ALLOWABLE HYDROGEN PERMEATION RATE FROM ROAD VEHICLE COMPRESSED GASEOUS STORAGE SYSTEMS IN GARAGES; PART 1 – INTRODUCTION, SCENARIOS, AND ESTIMATION OF AN ALLOWABLE PERMEATION RATE

P. Adams[1], A. Bengaouer[2], B. Cariteau[3], V. Molkov[4], A.G. Venetsanos[5]
[1] Volvo Technology Corp., Dept. 06120 Chalmers Teknikpark, 412 88 Gothenburg, Sweden
paul.adams@volvo.com
[2] Heat Transfer and Fluid Mechanics Laboratory, (DEN/DANS/DM2S/SFME/LTMF), CEA, F-91191 Gif-sur-Yvette Cedex, France
[3] Experimental Fluids mechanics Laboratory (DEN/DANS/DM2S/SFME/LEEF), CEA, F-91191 Gif-sur-Yvette Cedex, France
[4] HySAFER Centre, University of Ulster (UU), Newtownabbey, BT37 0QB, N. Ireland, UK
[5] Environmental Research Laboratory, National Centre for Scientific Research Demokritos, 15310 Aghia Paraskevi, Attikis, Greece

ABSTRACT

The paper presents an overview of the main results of the EC NoE HySafe activity to estimate an allowable hydrogen permeation rate for automotive legal requirements and standards. The work was undertaken as part of the HySafe internal project InsHyde.

A slow, long term hydrogen release such as that due to permeation from a vehicle into an inadequately ventilated enclosed structure is a potential risk associated with the use of hydrogen in automotive applications. Due to its small molecular size hydrogen permeates through the containment materials found in compressed gaseous hydrogen storage systems and is an issue that requires consideration for containers with non-metallic (polymer) liners. Permeation from compressed gaseous hydrogen storage systems is a current hydrogen safety topic relevant to regulatory and standardisation activities at both global and regional levels.

Various rates have been proposed in different draft legal requirements and standards, based on different scenarios and the assumption that hydrogen disperses homogeneously. This paper focuses on the development of a methodology by HySafe Partners (CEA, NCSRD, University of Ulster and Volvo Technology) to estimate an allowable upper limit for hydrogen permeation in automotive applications, by investigating the behaviour of hydrogen when released at small rates with a focus on European scenarios. The background to the activity is explained, reasonable scenarios are identified, a methodology proposed and a maximum hydrogen permeation rate from road vehicles into enclosed structures is estimated. The work is based on conclusions from the experimental and numerical investigations described by CEA, NCSRD and the University of Ulster in related papers.

1.0 INTRODUCTION

The primary goal of the HySafe permeation study has been to assist the safe introduction of hydrogen road vehicles with the minimum of restrictions for customers and manufacturers. In effect, to avoid the restrictions imposed by some countries on alternative fuel vehicles in parking facilities, e.g. the ban on LPG vehicles entering underground parking facilities in Belgium. The technical goals of the HySafe work can be summarised as:

- Identifying how hydrogen behaves when released into an enclosed volume at very low release rates, i.e. will a homogeneous mixture develop in the volume or will a stratified layer develop or a combination varying with time?

- Reassessment of the assumptions and simple calculations behind earlier proposals for an allowable permeation rate.
- Proposal of an allowable permeation rate considering, in particular, European scenarios.

2.0 HYDROGEN FUELLED ROAD VEHICLES

The majority of vehicle manufacturers consider using hydrogen in combination with fuel cells and electric drivetrains, as this combination provides the best potential gains in efficiency compared with current internal combustion engine technology.

For city buses, hydrogen is typically considered for use by various manufacturers with fuel cell based drivetrains and to a lesser extent internal combustion engines. For a typical full size city bus, e.g. a 12m long non-articulated single deck city bus, the maximum quantity of hydrogen required is in the order of 40-50kg, typically stored in a 35MPa compressed gaseous hydrogen (CGH_2) storage system. The maximum storage pressure could vary between 25MPa and 70MPa depending on the application, storage requirements and future industry norms. For city buses CGH_2storage systems are usually roof mounted, with the fuel cells either mounted on the roof or at the back of the vehicle. Liquid hydrogen storage systems could also be used.

For passenger cars, hydrogen is typically considered for use with fuel cell /electric drivetrains by many manufacturers and to a much lesser extent in internal combustion engine applications. For a typical passenger car the maximum quantity of hydrogen required is in the order of 3-10kg depending on the size and characteristics of the car. Typically CGH_2storage systems are used, however, some manufacturers have adopted liquid hydrogen storage systems. Until recently, CGH_2systems for prototype cars were typically based on a storage pressure of 35MPa, however, 70MPa systems are now available and are likely to become the norm due to the range and packaging demands of passenger cars. For passenger cars, hydrogen storage systems are usually mounted near the rear axle as this position offers the best protection in the event of a traffic accident. In prototype vehicles, the storage system has often been located in the luggage area.

