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InsHyDe, Sub-task IP1.3

### ALLOWABLE HYDROGEN PERMEATION RATE FOR AUTOMOTIVE APPLICATIONS

Deliverable D74 (InsHyde) – FINAL with Corr.1

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The use of this report is entirely voluntary, and its applicability and suitability for any particular use is the sole responsibility of the user. The recommendations contained in this report provide a methodology for estimating an allowable permeation rate only for use in vehicle regulations or standards. Furthermore, the results and recommendations of this study are limited to the scenarios considered in this study together with the assumptions that have been made. Actual building and vehicle configurations and usage patterns may differ from those described in this report. Neither the HySafe Partners, nor any author of this report make any representation, nor gives any warranty, express or implied, as to the accuracy or completeness of this report or any of the information contained herein. The reader/user is fully liable for any conclusions drawn from its examination of the report and the information therein. No responsibility is assumed by the authors for any injury and/or damage to persons or property as a matter of product liability, negligence or otherwise, or from any use of the proposals contained herein.

#### Corrigendum 1

The "Free Volume In Enclosure" (see Table 3) for Car Scenario 1 "Large Car" should have been 48m<sup>3</sup> in accordance with Table 3 and the underlying assumptions. However, a "Free Volume In Enclosure" of 35m<sup>3</sup> was used for Car Scenario 1 "Large Car" in the calculation spreadsheet. The other scenarios considered were correct. The opportunity was taken to improve consistency between scenarios and the large car "Impermeable Material Volume" was increased from 2m<sup>3</sup> to 4m<sup>3</sup>, giving a final "Free Volume In Enclosure" of 46m<sup>3</sup>. As the large car scenario is the critical scenario for both cases in determining the allowable permeation rate, the proposed allowable permeation rate increases from 6.0 to 8.0NmL/hr/L water capacity at 20°C, and from 4.6 to 6.0NmL/hr/L water capacity at 15°C. Appropriate changes are made throughout the document. The changes do not affect the proposed value (90NmL/min per standard passenger vehicle) at the SAE test conditions.

For clarity the equivalent allowable permeation rate (90NmL/min per standard passenger vehicle) at the current SAE test conditions of 55°C or greater and at simulated end of life is added to Tables 12, 13, 19 and 20 and the Executive Summary. The value was previously only indirectly indicated in Table 16.

### **EXECUTIVE SUMMARY**

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 manufacturers and customers by avoiding the restrictions imposed by some countries on alternative fuel vehicles in parking facilities. The HySafe activity was initiated as the rates proposed in the draft ECE compressed gaseous hydrogen regulation and the various versions of ISO/DIS15869 (Gaseous Hydrogen And Hydrogen Blends - Land Vehicle Fuel Tanks) were believed to be overly restrictive. As a result HySafe undertook a scientifically based study to investigate if the existing rates could be relaxed safely. Discussions also took place with the SAE Fuel Cell Safety Working Group. The focus of this work is on providing an allowable permeation rate for the draft EC regulation for type-approval of hydrogen powered motor vehicles and the container requirements in the UN ECE WP29 GTR proposal.

Due to its small molecular size, hydrogen permeates through the containment materials found in compressed gaseous hydrogen storage systems. Permeation increases with increasing storage pressure, material temperature and the number of pressure cycles that the container is exposed to. For metallic containers or containers with metallic liners the permeation rate is considered to be negligible. However, hydrogen permeation is an issue for containers with non-metallic liners which are constructed from a non-load bearing polymer liner over wrapped with structural fibres set in a resin matrix (commonly known as Type 4 containers). Proposals for vehicle regulations and standards for hydrogen systems give limits on the allowable permeation rate from Type 4 containers during approval testing.

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.

The first part of this report provides an introduction to the subject area, while the second part explains the methodology, assumptions and scenarios on which the proposal is based, supporting the presentations made to the UN ECE WP29 HFCV-IG SGS and the EC Hydrogen Working Group in early 2009. The third part of the report compares the HySafe proposal with other proposals.

In estimating an allowable permeation rate the following assumptions have been made:

- Allowance must be made for the wide variation of vehicles, buildings, ventilation characteristics, and the numerous resulting combinations of vehicles and garages.
- The allowable permeation rate will be specified in the same manner as the rate in the draft EC regulation and ISO/TS15869, i.e. NmL/hr/L water capacity.
- Permeated hydrogen can be considered to disperse homogeneously following experimental and modelling work by the HySafe partners.
- 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°C.

Based on the above assumptions the following allowable permeation rates were <u>originally</u> proposed during presentations to the UN ECE WP29 HFCV-IG SGS and the EC Hydrogen Working Group in <u>January 2009</u>:

Minimum Lesting	Original
Temperature ( <sup>0</sup> C)	Maximum Allowable
	Permeation Rate
	(NmL/hr/L water
	capacity)
10	2.7*
15	3.1*
20	3.5*

Superseded by values in the following table.

Note: \* The value to be adopted depends on the definition of ambient temperature, i.e. with a definition of  $20\pm10^{\circ}$ C the allowable permeation rate should be 2.7NmL/hr/L, but if the test is specified at  $15^{\circ}$ C the allowable permeation rate would be 3.1NmL/hr/L.

Following those presentations, material temperature/permeation data provided by GM Powertrain Germany has allowed the original HySafe proposal to be optimised using the same methodology. As very few scientific results have been published, the factors for temperature and aging effects should be reviewed as and when further results become available. Based on the new data, revised allowable permeation rates were proposed and were presented to the EC Hydrogen Working Group on 10 March 2009 with the support of ACEA. The proposals were also presented to UN ECE WP29 HFCV-IG SGS on 29 May 2009, and provided to ISOTC197 WG6 and SAE Fuel Cell Safety Work Group. The values were subsequently updated by Corrigendum 1 to this report as shown in the table below and presented to the EC group on 16 June 2009.

