\text{CO}_2(\text{g})$ could be formed from degradation of the membrane. No quantitative assessment of the amount of $\text{CO}_2(\text{g})$ potentially formed from degradation of membrane or oxidation of organic material (and subsequent transport through membrane) could reach an amount fraction higher than the ISO 14687 specification. For the abovementioned reason the probability of risk for CO_2 was set to very rare (value of 1).

Oxygen

Oxygen can be present in the process [31]. As no analysis or monitoring has been previously done, it is unclear if oxygen will be absent or present at level higher than $5\ \mu\text{mol mol}^{-1}$. For this reason, the probability of occurrence was set at 0 or 4. As a conservative estimate, it will be set to 4 until report of analysis

Table 7 – Probability of contaminant presence in the three models presented in this article (SMR + PSA, PEM water electrolysis + TSA and chlor-alkali membrane electrolysis + TSA).

Probability of contaminant presence	Steam methane reforming with PSA	Chlor-alkali process (membrane cell process)	PEM water electrolysis process with TSA
Frequent	CO	O ₂	None identified
Possible	N ₂	None identified	None identified
Rare	CH ₄ , H ₂ O and Ar	N ₂ and H ₂ O	N ₂ , O ₂ and H ₂ O
Very rare	CH ₂ O	CO ₂	CO ₂
Unlikely	He, CO, O ₂ , CH ₂ O ₂ , NH ₃ , sulphur compounds, hydrocarbons compounds, halogenated compounds	He, Ar, CO, CH ₄ , CH ₂ O, CH ₂ O ₂ , NH ₃ , sulphur compounds, hydrocarbons compounds, halogenated compounds	He, Ar, CO, CH ₄ , CH ₂ O, CH ₂ O ₂ , NH ₃ , sulphur compounds, hydrocarbons compounds, halogenated compounds

demonstrates the presence or absence of oxygen in hydrogen from chlor-alkali process.

Considerations on other production methods

Alkaline electrolysis

The alkaline electrolysis was presented in the annex of ISO/DIS 19880-8 [12]. Alkaline electrolysis uses a diaphragm to separate the gases. Due to the better kinetics at high pH, Pt electrodes can be replaced by Ni. In this annex, nitrogen was classified with a probability of 3. The nitrogen origin is linked to insufficient purging after shutdown. The barrier considered is the electrolyser process control. If the operating procedure is defining clear procedure and purging time and the operating pressure of hydrogen is above the nitrogen pressure, the probability might be reduced to 2.

Oxygen and water were classified with a probability of 2, which is coherent with the water PEM electrolyser evaluation in Section [Steam methane reforming model system](#).

Argon was classified with a probability of 1 due to insufficient purging after shutdown. The argon is a contaminant in the nitrogen used for purging. Considering the reason mentioned for nitrogen above, the risk of argon might be reduced to 0 if operating procedure are well implemented.

The electrolyte is most commonly KOH or NaOH. The presence of water may be correlated with high ion content K⁺ or Na⁺ from the electrolyte. A high probability of water (like 2) may induce probability of presence of cations K⁺ and Na⁺. The international standard SAE J2719-2011 [32] specified a threshold for sodium (limit of 50 nmol mol⁻¹) and potassium or potassium hydroxide (limit of 50 nmol mol⁻¹). The probability of cations presence might be classified as 2 and may be reduced based on barrier and control in place in the production plant evaluated.

New production methods

When considering new production methods, the exercise presented in this article should be extended to impurities that may be present but not already regulated. This exercise will require to determine the severity of the new impurities founds in the hydrogen to realise the risk assessment (probability of presence x severity of effects on fuel cell). For example, recent

development around ammonia as energy carrier [33] will require to follow a similar risk assessment in order to evaluate the probability of presence of impurities from hydrogen produced using ammonia.

Summary

The contaminants potentially present in the hydrogen are depending on the process technology and on the purification step. The existing barrier may be different for every production installation therefore the probability of contaminant presence has to be investigated on a case by case basis for each site considering the barrier implemented locally and the working procedure as it may influence the results. Nevertheless, the general process can be used for the first approach. Based on the production model discussed in this article, the probability of contaminant presence in hydrogen for SMR and electrolysis model were summarized in [Table 7](#) considering the existing barriers in the model. The probability of contaminant presence will provide the first evidence to develop a hydrogen quality assurance plan based on the real process evaluated. Performing a similar study for each hydrogen production facility will help hydrogen providers or hydrogen integrators to focus and monitor the contaminants that could be present.

The next step for the project *Metrology for sustainable hydrogen energy applications* will be to confirm the assumption made with hydrogen quality measurement on several SMR and electrolyser. Then the risk assessment can be determined based on the probability of contaminant presence and on the severity of the contaminant against the fuel cell system.

Acknowledgements

The in-progress Joint Research Project «Metrology for sustainable hydrogen energy applications» is supported the European Metrology Programme for Innovation and Research (EMPIR). The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States.

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