3.0 TYPICAL COMPRESSED GASEOUS HYDROGEN CONTAINERS

For road vehicle applications, CGH_2systems typically have a maximum storage pressure of 35 or 70MPa and are designed to operate within normal ambient temperature ranges. In (draft) legal requirements and standards, CGH_2containers are normally categorised into the following four types:

- Type 1 - Metallic container,
- Type 2 - Hoop wrapped container with a metallic liner,
- Type 3 - Fully wrapped container with a metallic liner,
- Type 4 - Fully wrapped container with a non-metallic liner.

In Type 3 and 4 containers, the main purpose of the liner is containment of the hydrogen gas, while the overwrapping provides the structural strength of the container. Current Type 4 containers use a polymer as the liner, e.g. HDPE, typically overwrapped with carbon fibres set in a resin matrix. Other fibres such as glass or aramid may also be used, but most automotive hydrogen applications use carbon fibre. The overwrap varies in thickness around the container depending on the stresses, as does the direction of the fibres. Type 3 or Type 4 containers are used for almost all current automotive CGH_2applications.

4.0 HYDROGEN PERMEATION

Permeation in the context of CGH_2storage systems may be defined as “molecular diffusion through the walls or interstices of a container vessel, piping or interface material” [1]. Permeation may be categorised as a slow long term hydrogen release from a CGH_2 storage system. Due to its small molecular size, hydrogen permeates through the containment materials found in CGH_2 storage systems. Permeation increases with increasing storage pressure, material temperature and aging. For metallic containers or containers with metallic liners (commonly known as Types 1, 2 or 3) the permeation rate is considered to be negligible. However, hydrogen permeation is an issue for containers with non-metallic (polymer) liners (commonly known as Type 4) which readily allow the permeation of hydrogen.

Proposals for vehicle regulations and standards for hydrogen systems give limits on the allowable rate of hydrogen permeation from Type 4 containers during approval testing.

5.0 TYPICAL ENCLOSED STRUCTURES

Typical enclosed structures used by road vehicles include:

- Tunnels, or wide over bridges,
- Domestic single or multi-vehicle garages,
- Partially enclosed public parking, e.g. multi-storey parking with semi-open sides,
- Fully enclosed public parking, e.g. underground parking,
- City bus garages,
- Maintenance facilities,
- Showrooms,
- Covered bus stations, e.g. beneath shopping centres,
- Covered loading bays,
- Ferries,
- Train transport, e.g. Channel Tunnel.

With respect to hydrogen permeation, the critical cases were considered to be domestic garages for passenger cars and city bus maintenance facilities as being a representative case for commercial vehicles. One of the key challenges in estimating an allowable hydrogen permeation rate is the very wide variation in the design, construction and ventilation requirements for domestic garages.

6.0 METHODOLOGY FOR THE ESTIMATION OF AN ALLOWABLE HYDROGEN PERMEATION RATE

6.1 Introduction

The estimation of an allowable permeation rate for hydrogen containers used in road vehicle applications requires consideration of the issues identified below:

- Dispersion behaviour of hydrogen at the very low flow rates associated with permeation
- Vehicle scenarios
    - Quantity of hydrogen to be stored
    - Nominal working pressure
    - Size of hydrogen containers
    - Key vehicle dimensions
    - Maximum material temperature
- Environment scenarios
    - Enclosure dimensions
    - Reasonable minimum enclosure ventilation rate
    - Maximum prolonged ambient temperature
- Testing
    - New container or simulated end of life container
    - Testing temperature
- Level of safety required to take account of:
    - If the test is on a “new” container, the allowable rate must be such that the permeation from the container is safe at the end of its life
    - Different materials

- Test temperature relative to maximum prolonged material temperature
- Scenarios versus real world conditions

6.2 Hydrogen Dispersion

Understanding the dispersion of permeated hydrogen in confined spaces is crucial to the assessment of an allowable hydrogen permeation rate. No published experimental work undertaken prior to this study was identified to confirm the behaviour of hydrogen at the very low flow rates comparable with permeation. The closest published experimental work on low flow rate releases prior to this study showed some degree of stratification of the released gas, however, the releases were 3-4 orders of magnitude greater than the releases considered in this study, with the release from a concentrated source and helium used instead of hydrogen [2]. General opinion has been that the permeated hydrogen would disperse homogeneously.