New Or	Minimum	Allowable Permeation Rate
Simulated	Testing	
End Of Life	Temperature	
Container	$(O^{0})$	
	10	4.2* NmL/hr/L water capacity
New	15	6.0* NmL/hr/L water capacity
	20	8.0* NmL/hr/L water capacity
Simulated	55.	90 NmL/min per standard
End of Life	55+	passenger vehicle

Notes: \* The value to be adopted depends on the definition of ambient temperature.

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

The HySafe proposals for allowable hydrogen permeation rates are intended only for use in appropriate road 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 as appropriate, in the relevant vehicle regulations or standards are necessary to avoid the potential development of flammable hydrogen/air mixtures.

The HySafe permeation work is also covered in a series of four papers that have been submitted to the 3rd International Conference on Hydrogen Safety, 16-18 September 2009, Corsica, France:

 Adams, P., et al, "Allowable Hydrogen Permeation Rate From Road Vehicle Compressed Gaseous Storage Systems In Garages; Part 1 – Introduction, Scenarios, And Estimation Of An Allowable Permeation Rate".

- Venetsanos A.G., et al, "Estimation of an Allowable Hydrogen Permeation Rate From Road Vehicle Compressed Gaseous Hydrogen Storage Systems In Typical Garages; Part 2 – CFD dispersion calculations using the ADREA-HF code and experimental validation using helium tests at the GARAGE facility".
- Saffers J-B., et al, "Estimation of an Allowable Hydrogen Permeation Rate from Road Vehicle Compressed Gaseous Hydrogen Storage Systems in Typical Garages; Part 3: Modelling and Numerical Simulation of Permeation in a Garage with Adiabatic Walls and Still Air".
- Cariteau B., et al, "Experiments on the distribution of concentration due to buoyant gas low flow rate release in an enclosure".

Source	Justification Reference	New Or Simulated	Minimum Testing	Maximum Allowable Permeation Rate
		Container	remperature (°C)	– except where indicated)
HySafe Proposal	See Part II		10	4.2
	of this	New	15	6.0
	report		20	8.0
Alternative	See Pt III of	Sim. End of	Min.55	90 NmL/min per
	this report	life		standard passenger
				vehicle
Early ISO15869 & draft ECE	LLNL (2000)	New	Ambient	1.0
Draft EU Reg	LLNL (2000)	New	20±10	1.0
ISO/DIS15869.2&.3 & ISO/TS15869:2009 Option i) Test B16	JARI (2004)	New	Ambient	2.0@35MPa & 2.8@70MPa
ISO/TS15869:2009 Option ii) Test E5	-	Simulated end of life	20	75 NmL/min per container
JARI for GTR	-	?	15	5
Initial ACEA proposal for EU Regulation	LLNL (2000)	New	20±10	10
SAE J2579: Jan. 2009	-	Simulated end of life	Min. 55	150 NmL/min per standard <u>vehicle</u>

A comparison with allowable permeation rates from other sources is given below:

A critical issue has been identified that relates to the allowable permeation rate in ISO/TS15869: 2009 Test E5. It was understood that the rate was based on SAE J2579 (January 2008), however, with respect to the permeation test there is a fundamental and significant difference between the rate specified in the ISO and SAE standards. A comparison of the two standards shows two major differences between the SAE test and the ISO Option ii) test:

- i) The test temperature is reduced from 85°C (now 55°C) in the SAE document (depending on the revision) to 20°C in the ISO document.
- ii) The SAE rate is per vehicle, whereas the ISO rate is per container and it is likely that a typical passenger car would use more than one container, possibly two but say four as a reasonable upper limit.

Taken together the changes introduced into the ISO Option ii) rate imply a significant relaxation of the hydrogen permeation rate in comparison to the SAE standard on which it is understood that it was based. If the ISO Option ii) rate is considered in relation to the HySafe Scenarios it represents an increase in the allowable permeation rate in comparison to the

HySafe proposal by a factor of 1.1 to 3.0 if only one container is fitted per vehicle but an increase of 3 to12 times if four containers are fitted per vehicle. In addition there is a significant inconsistency between the rates given in Options i) and ii). On this basis it is proposed that the test specified in ISO/TS15869: 2009 "Gaseous hydrogen and hydrogen blends — Land vehicle fuel tanks" Annex E, E.5 is reviewed as a matter of priority.

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ACKNOWLEDGEMENTS

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- APPENDIX 1: Presentation To The EC Hydrogen Working Group On 27 January 2009
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#### 1. 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 both manufacturers and customers. In effect, to help avoid the restrictions imposed by some countries on alternative fuel vehicles in parking facilities.

The HySafe activity was initiated as the rates proposed in the draft ECE compressed gaseous hydrogen regulation and the various versions of ISO/DIS15869 (Gaseous Hydrogen And Hydrogen Blends - Land Vehicle Fuel Tanks) were believed to be unnecessarily restrictive. As a result HySafe undertook a scientifically based study to investigate if the existing rates could be relaxed safely. Discussions also took place with the SAE Fuel Cell Safety Working Group (Glenn Scheffler). The focus of the work was on identifying an allowable permeation rate for the proposed EC regulation for type-approval of hydrogen powered motor vehicles and the container requirements in the UN ECE WP29 GTR proposal.

Due to its small molecular size, hydrogen permeates through the containment materials found in compressed gaseous hydrogen storage systems. Permeation increases with increasing storage pressure, material temperature and the number of pressure cycles that the container is exposed to. For metallic containers or containers with metallic liners the permeation rate is considered to be negligible. However, hydrogen permeation is an issue for containers with non-metallic liners (commonly known as Type 4) which are constructed from a non-load bearing polymer liner over wrapped with structural fibres set in a resin matrix. Proposals for vehicle regulations and standards for hydrogen systems give limits on the allowable rate of permeation from Type 4 containers during type approval tests.

In the context of this work, the underlying principle of regulatory control must be clarified. 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 their use it is necessary to ensure that vehicle regulations are compatible with building and infrastructure regulations and vice versa. The draft EC regulation and the UN ECE WP29 GTR proposal will apply to prototype, pre-production and production vehicles.