The work undertaken by the HySafe Partners Commissariat à l'Énergie Atomique (CEA), National Centre for Scientific Research Demokritos and the University of Ulster has been focussed on the subject of hydrogen dispersion at very low flow rates, including "direct" numerical simulation of permeation. The work includes experiments, modelling and numerical studies. The details of the work are published in separate papers at the 3rd International Conference on Hydrogen Safety [3, 4 & 5]. The conclusion from the activities was that while some degree of stratification was observed in the experimental and modelling activities with 100% of the released hydrogen concentrated at a source [4, 5], it was very small in practical terms. The numerical study with "direct" simulation of permeation from the surface of a hydrogen container into a garage showed a negligible difference in hydrogen concentration at the ceiling and floor levels (below 0.01% by volume compared with the lower flammability limit of 4% by volume). For the purposes of estimating an allowable permeation rate, the studies concluded that it would be valid to assume homogeneous distribution of hydrogen at the permeation rates and ventilation rates considered.

6.3 Proposed Scenarios

With respect to hydrogen permeation, the critical cases were considered to be domestic garages for passenger cars and city bus maintenance facilities as being representative for commercial vehicles. Three passenger car and three city bus scenarios were considered and are summarised in Table 1.

The passenger car scenarios included a large car scenario with a maximum stored hydrogen quantity = 10kg [6]. The GM HydroGen4 SUV has a 70MPa system with a capacity of 4.2kg giving a range of 320km [7]. On this basis 10kg of hydrogen would give a range of 760km comparable to conventional vehicles. Other scenarios included a small/medium car scenario, e.g. Ford Fiesta, with a maximum stored hydrogen quantity = 6kg [6], and a minimum garage/micro car scenario based on the smallest prefabricated garages available from a number of UK manufacturers.

With respect to city bus maintenance or storage garages, in most situations it would be reasonable to assume that some type of forced ventilation would be required, however, the scenarios serve to cover concerns regarding failure of the forced ventilation and give an indication of the applicability of the allowable permeation rate. Typical full size, single deck, non-articulated city bus dimensions are length = 12.0m, width = 2.55m, height = 3.0m, with a reasonable maximum hydrogen storage quantity = 50kg [8]. The proposed scenario is based on the minimum maintenance volume for a single bus. Typical working space requirements for a city bus maintenance facility are approximately 2.0m to each side of the bus including the front and rear [9]. Above the bus, 2.0m is necessary to be able to lift the bus to work beneath it and a further 1.5m is necessary for lighting and other services, giving a total distance between the floor and roof of approximately 6.5m. An alternative scenario is a storage garage where the buses are parked close together. In such a facility the minimum dimensions for each bus are estimated as follows, bus length + 0.6m, bus width + 1.0m, bus height + 2.5m.

	Car Scenarios			City Bus Scenarios		
	1	2	3	4	5	6
	Large Car	Small Car	Min. Garage/ Micro Car	35MPa Bus Maint. Garage	70MPa Bus Maint. Garage	Min. Bus Garage
Enclosure Length (m)	6.5	5.0	3.7	16.00	16.00	12.60
Enclosure Width (m)	3.5	3.0	2.4	6.55	6.55	3.55
Enclosure Height (m)	2.2	2.2	2.1	6.50	6.50	5.50
Enclosure Volume (m^3)	50	33	19	681	681	246
Impermeable Material Vol.* (m^3)	4	2	1	5	5	5
Free Vol. in Enclosure** (m^3)	46	31	18	676	676	241
Storage pressure (MPa)	70	70	70	35	70	35
Hydrogen Stored (kg)	10	6	3	50	50	50
Storage Volume (L)	249	149	75	2082	1244	2082

Notes:

i) Hydrogen density at 35MPa/15^{0}C = 24.02kg/m^3 [10]

ii) Hydrogen density at 70MPa/15^{0}C = 40.18kg/m^3 [10]

iii) Scenario 3 is based on smallest garage that is easily available in the UK market

iv) * Volume of impermeable materials, tyres, etc. includes the volumes of those parts of the vehicle without the possibility of air movement (assuming that hydrogen can enter the passenger and other compartments). Assume 2m^3 for a car, 1m^3 for the micro-car, and 5m^3 for a city bus.

v) ** Free volume in facility = Facility Volume – Volume of impermeable materials, tyres, etc

Table 1: Summary Of HySafe Vehicle Scenarios

6.4 Maximum Prolonged Material Temperature

One of the key parameters affecting the actual permeation rate of hydrogen through the walls of a hydrogen container is the temperature of the container materials [11]. The temperature is influenced by thermodynamic processes inside the container during refilling for example, and by the ambient temperature. Temperature increases due to fires for example are not considered as they are not part of normal usage. All current draft legal requirements and standards specify a maximum material temperature of 85^{0}C for container materials, and although there may be variations in the precise definition the same figure is adopted. In terms of normal usage of the system the only time that a temperature of 85^{0}C would be experienced inside the container is immediately after fast refilling. Tests have repeatedly shown that the temperature falls rapidly after the refilling is complete so that the material temperature should drop below 50^{0}C in a few minutes [12].

The alternative way in which the material temperature increases during normal usage is due to a high ambient temperature. The hottest recorded air temperature anywhere in the world is 57.8^{0}C recorded in El Azizia in Libya in 1922, while in Europe it is 48.0^{0}C recorded in Athens in 1977 [13]. However, these are extreme peak temperatures that last for 1-2 hours at most [14]. With respect to long term average temperatures, which are of more relevance to the permeation phenomenon, the maximum figures are somewhat lower. The average monthly maximum and minimum temperatures over a 38 year period between 1913 and 1951 for El Azizia in Libya show that for the hottest month, July, the average maximum and minimum temperatures are 37 and 20^0C [15]. A statistical analysis of temperature loads for the EC StorHy project showed that in an example city (Athens) over a 30 year period average temperatures only exceeded 35°C for 5% of the year [16].

In summary, a prolonged material temperature of 85°C in an enclosed facility or beneath a vehicle is extreme and would only be experienced for short periods in comparison to the time necessary for permeation to produce hazardous concentrations of hydrogen. In addition the permeation barrier, i.e. the liner, would be insulated by thick carbon fibre wrapping. Based on the above and allowing for the effect of some additional warming inside an enclosed structure, a reasonable and conservative maximum prolonged material temperature could be taken as 55^0C, which harmonises with SAE [1].

6.5 Reasonable Minimum Enclosure Ventilation

One of the key issues in determining an acceptable permeation rate is the minimum natural ventilation rate of enclosed structures such as garages, since it affects the size of the hydrogen release that would cause the hydrogen concentration to reach a given level. A review of ventilation requirements undertaken by the HySafe internal project “InsHyde” indicated that in some European countries there are no minimum requirements for natural garage ventilation [17]. Although natural ventilation rates in many types of homes and other buildings are well studied, those for residential garages are not.

Six references were identified providing detailed sets of real world measurements of natural garage air ventilation including two studies by the Canada Mortgage & Housing Association [18 & 19], two studies by the Electric Power Research Institute (EPRI) in the USA [20 & 21], and a study by TIAX in the USA [22 & 23]. In addition an experimental study by HySafe Partner CEA was undertaken [4].In total 17 measurements in real world garages have been identified plus 8 measurements in an experimental facility. The lowest measurement identified in real world conditions was 0.03ac/hr from the TIAX study [22 & 23].

Experiments in a well sealed experimental garage at CEA (all vents closed and doors sealed), gave a natural helium leak rate of 6NL/min, which is equivalent to 0.01ac/hr assuming that the helium leak rate corresponds to the natural ventilation rate [4]. This value increased to 0.06ac/hr when the lower vent was opened and further to 0.07ac/hr when sealing was removed from the access door. With the upper vent open and both doors sealed the rate increased to 0.27ac/hr. With sealing removed from both doors the lowest value with both vents closed was 0.36ac/hr, increasing to 0.42ac/hr with the lower vent open, 1.72ac/hr with the top vent open and 3.29ac/hr with both vents open.

Prior to this study it had been assumed that domestic garages benefited from reasonable natural ventilation, however, the available measurements clearly demonstrate that many can be relatively poorly ventilated. The available real world measurements (excluding the data from the Canadian study which were considered to be unreasonably high for a reasonably weather proofed garage, ranging from 17-47ac/hr) are shown in Figure 1. The data shows that the natural ventilation rate for approximately 70% of garages can be expected to be less than 1ac/hr if the garages measured are representative of real world garages. This range is supported by the CEA measurements in an experimental garage and values calculated from the dimensions of real world garages in the TIAX study (see Figure 1).

Based on the previous sections it is proposed that a reasonable minimum garage natural ventilation rate is 0.03ac/hr. The value lies at a level reasonably above the assumed absolute minimum value of 0.01ac/hr (CEA measurement in a sealed experimental facility) and is equal to the lowest measured value for real world garages of 0.03ac/hr (TIAX, San Jose measurement). The rate is also harmonised with the SAE Fuel Cell Safety Work Group.