Adequate allowance must be made in the estimation of an allowable permeation rate in recognition of the fact that it can only be an estimate given the wide variation of vehicle types, building designs, ventilation characteristics and requirements, and the numerous resulting combinations of vehicles and buildings.

The first part of this report provides an introduction to the subject area, while the second part explains the methodology, assumptions and scenarios on which the HySafe proposal is based, supporting the presentations made to the UN ECE WP29 HFCV-IG SGS and the EC Hydrogen Working Group in January and March 2009. The third part of the report compares the HySafe proposal with other proposals.

Note:  $1NmL = 1Ncm^3 = 1Ncc$ . Normal temperature is taken as  $20^{\circ}C$ .

Part I

## BACKGROUND

### 2. GOALS

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.
- Acceptability of the rate for heavy duty vehicles in maintenance facilities in addition to passenger cars in domestic garages.

#### 3. INTRODUCTION TO 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. In the medium term typical hydrogen powered road vehicles are likely to appear in applications which largely operate on start/stop cycles in urban environments and can exploit the potential efficiency gains of fuel cell/electric drivetrains. Such applications may include passenger cars, city buses and distribution trucks.

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 storage system. With a single deck city bus the storage space is not as critical as for other applications since there is significant usable roof space. The maximum storage pressure could vary between 25MPa and 70MPa depending on the application, storage requirements and future industry norms. For city buses compressed gaseous hydrogen storage systems are usually roof mounted, with the fuel cells either mounted on the roof or at the back of the vehicle, with both locations offering good ventilation that maximises the use of the strong buoyancy characteristics of hydrogen in the event of leaks. Roof mounted systems are also outside of the impact zones of traffic accidents. 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 compressed gaseous hydrogen storage systems are used, however, some manufacturers have adopted liquid hydrogen storage systems. Until recently, compressed gaseous hydrogen systems for prototype cars were typically based on a storage pressure of 35MPa, however, 70MPa systems are now available and may 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, with the fuel cells mounted in the "engine" compartment or underneath the car. In

many prototype vehicles, the hydrogen storage system has been located in the boot or luggage area.

Although there are a number of different technologies available for hydrogen storage using, for example, metal hydrides, nano materials or hybrid compressed cryogenic systems, there are currently only two mature storage options [1]:

- Compressed gaseous hydrogen storage,
- Liquid hydrogen storage.

#### 4. TYPICAL COMPRESSED GASEOUS HYDROGEN CONTAINERS

For road vehicle applications, compressed gaseous hydrogen systems typically have a maximum storage pressure of 35 or 70MPa and are designed to operate within normal ambient temperature ranges, see Figure 1.



Figure 1: Typical Compressed Gaseous Hydrogen Storage System

In (draft) legal requirements and standards, compressed gaseous hydrogen containers are normally categorised into the following four types, see Figure 2:

- 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.



Figure 2: Types Of Compressed Gaseous Hydrogen Containers

In Type 3 and Type 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 increasingly Type 4 containers are used for almost all current automotive compressed gaseous hydrogen applications.

#### 5. HYDROGEN PERMEATION

Permeation in the context of compressed gaseous hydrogen storage systems may be defined as "molecular diffusion through the walls or interstices of a container vessel, piping or interface material" [3]. Permeation may be categorised as a slow long term hydrogen release from a compressed gaseous hydrogen storage system. Other releases with similar flow rates may be small leaks from fittings or seals for example.

Due to its small molecular size, hydrogen permeates through the containment materials found in compressed gaseous hydrogen 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.

#### 6. TYPICAL ENCLOSED STRUCTURES

Typical enclosed structures used by road vehicles include (see Figure 3):

- 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.



Figure 3: Typical Enclosed Structures Used By Road Vehicles

One of the key challenges in assessing an allowable hydrogen permeation rate is the very wide variation in the design, construction and ventilation requirements for domestic garages, as illustrated in Figure 4.



Source: Various

Figure 4: Wide Variation Of Domestic Garage Designs

#### 7. LITERATURE SEARCH

A literature search was undertaken to identify any work dealing with the behaviour of hydrogen releases at the levels associated with permeation or small leaks (of the sizes quoted in draft legal requirements and standards). The following reports were identified:

- EIHP2 Sub-Task 5.3: Gaseous Hydrogen City Bus Safety Analysis Scenarios & Conclusions [4]. Discusses the CFD modelling work initiated in EIHP2 for permeation from city buses in maintenance facilities and provides possible scenarios.
- Vehicular Hydrogen Storage Using Lightweight Tanks [5 & 6]. Discusses the rationale behind the allowable permeation rate adopted in the draft UN ECE compressed gaseous hydrogen [7 & 8] regulation and in early drafts of ISO/DIS15869.
- Investigation Of Hydrogen Permeation And Hydrogen Safety In Garage [9]. Discusses the rationale behind the allowable permeation rate used in ISO/DIS15869.2 and .3 and ISO/TS15869: 2009 Option i).
- Hydrogen Related Risks Within A Private Garage: Concentration Measurements In A Realistic Full Scale Experimental Facility [10]. Discusses experimental releases designed to simulate releases from a vehicle into a garage. Stratification of the released gas was found, however, the releases are 3-4 orders of magnitude greater than the releases considered in this study, and helium was used instead of hydrogen.

In summary, no previously published research was identified that confirmed the dispersion behaviour of hydrogen at the release rates considered in this report. Additionally the rationales behind existing allowable rates were found to be focussed exclusively on passenger cars in domestic garages.

#### 8. ALLOWABLE RELEASE RATES IN VEHICLE REGULATIONS AND STANDARDS

#### 8.1 Introduction

Vehicle regulations are mandatory requirements that a manufacturer must fulfil in order to gain type approval for a vehicle, system or component, allowing it to be used on the public road. Within Europe, vehicle regulations can be:

- European Commission (EC) Directives (requiring enactment through national legislation)
- EC Regulations (enforceable directly without enacting national legislation),
- national legislation
- United Nations Economic Commission for Europe (UN ECE) Regulations or Global Technical Regulations if adopted by the country or region under the so called 1958 or 1998 Agreements respectively.