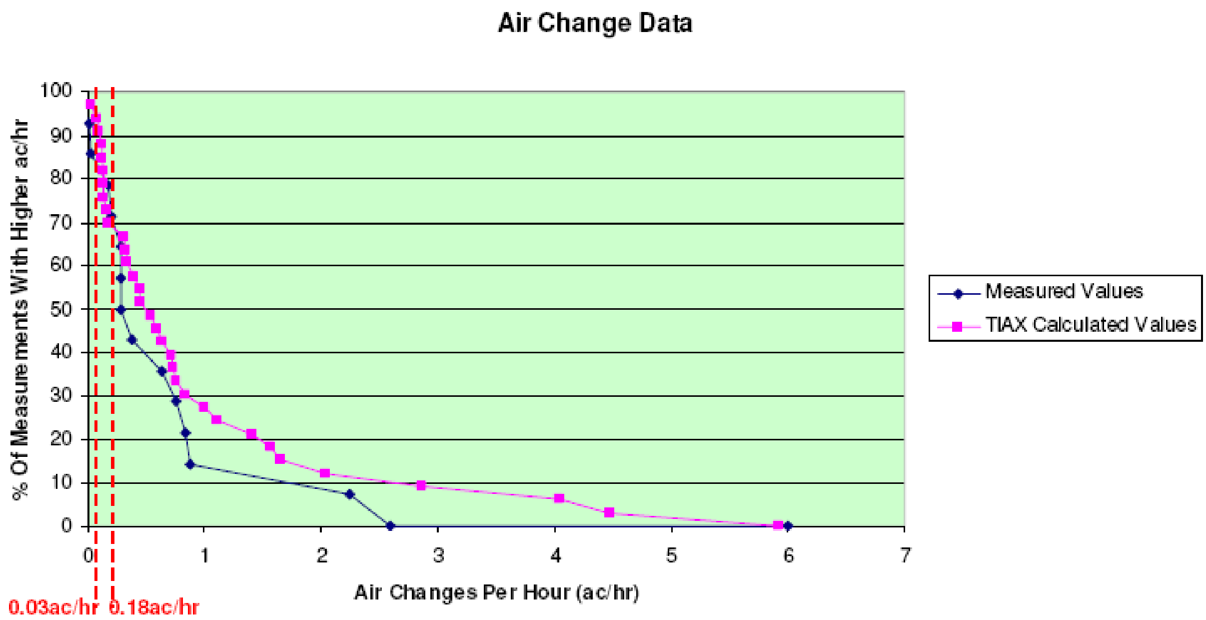


Figure 1: Available Real World Measurements (exc. Canadian studies) & TIAX Calculated Ventilation Rates

6.6 Testing

The specification of the testing conditions under which the actual permeation rate will be measured influences the allowable permeation rate. The allowable permeation rate could be set such that the conditions under which the test are performed represent the worst case for permeation from the container, i.e. the approach adopted in SAEJ2579 [1]. Alternatively, if the test conditions are not representative of the worst case conditions the allowable rate must reflect the difference between the test conditions and the worst case conditions. Factors to be considered include:

<u>New Container or Simulated End of Life Container (Aging)</u>

It has been reported that the carbon fibre overwrap on new containers could significantly restrict the permeation rate. In a container that is reaching the end of its service life the carbon fibre/resin matrix will be affected by a significant amount of micro-cracking that will not affect the structural integrity, however, it could allow an increase in hydrogen permeation. The increase in permeation for a container at end of life has been suggested to be twice that of a new container [11]. Further investigations revealed conflicting opinions based on testing experience. One test centre cast doubt on the validity of the underlying assumption based on their test results, and due to the nature of complete containers relative to material samples, e.g. varying overwrap thickness on a container, and at the weakest point, e.g. the interface with the metal end boss [24]. Other organisations suggest that the phenomena has been observed [25] and also that a factor of 2 may not be adequate [26]. The investigations suggest that the effects of aging are not adequately understood and further research should be carried out, as is being done by the French national project ENDEMAT [27] for example. As a result it is considered necessary to retain an arbitrary factor of two reduction between end of life and new containers which would provide allowance allow for:

- Unknown aging effects
- Use of new materials
- Statistical variation around limited existing data

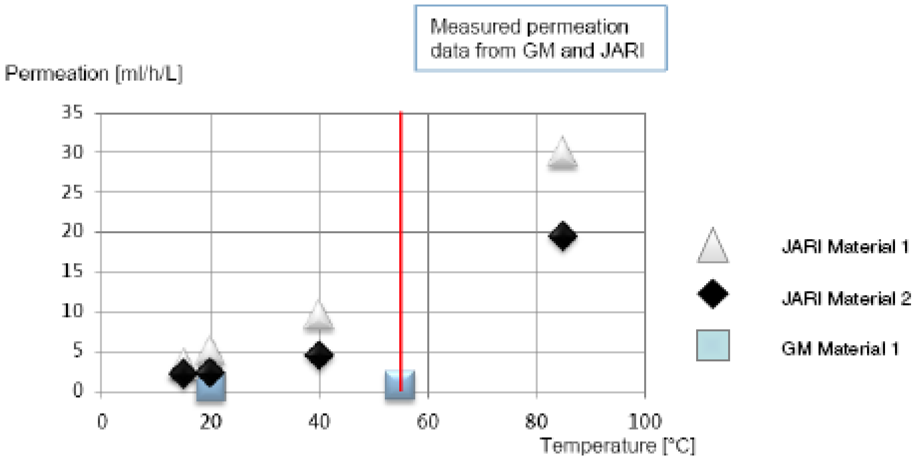
<u>Test Pressure</u>

Testing should be undertaken at the nominal working pressure.