In contrast to legal requirements the adoption of a standard is voluntary, unless a particular standard is referenced in a vehicle regulation in which case its adoption becomes mandatory. Standards can be developed by any standard developing organisation. International standards affecting the use of hydrogen in automotive applications are developed by the International Organization for Standardisation (ISO), but supra-national or national standards are being developed in particular by organisations in North America such as SAE.

This section identifies the current allowable hydrogen permeation rates from compressed gaseous hydrogen storage systems quoted in current or draft legal requirements or standards. It also gives total hydrogen discharges from a vehicle. Many of the values quoted are subject to ongoing discussions in the appropriate body developing the vehicle regulation or standard, but are correct at the time of writing.

#### 8.2 Allowable Permeation Rates From The Hydrogen Storage System

There are a number of draft vehicle regulations and standards defining allowable permeation rates:

i) The draft European Regulation On Type Approval Of Hydrogen Powered Motor Vehicles\* [11], formerly the draft UN ECE regulation (in turn often referred to as the EIHP proposal) for the storage of compressed gaseous hydrogen on-board road vehicles [7 & 8] and early versions of draft ISO/DIS15869: Gaseous hydrogen and hydrogen blends – Land vehicle fuel tanks:

For Type 4 containers "*The steady state permeation rate shall be less than 1.0Ncm*<sup>3</sup> *per hour of hydrogen per litre internal volume of the container.*" The test is conducted at ambient temperature and nominal working pressure, i.e. the settled pressure at  $15^{\circ}$ C for a full container. The test is representative of a start of life container. In the current draft of the EU Regulation ambient temperature is defined as  $20^{\circ}$ C ±  $10^{\circ}$ C.

- ii) ISO/TS15869: 2009 Option i) Test B16 [12] & ISO/DIS 15869.2 & .3: Gaseous hydrogen and hydrogen blends Land vehicle fuel tanks [13 & 14]: For Type 4 containers "*The steady state permeation rate for hydrogen gas shall be less than 2,00 cm<sup>3</sup> of hydrogen per hour according to litre water capacity at 35 MPa, and 2,8 cm<sup>3</sup> per hour according to litre water capacity at 70 MPa." The test is conducted at ambient temperature and nominal working pressure, i.e. the settled pressure at 15<sup>o</sup>C for a full container. The test is representative of a start of life container.*
- iii) ISO/TS15869: 2009 Option ii) Test E5 [12]:
   "...the steady state hydrogen discharge rate due to leakage and permeation does not exceed 75 cm<sup>3</sup>/min (at 20 ℃ and 101,325 kPa) for use in standard passenger cars. For fuel tanks to be used in larger vehicles, such as buses, the allowable leakage may be increased in proportion to the enclosure volume for the vehicle."
- iv) SAE J2579, Jan. 2009 Technical Information Report For Fuel Systems In Fuel Cell And Other Hydrogen Vehicles [3]:

"The fully filled storage system shall be held at a temperature of at least 55  $\,^{\circ}$ C to stabilize and measure the total discharge rate due to leakage and permeation according to procedures given in Appendix C.7. This test may be conducted coincidentally with the last half of testing in 5.2.2.1.2 (at 85  $\,^{\circ}$ C) or after testing in 5.2.2.1.2 is completed with the system temperature held at least 55  $\,^{\circ}$ C for the measurement. The maximum allowable discharge from the compressed hydrogen storage system is 150 Ncc/min for standard passenger vehicles. The maximum allowable discharge for systems in larger vehicles is R\*150 Ncc/min where R = (Vwidth+1)\*(Vheight+0.5)\*(Vlength+1)/30.4 and Vwidth, Vheight, Vlength are the vehicle width, height, length (m), respectively."

The test is representative of an end of life container.

It is important to note that there are fundamental specification differences between the rates given above:

- In i) and ii) above, the allowable rates are linked to the size of the hydrogen storage system as determined by its water capacity. Whereas in iv) the rate is given for a vehicle regardless of the size of the hydrogen storage system, however, larger vehicles may be catered for by increasing the size of the test enclosure in relation to the size of the vehicle. In iii) the rate applies to an individual container and so the total rate per vehicle could be larger than the similar rate specified in iv).
- In i), ii) and iii) the permeation rate is at ambient temperature. In iv) the permeation rate is for the maximum prolonged material temperature
- In i) and ii) the test is on a new container, while for iii) and iv) the test is near the end of an expected service performance test sequence, i.e. the container should be in a condition similar to that of an end of service life container.
- The rates specified in i) and ii) are per hour, whereas in iii) and iv) they are per minute.

#### 9. SCENARIOS USED AS THE BASIS OF THE CURRENT ALLOWABLE RATES

#### 9.1 Earlier ISO & Draft UN ECE Rates

The permeation rate used in the draft UN ECE regulation [7 & 8] and earlier ISO drafts was based on a simple calculation by Lawrence Livermore National Laboratory (LLNL) [6]. The calculation was based on the time taken to fill a garage with a 4% hydrogen/air mixture, assuming a SUV with a large hydrogen storage system in a small garage and a minimum ventilation rate. The scenario dimensions used in the LLNL scenario are given in Table 1. The resulting permeation rate is linked to the water capacity of the hydrogen storage system, i.e. a larger system may permeate more hydrogen. It should be noted that the LLNL calculation recommended a rate of 10NmL/hr compared to 1NmL/hr that was adopted in the drafts.