Test Temperature

With respect to increasing material temperature, hydrogen permeation through polymer materials has been found to increase by an order of magnitude between approximately 24^0C and 82^0C [28]. However, the behaviour will vary from material to material. Material temperature/permeation characteristics are a key factor in determining the allowable permeation rate at a specified test temperature different to the maximum prolonged material temperature. Recently published material temperature/permeation rate data for three different materials has been used [29], see Figure 2.

If the testing is under taken at ambient temperature, the allowable permeation rate should be set such that the equivalent permeation at the maximum prolonged material temperature, previously proposed as 55^0C, results in a safe condition. The factor depends on the actual specified testing temperature and material behaviour. Based on Figure 2, the following factor was estimated; for a test conducted at a temperature of 20^0C, a factor of 3.5 will be used to convert from the maximum prolonged material temperature to the test temperature (4.7 at a temperature of 15^0C).



Source: Rothe [29]

Figure 2: Material Temperature/Permeation Characteristics

6.7 Level of Safety Required

The level of safety required has to take into account the probable and reasonably foreseeable variations around the values chosen for the other factors that influence the allowable permeation rate.

The long term maximum material temperature could be argued to have been set very conservatively at 55^0C. Equally the minimum natural ventilation rate may appear to have been set very low at 0.03ac/hr, however, the available measurements suggest that this is not the case.

The lower flammability limit (LFL) of hydrogen in air is recognised as 4% by volume. The flammability limits of hydrogen expand with temperature, e.g. the lower flammability limit for an upward propagating flame reduces from 4% at NTP to 3% at 100oC. The flammability limits also vary with the direction of flame propagation. The lower flammability limit increases to 7.2% for horizontally propagating flames and 8.5-9.5% for downward and spherically propagating flames [30]. In some forums this has been the reasoning for suggestions that 4% could be considered as a safe level, particularly for localised short term releases. However, permeation is a continuous long term release. Again it could be argued that most vehicles are used regularly so long term build ups are not relevant particularly for commercial vehicles, however, for passenger cars there are scenarios where this is not true. For example, someone may refill a passenger car and then take a long holiday leaving the car in a garage for say 4 weeks. An alternative and not unreasonable scenario is that someone refills the car, parks it in a garage and is then taken seriously ill and does not use the car for some months or maybe years.

The major sources of possible variations around the assumed scenarios lie in the on-board storage system size and particularly the vehicle and garage dimensions and the myriad combinations of them. The garage dimensions have been set at a reasonably comfortable level rather than particularly tightly as it is very subjective how small an accessible distance around the car should be. A further major source of possible variations is in the relationship of the permeation rate to material temperature for different materials. Based on the above it can be argued that there is need for an additional safety margin in addition to that already built into the various factors. For this reason it is proposed that the allowable concentration of hydrogen is taken as 25% LFL, i.e. 1% of hydrogen in air by volume in accordance with IEC and NFPA guidelines for explosive atmospheres. It is also harmonised with the SAE FC Safety Work group [1].

6.8 Calculation of an Allowable Permeation Rate

The following assumptions have been made:

- The allowable permeation rate will be specified in the same manner as the rate in the draft EC proposal [31], i.e. NmL/hr/L water capacity. *
- Releases similar in size to permeation can be considered to disperse homogeneously.
- Minimum natural ventilation rate for a domestic garage = 0.03ac/hr.
- Maximum permitted hydrogen concentration = 1% by volume, i.e. 25% LFL.
- Maximum long term material temperature = 55^0C.
- New container, with a factor of 2 to convert from the worst case end of life condition.
- For a test conducted at a temperature of 20^0C, a factor of 3.5 is used to convert from the maximum prolonged material temperature to the test temperature (4.7 at 15^0C). *

Note: * It should not be implied that these are considered to be the best test conditions. The aim of this work is to identify an allowable permeation rate rather than test conditions.

The basic method requires the calculation of the "safe" permeation rate at the end of life condition for the container combined with the maximum prolonged material temperature, and then the value is reduced to that for a new container at the nominal test temperature, e.g. 20^0C. Values were also derived for an alternative test temperatures of 15 ^{0}C.