#### 9.2 Current ISO Rates

The permeation rates stated in Option i) of ISO/TS15869: 2009 [12] and the earlier draft versions ISO/DIS15869.2 and .3 [13 & 14] are based on simple calculations by the Japan Automobile Research Institute (JARI), assuming a minimum natural ventilation rate and a single small hydrogen container [9]. The scenario dimensions used in the calculations are given in Table 1. The permeation rates are linked to the water capacity of the hydrogen storage system, i.e. a larger system may permeate more hydrogen. However, to take into account the fact that 70MPa systems will have a smaller water volume than 35MPa systems but will permeate more due to increased pressure, the rate for 70MPa systems is increased to 2.8NmL per hour per litre water capacity compared with 2.0NmL per hour per litre water capacity for 35MPa systems.

The justification behind the permeation rates stated in Option ii) of ISO/TS15869: 2009 [12] is not known, however, it corresponds to the numerical value used in the then current version of SAE J2579: Jan. 2008 [15] of 75NmL/hr at 85°C. However, it should be noted that the SAE figure was for a "standard passenger vehicle" whereas the ISO figure is per container.

#### 9.3 Summary Of Scenarios Used In Calculations To Justify Existing Rates

Table 1 summarises the facility and vehicle dimensions used (or estimated) in the scenarios justifying the existing permeation rates.

	1	2
Scenario	LLNL Garage (see Sect. 9.1)	JARI Garage (see Sect. 9.2)
Facility Length (m)	5.0*	6.00
Facility Width (m)	3.0*	2.43
Facility Height (m)	2.0*	2.40
Facility Volume (m <sup>3</sup> )	30	35
Storage pressure (MPa)	35	35 & 70
Hydrogen Stored (kg)	13	1.4** & 2.4**
Storage Volume (L)	540	60
Min. Natural Ventilation Rate (ac/hr)	0.18	0.18

Notes:

Estimated from volume

\* From storage pressure/volume

#### Table 1: Scenarios Used To Justify Current Permeation Rates

#### 9.4 Issues With The Existing Scenarios

A review of the scenarios and calculations that have been used to justify the existing allowable permeation rates highlights a number of issues that raise questions over the validity of the current allowable rates, and also from a European perspective:

- The justifications use a single scenario based on cars in domestic garages, and do not consider commercial vehicles in, for example, maintenance facilities,
- Are the minimum natural ventilation rates valid?
- Are the enclosure dimensions valid in a European context?
- All of the scenarios assume that a homogeneous mixture develops in the enclosure, although stratification occurs at the smallest release levels in previously published experimental work albeit 3-4 orders of magnitude greater than the release levels considered here. How does the hydrogen disperse at the levels being considered?
- The LLNL scenario represents a very severe upper bound scenario<sup>1</sup>. Conversely the JARI scenario represents a lower bound scenario<sup>2</sup>. What is a reasonable upper hydrogen quantity, and what are reasonable minimum enclosure dimensions and ventilation rates?

Notes:

1. The mass of hydrogen stored (13kg) is very large and the storage volume at 35MPa is unreasonable for a car and probably even the largest SUV from a packaging perspective. In comparison the GM HydroGen4 SUV has a 70MPa system with a capacity of 4.2kg giving a range of 320km [16]. On this basis 10kg would give a range of 760km comparable to conventional vehicles.

2. The mass of hydrogen stored is low and appears more appropriate for a prototype, micro-car or range extender system than a typical car with a marketable range.

Part II

### **HYSAFE PROPOSAL**

#### 10. ORIGINAL ESTIMATION OF THE HYSAFE ALLOWABLE PERMEATION RATE

This section describes the HySafe methodology and the original estimation which was published in support of presentations made to the UN ECE WP29 HFCV-IG SGS and the EC Hydrogen Working Group on 21<sup>st</sup> and 27<sup>th</sup> January 2009 respectively. Section 11 contains a revised estimation based on new testing data made available after the January presentations which was the basis of the presentation to the EC Hydrogen Working Group on 10<sup>th</sup> March 2009.

#### 10.1 Introduction

The estimation of an allowable permeation rate for hydrogen containers used in road vehicle applications requires consideration of the issues identified in Figure 5.



Figure 5: Key Issues Behind An Allowable Permeation Rate

The issues identified in Figure 5 are broken down into more detail 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
  - o Enclosure dimensions
  - o 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, i.e. aging
  - o Different materials
  - o Test temperature relative to maximum prolonged material temperature
  - Scenarios versus real world conditions
  - Statistical variation around limited existing data

The HySafe permeation work is also covered in a series of four papers that have been submitted to the 3rd International Conference on Hydrogen Safety, 16-18 September 2009, Corsica, France, see Section 17.

#### 10.2 Hydrogen Dispersion

#### 10.2.1 Introduction

Understanding the dispersion of permeated hydrogen in confined spaces is a crucial issue in 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 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 [10]. General opinion has been that the permeated hydrogen would disperse homogeneously.

The work undertaken by the HySafe Partners Commissariat à l'Énergie Atomique, 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. Papers detailing the work undertaken by those Partners will be published at 3<sup>rd</sup> International Conference on Hydrogen Safety, 16-18 September 2009, Corsica, France, see Section 17. Further details of the work are given below.

#### 10.2.2 Commissariat à l'Énergie Atomique (CEA)

The GARAGE facility developed at CEA has been used to perform a set of experiments on natural ventilation and low flow rate releases. This installation is an indoor enclosure of 40.92m<sup>3</sup>, 5.76m long, 2.96m wide and 2.42m high. As a typical domestic garage it is fitted with a tilting door for vehicle access, a back door for human access, and two vents that are 200mm in diameter. One vent is located near the floor and links the enclosure with the experimental hall in which the garage is situated. The second is near the ceiling and links the enclosure with the exterior of the experimental hall. Both vents are placed along the middle of the back wall (opposite the vehicle access door). Hydrogen releases are simulated with helium injected through various nozzles at a regulated mass flow rate. Concentration measurements are made using thermal conductivity probes named mini-katharometers. They are located at different heights along vertical masts distributed in the enclosure. The temperature is measured using thermocouples placed near the top and bottom of the enclosure.