The perfect mixing equation can be used to calculate the hydrogen release rate required to give a steady state hydrogen concentration [32]:

$$C_{\%} = \frac{100 \cdot Q_g}{Q_a + Q_g}$$

where:
$C_{\%}$ = Steady state gas concentration (% by volume),
Q_a = Air flow rate (m^3/min),
Q_g = Gas leakage rate (m^3/min).

Based on the above, the maximum allowable hydrogen permeation rate is given as follows:

$$Qp_x = \frac{Q_a \cdot C_{\%}}{100 - C_{\%}} \cdot \frac{60 \cdot 10^6}{V \cdot f_a \cdot f_t}$$

where:
Qp_x = Allowable permeation rate (mL/hr/L water capacity) at a test temperature of x^0C,
V = Water capacity of hydrogen storage (L),
f_a = Aging factor, taken to be 2,
f_t = Test temperature factor = 3.5 at a test temperature of 20^0C or 4.7 at 15^0C.

Based on the above assumptions, scenarios and methodology, the theoretical allowable permeation rates to give a hydrogen concentration less than 1% are given in Table 2.

Minimum Testing Temperature (^{0}C)	Maximum Allowable Permeation Rate (mL/hr/L water capacity)	
	Passenger Car	City Bus
15	**6.0**	3,7
20	**8.0**	5.0

Table 2: Theoretical Allowable Permeation Rates

It is proposed that the passenger car rates can be accepted for the city bus scenarios, as the worst case bus scenario is the “minimum” garage with failed forced ventilation and even in this situation the passenger car rates would still give a hydrogen concentration significantly lower than 4%. The proposed allowable permeation rates are given in Table 3 along with an alternative value based on similar methodology at the SAE test conditions.

New or Simulated End of Life Container	Minimum Testing Temperature (^{0}C)	Maximum Allowable Permeation Rate
New	15	**6.0*** mL/hr/L water capacity
	20	**8.0*** mL/hr/L water capacity
Simulated end of life	55+	**90** mL/min

Note: * The value to be adopted depends on the definition of ambient temperature, i.e. with a definition of 20^{0}C the allowable permeation rate should be 6.0NmL/hr/L, but if the test is specified at 15^{0}C the allowable permeation rate would be 4.6NmL/hr/L.

Table 3: Proposed Allowable Permeation Rate

The HySafe proposals for allowable hydrogen permeation rates are intended only for use in appropriate vehicle regulations and standards. The proposals are based on a range of garage scenarios that are considered to be representative of real world situations allowing the safe use of vehicles in typical enclosed structures such as domestic garages or maintenance facilities. The rates should not be applied to other situations or applications without further consideration. The proposed allowable hydrogen permeation rates are not applicable to hydrogen permeation into vehicle compartments. For hydrogen permeation into vehicle compartments the adoption of appropriate performance based requirements, or other requirements as appropriate, in the relevant vehicle regulations or standards are necessary to avoid the potential development of flammable hydrogen/air mixtures.

A comparison with allowable permeation rates from other (draft) legal requirements and standards is given in Table 4.

Source	Justification Reference	New or Simulated End of Life Container	Minimum Testing Temperature (^{0}C)	Maximum Allowable Permeation Rate (NmL/hr/L water capacity – except where indicated)
HySafe Proposal	-	New	15	**4.6**
			20	**6.0**
Alternative	-	Simulated end of life	55+	**90 mL/min per standard vehicle**
Early ISO15869 & draft ECE [33]	LLNL [11]	New	Ambient	**1.0**
EU Reg [31&34]	LLNL [11]	New	20±10	**1.0**
ISO/TS15869:2009 Option i) Test B16 [35]	JARI (2004) [36]	New	Ambient	**2.0@35MPa &** **2.8@70MPa**
ISO/TS15869:2009 Option ii) Test E5 [35]	-	Simulated end of life	20	**75 NmL/min per container**
JARI for UN ECE HFCV GTR	-	-	15	**5**
ACEA for EC Hydrogen Regulation	LLNL [11]	New	20±10	**10**
SAE J2579: 01 2009 [1]	-	Simulated end of life	Min. 55	**150 NmL/min per standard vehicle**

Table 4: Alternative Proposals For An Allowable Permeation rate

7.0 CONCLUSIONS AND RECOMMENDATIONS

The work undertaken by HySafe has:

- Addressed a number of issues in relation to hydrogen permeation in automotive applications,
- Provided a new methodology for justifying permeation rates based on published data,
- Been presented to the UN ECE WP29 HFCV-IG SGS and EC Hydrogen Working Group and subsequently to ISO TC197 WG6 and the SAE Fuel Cell Safety Working Group,

The allowable permeation rate for hydrogen has been estimated based on a number of key assumptions:

- A structure should be safe regardless of the vehicle that enters it, although what vehicle can physically enter the structure is a limit in itself,
- The allowable rate should be set so the vehicle is safe throughout its intended service life,
- The allowable rate should not rely on regulations affecting the structure to ensure safety, i.e. safety should be assured independent of the combination of vehicle and structure.