Two sets of experiments were performed. First the leak rate of the enclosure was measured with different sealing conditions. Secondly, the distribution of helium was measured in the enclosure during very low flow rate injections in the most air tight configuration of doors and vents.

Sealing conditions of the enclosure can be varied by different combinations of opened or closed vents, obstruction of the tilting door and sealing of the back door joints with aluminium tape. The leak rate of the enclosure was measured with the tracer decay method. The decrease in concentration of a homogenous air/helium mixture initially at a volume fraction of 2% helium was monitored. From this measurement, the leak rate is deduced from an exponential decay fit of the data. In the experiments the enclosure can be considered to be in thermal equilibrium with the hall. Thus, except in case of the opened upper vent, gas exchanges between the enclosure and the exterior may be mainly driven by the density gradient due to the presence of helium in the enclosure rather than thermal gradient. With a helium volume fraction of 2%, the density gradient due to helium is equivalent to a temperature gradient of 5 °C between the interior and the exterior of the enclosure. Table 2 gives the air change measurements for the configurations tested. For configurations 3, 4 and 8, the previous assumption of thermal equilibrium between the hall and the enclosure is not valid as the upper vent is opened to the exterior of the building. The resulting measurements of the air changes in these configurations are strongly dependent on meteorological conditions.

No.	Lower Vent Position	Upper Vent Position	Vehicle Door	Access Door	Air Change (ac/hr)
1	Close	Close	Normal	Normal	0.36
2	Open	Close	Normal	Normal	0.42
3	Close	Open	Normal	Normal	1.72
4	Open	Open	Normal	Normal	3.29
5	Close	Close	Sealed	Sealed	0.01
6	Open	Close	Sealed	Sealed	0.06
7	Open	Close	Sealed	Normal	0.07
8	Close	Open	Sealed	Sealed	0.27

Table 2: CEA Test Garage Natural Ventilation Measurements

The vertical concentration distribution was measured during a low flow rate injection in the tightest configuration (No.5 in Table 2). Two flow rates were tested; 30NmL/min and 1000NmL/min. A vertical, 70mm diameter nozzle was used for the injection centred in the enclosure near the floor. At a flow rate of 30NmL/min the injection was performed over 64hrs. The average final concentration was 0.4%. Although a clear increase of concentration was observed this value is at the limit of the measurement capability. No stratification is found to occur but for such a low concentration the sensors may not be able to detect any stratification. The noise amplitude of the sensors is approximately 0.1%. The results obtained for the 1000NmL/min flow rate are more relevant. Injection was not reached. Figure 6 shows the time variations of the concentration at different levels. The straight black line on this graph is the theoretical increase of concentration for a homogeneous mixture and a perfectly sealed enclosure.

Stratification is found throughout the injection except between 60000s and 80000s. Inspection of this homogenization event shows that it does not occur on a diffusive time scale. During this event, temperature measurements show thermal homogeneity whereas before and after, there is a stable vertical temperature gradient of less than  $0.3 \,^{\circ}$ C. Hence, one can suppose that homogenization occurs due to an inversion of the temperature gradient. Indeed, the density variation associated with the helium concentration is of the same order of magnitude as that due to a temperature gradient of  $0.6 \,^{\circ}$ C. The vertical concentration difference ( $\Delta X$ ) varies with time, see Figure 7. After an initial increase it reaches a constant value of 0.22% between 10000s and 60000s. After the homogenous stage, the temperature gradient is again stable and the stratification appears again. At that stage and during the rest of the experiment,  $\Delta X$  constantly increases and the ratio of the concentration difference to the average concentration tends to be constant. This characteristic has also been found for higher flow rates except for the homogenous stage, so it can be inferred that there is no significant influence of the homogenization on the increase in  $\Delta X$ .



Note: Levels N1 to N5 are regularly vertically spaced between 0.2m and 2.2m

Figure 6: Helium Concentration Variation With Time During 1000NmL/min Release



Figure 7: Variation Of The Vertical Concentration Difference With Time During The 1000NmL/min Injection

#### 10.2.3 National Centre for Scientific Research Demokritos (NCSRD)

NCSRD investigated the time and space evolution of the distribution of hydrogen in confined settings due to permeation from typical compressed gaseous hydrogen storage systems used in city buses or cars, using the three dimensional CFD dispersion code ADREA-HF. The main goal was to examine whether hydrogen is distributed homogeneously within the given facility or whether stratified conditions develop under certain conditions. The nominal hydrogen flow rate considered was 1087NmL/min for a city bus based on the then current SAE permeation rate [15] for Type 4 hydrogen containers at simulated end of life conditions and maximum material temperature and a bus facility volume of 681m<sup>3</sup> (Rate = 75NmL/min x bus garage

vol./47). The release was assumed to be directed upwards from a 0.15m diameter hole located at the middle part of the cover over the roof mounted containers on a bus. Ventilation rates up to 0.03ac/hr were considered. Simulated time periods extended up to 20 days. The CFD simulations showed that fully homogeneous conditions exist for low ventilation rates, while stratified conditions prevail for higher ventilation rates. Regarding flow structure it was found that the vertical concentration profiles can be considered as the superposition of the concentration at the floor (driven by laminar diffusion) plus a concentration difference between floor and ceiling (driven by buoyancy forces). In all cases considered this concentration difference was found to be less than 0.5%. ADREA-HF was also validated against experiments performed by CEA at the GARAGE facility, using helium, with good agreement between predicted concentrations and experimental data.

### 10.2.4 University of Ulster (UU)

UU undertook a numerical investigation of a permeation scenario in a typical garage with quiescent air in order to understand:

- The dynamics of hydrogen concentration on the container's surface (would the hydrogen concentration on the surface remain constant or increase with time?),
- The interplay between the two mechanisms affecting hydrogen dispersion in a closed space, i.e. buoyancy and diffusion,
- The uniformity of hydrogen distribution in an enclosure such as a typical garage due to permeation (is the hydrogen-air mixture formed during permeation practically uniform or there is a layer of flammable hydrogen-air mixture under the ceiling?).