In determining allowable hydrogen release rates from a vehicle, it is necessary to consider the real world and the different regulatory regimes that govern vehicles and buildings. The automotive industry increasingly has regulations harmonised at a global or regional level, however, vehicle regulations do not regulate the design of structures associated with vehicle use. In contrast, buildings and infrastructure are regulated at a national or local level by different authorities to those developing vehicle regulations. To achieve the safe introduction of hydrogen vehicles without unnecessary restrictions on use, it is necessary to ensure that vehicle regulations are compatible with building and infrastructure regulations and vice versa. Any one vehicle may be driven into a wide range of garages during its service life, and the discharges should be safe for all reasonably foreseeable conditions. Conversely a garage or other enclosed structure can contain different vehicles during its life, however, the structure needs to be safe regardless of what vehicle is put inside. Vehicle regulations can only control the approval of a vehicle type and additional regulations control future roadworthiness inspections. Vehicle regulatory authorities are different agencies to those that deal with building regulations, so it is very difficult to link the two issues.

A key issue that has been identified during this study, is what the maximum allowable concentration of hydrogen should be. Clearly the lower flammability limit (LFL) should not be exceeded, though as it is difficult to achieve stable combustion below approximately 7% even this could be debated. However, the limit will probably be set by the building authorities or insurers rather than the vehicle manufacturers. 25% LFL is a common upper concentration limit and is recommended by international standards, e.g. IEC 60079-10. Further research could consider whether there is justification for raising this threshold for hydrogen.

Prior to this study it had been assumed that domestic garages benefited from reasonable natural ventilation, however, the available measurements clearly demonstrate that they can be relatively poorly ventilated. The available data suggests that the natural ventilation rate for many garages can reasonably be expected to be less than 1ac/hr. Prior to this study the reasonable minimum natural ventilation rate used in other garage studies was 0.18ac/hr. However, based on available measurements and research conducted for this report, this has now been agreed with the SAE as 0.03ac/hr. Similarly a reasonable maximum prolonged material temperature has been agreed with the SAE as 55^0C.

The dispersion behaviour of hydrogen at flow rates as low as those associated with hydrogen permeation was not described in previously published work. Experimental and numerical work by CEA, NCSRD and UU concluded that while some degree of stratification was observed in the experimental and modelling activities, it was so small in practical terms that it can be neglected. For the purposes of estimating an allowable permeation rate, the studies concluded that it would be valid to assume homogeneous distribution of hydrogen at the flow rates and ventilation rates considered.

The effects of aging on the permeation behaviour of complete containers appears uncertain with conflicting reports from different sources and further research may be necessary. As a result it is considered necessary to retain an arbitrary factor of two reduction between end of life and new containers to provide some allowance allow for unknown aging effects, use of new materials and the statistical variation around limited existing data.

The work has involved the development of a methodology, assumptions and scenarios on which the HySafe proposal has been based and subsequently optimised with the publication of new data. The work has compared the HySafe proposal with other proposals and has been the basis of presentations made to the UN ECE WP29 HFCV-IG SGS and the EC Hydrogen Working Group.

Key assumptions used in the HySafe estimation of an allowable permeation rate include:

- Allowance must be made for the wide variation of vehicles, buildings, ventilation characteristics, and the numerous resulting combinations of vehicles and buildings.
- Permeated hydrogen can be considered to disperse homogeneously
- Reasonable minimum natural ventilation rate for a domestic garage = 0.03ac/hr.
- Maximum permitted hydrogen concentration = 1% by volume, i.e. 25% LFL.

- Maximum prolonged material temperature = 55^0C.

Based on the above assumptions the allowable permeation rates indicated in Table 3 have been proposed to UN ECE WP29 and EC regulatory working groups in 2009. As very few scientific results have been published, the factors adopted for temperature and aging should be reviewed as and when further results become available. The HySafe proposals for allowable hydrogen permeation rates are intended only for use in appropriate vehicle regulations and standards. The proposals are based on a range of garage scenarios that are considered to be representative of real world situations. The rates should not be applied to other situations or applications without further consideration. The proposed allowable hydrogen permeation rates are not applicable to hydrogen permeation into vehicle compartments. For hydrogen permeation into vehicle compartments the adoption of appropriate performance based requirements, or other requirements as appropriate, in the relevant vehicle regulations or standards are necessary to avoid the development of flammable hydrogen/air mixtures.

Further details of the work can be found in HySafe report no. D74 [37].

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