A conservative approach was applied for this study, i.e. "extreme" parameters that increased the permeation rate were chosen for numerical simulations, i.e. the container's material, thickness, temperature and pressure.

The hydrogen concentration on the surface grew as the square root of time when considering only the diffusion process without buoyancy, e.g. in microgravity. However, it was shown by both simple estimates and the numerical simulations that buoyancy prevails over diffusion in a matter of tens of seconds after the assumed start of permeation. One of the questions was what the hydrogen concentration increase on the surface would be before buoyancy effects raised the hydrogen-air away from the surface? Numerical simulations confirmed that at a time of about 80s from the assumed start of permeation, the buoyancy distortion of the "cylindrical" symmetry of hydrogen propagation by diffusion is obvious. Simulation showed that the maximum volumetric hydrogen concentration is on the top of the tank and its value is about 8.2×10<sup>-3</sup>% by volume (the lower flammability limit is 4% by volume). This value slowly grows when the "hydrogen-air layer" descends down to the tank. The numerical simulations clearly demonstrated that across the garage height, the hydrogen concentration is distributed nearly linearly, and the difference in hydrogen concentration at the ceiling and floor is only about 3x10<sup>-3</sup>% (i.e. three orders of magnitude below the lower flammability limit of 4% by volume). In practical terms during permeation in a garage a uniform mixture will be formed as the results suggest that if the hydrogen concentration at the top was 4.003% the concentration at the bottom of the garage would be 4.000%. The results allow the application of simple engineering formulas for natural ventilation in garages with hydrogen fuelled cars.

#### 10.2.5 Summary

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, 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.

#### 10.3 Proposed Scenarios

#### 10.3.1 Introduction

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. Three passenger car and three city bus scenarios were considered.

#### 10.3.2 Passenger Car Scenarios

- Large Car Scenario
  - Example dimensions of large European cars:

Audi Á8 -	5.1m x 2.2m
BMW 7 Series -	5.2m x 2.2m
Mercedes S-Class -	5.2m x 2.2m
Volvo S80 -	4.9m x 2.2m

- Typical large car dimensions based on the above: Length = 5.2m, width = 2.2m (1.9m exc. mirrors), height = 1.5m
- Reasonable maximum hydrogen quantity = 10kg [17] Note: GM HydroGen4 SUV has a 70MPa system with a capacity of 4.2kg giving a range of 320km [16]. On this basis 10kg of hydrogen would give a range of 760km comparable to conventional vehicles.
- Assume storage pressure = 70MPa, giving a water volume of 249L
- Reasonable minimum garage plan dimensions: 6.5m x 3.5m x 2.2m to eaves
- Small/Medium Car Scenario
  - Example dimensions of small European cars:

Citroen AX -	3.6m x 1.9m
Ford Fiesta -	3.8m x 1.9m
Peugeot 106 -	3.6m x 1.9m
Renault Twingo -	3.5m x 1.9m

- Typical small car dimensions based on the above:
- Length = 3.6m, width = 1.9m (1.6m exc. mirrors), height = 1.4m
- Reasonable maximum hydrogen quantity = 6kg [17]
- Assume storage pressure = 70MPa, giving a water volume of 149L
- Reasonable minimum garage plan dimensions: 5.0m x 3.0m x 2.2m to eaves
- Minimum Garage Scenario
  - Example dimensions of cars: Peugeot 107 - 3.4m x 1.7m Toyota iQ – 3.0m x 1.7m Smart - 2.7m x 1.8m
  - Typical micro car dimensions based on the above: Length = 2.7m, width = 1.8m (1.5m exc. mirrors), height = 1.5m

- Reasonable maximum hydrogen quantity = 3kg (estimated from the other car scenarios)
- Assume storage pressure = 70MPa, giving a water volume of 75L
- Reasonable minimum garage plan dimensions: 3.7m x 2.4m x 2.1m to eaves (based on the smallest prefabricated garages available from a number of manufacturers in the UK).

#### 10.3.3 City Bus Scenarios

In most situations it would be reasonable to assume that some type of forced ventilation would be required in city bus maintenance or storage garages, however, the scenarios serve to cover concerns regarding failure of the forced ventilation and gives an indication of the applicability of the allowable permeation rate.

- Typical city bus dimensions: Length = 12.00m, width = 2.55m, height = 3.0m
- Reasonable maximum hydrogen guantity = 50kg [18]
- Assume storage pressure = 35MPa, giving a water volume of 2082L, alternatively 70MPa gives a total water volume of 1244L
- Reasonable minimum garage plan dimensions: 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 [19]. 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. Based on these assumptions, reasonable minimum city bus maintenance facility dimensions are: 16.00m long, 6.55m wide, 6.50m high.
- Parking Garage Scenario (Minimum) An alternative scenario is a city bus storage garage where the vehicles 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 (also allows double deck buses). Based on these assumptions, minimum city bus garage dimensions are: 12.60m x 3.55m x 5.50m

#### 10.3.4 Summary

The various scenarios identified above are summarised in Table 3.

	Car Scenarios			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 <sup>3</sup> )	50	33	19	681	681	246
Impermeable Material Vol.* (m <sup>3</sup> )	4	2	1	5	5	5
Free Vol. in Enclosure** (m³)	46	31	18	676	676	241
Storage pressure (MPa)	70	70	70	35	70	35 (worst)
Hydrogen Stored (kg)	10	6	3	50	50	50
Storage Volume (L)	249	149	75	2082	1244	2082 (worst)

Notes:

i) Hydrogen density at  $35MPa/15^{\circ}C = 24.02kg/m^{3}$ [20]

ii) Hydrogen density at 70MPa/15<sup>o</sup>C = 40.18kg/m<sup>3</sup> [20]

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 4m<sup>3</sup>, 2m<sup>3</sup> and 1m<sup>3</sup> for large, small and micro cars respectively, and 5m<sup>3</sup> for a city bus.

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

vi) Scenario 3 is based on smallest garage that is easily available

#### Table 3: Summary Of HySafe Vehicle Scenarios

#### **10.4 Maximum Prolonged Material Temperature**

#### 10.4.1 Introduction

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 [5]. 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.

#### 10.4.2 Material Temperature

All current draft legal requirements and standards specify a maximum material temperature of 85°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°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°C in a few minutes [21].

#### 10.4.3 Ambient Temperature

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°C recorded in El Azizia in Libya in 1922, while in Europe it is 48.0°C recorded in Athens in 1977 [22] (some sources indicate 50.5°C for Seville, Spain in 1881). However, these are extreme peak temperatures that last for 1-2 hours at most [41].

With respect to long term average temperatures, which are of more relevance to the permeation phenomenon, the maximum figures are somewhat lower. Figures 8 and 9 show the average monthly maximum and minimum temperatures over a 38 year period between 1913 and 1951 for El Azizia in Libya [23]. At El Azizia it can be seen that for the hottest month, July, the average maximum and minimum temperatures are 37 and 20<sup>o</sup>C. 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 [24]. The following figures are extracted from web resources such as "Wikipedia" and "Infoplease":

- The highest average annual mean temperature anywhere in the world is 35°C recorded at Dallol, Ethiopia between October 1960 and December 1966.
- At Key West (USA) the 30-year normal temperature is 25.7°C.
- The longest recorded hot spell resulted in temperatures exceeding 38 °C for 162 consecutive days from 30 October 1923 to 7 April 1924 in Western Australia. Other sources suggest that the hottest average maximum temperature is 35.6 °C (Western Australia).

From the above, the hottest long term temperature is less than 40°C which is significantly smaller than the peak values and less than half of the maximum permitted material temperature.



#### Tripoli and Azizia Average Max Temperature

Source: [23]

Figure 8: Average Monthly Maximum Temperatures Over A 38 Year Period Between 1913 And 1951 At El Azizia



Tripoli and Azizia Average Min Temperature

Figure 9: Average Monthly Minimum Temperatures Over A 38 Year Period Between 1913 And 1951 At El Azizia

#### 10.4.4 Summary

A prolonged material temperature of 85°C in an enclosed facility or beneath a vehicle is extreme, and would only be experienced for relatively short periods in comparison to the lengths of 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°C, which harmonises with SAE [3].

#### **10.5 Reasonable Minimum Enclosure Ventilation**

#### 10.5.1 Introduction

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. Although natural ventilation rates in many types of homes and other buildings are well studied, those for residential garages are not.

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 [25]. As a result, additional research was undertaken. Initially the additional research aimed to identify any studies that had been published dealing with measured minimum natural ventilation rates in garages. Subsequently the work was extended to determine a reasonable minimum natural garage ventilation rate.

#### 10.5.2 Published Garage Natural Ventilation Rate Measurements

Six references were identified providing detailed sets of real world measurements of natural garage air ventilation including two Canadian studies, two USA studies, an experimental study by HySafe Partner CEA, and unreferenced data attributed to TNO

in the Netherlands. In total 17 measurements in real world garages have been identified plus 8 measurements in an experimental facility. The measurements include:

#### i) Canadian Studies (real world garages)

Two studies by the Canada Mortgage & Housing Association [26 & 27] gave measured garage air leakage rates of 17 ac/hr, 18 ac/hr, 37ac/hr, and 47ac/hr for sample houses in four different Canadian cities. However, by inspection these rates appear unreasonably high for a reasonably weatherproofed garage, especially 37 & 47ac/hr, i.e. one complete air change every 1.3 to 1.6 minutes.

#### ii) EPRI Study (real world garages)

A study for the Electric Power Research Institute (EPRI) in the USA [28] refers to an earlier study [29]. Residential garage air flows from the outside were measured at seven sites, with four measurements during different climatic conditions at one of the sites. The seven measurements at different sites with similar climatic conditions gave a mean of 1.19ac/hr, see Table 4. The minimum value measured was 0.38ac/hr, however, with different climatic conditions that dropped to 0.18ac/hr (which is the part of the justification of the minimum value quoted in both the LLNL and JARI (2004) calculations).

Site	Air Flow From Outside (ac/hr)
1	0.84
2	0.88
3	0.75
6	0.64
8 iv	0.38
9	2.60
10	2.24
Measured	under different climatic conditions
8 i	0.18
8 ii	0.29
8 iii	0.28

Table 4: Results OF EPRI Garage Ventilation Study [28 & 29]

iii) TIAX Study (real world garages)

A significant study was undertaken as part of a safety evaluation project for natural gas home vehicle refuellers by TIAX [30]. An additional reference provides more in-depth details of the study [31]. Although the TIAX study has been quoted as involving over 30 different garage measurements based on reference [30], it actually involved only 3 natural garage ventilation air measurements. The remaining values were calculated from dimensions/inspections of a further 30 residential garages spread across the USA. Furthermore, cross-checking the values for the three measurement sites indicated in the original presentation [30] with the data in the detailed report [31], showed that the values in the presentation are based on the calculations rather than actual measurements.

The three residential garages in which measurements were taken included two garages attached to houses and a town house garage in what is effectively a street canyon. More details of the measurements are given in Table 5. Although wind speed measurements are provided, no temperature measurements are given in the published data.

Location	Home & Garage Type	Vol. (m <sup>3</sup> )	Garage Characteristics	Comments	Actual Wind Speed (kph)	ASHRAE Average Wind Speed (kph)	Min. Measured Vent. Rate (ac/hr)	Calculated* Ventilation Rate (ac/hr)
Spring, Texas	1-story single family house with attached 2-car garage	86	2 panelled & overhead retracting garage "car" doors have significant air gaps	Home used for NREL building energy studies	2.3 av	20.4	0.28	2.86
Freemont, California	3-story town house with 2-car garage on bottom floor	115	1 panel overhead retracting garage "car" door with significant air gap at top	Substantial wind shielding due to adjacent 3- story houses	3.7 av	7.4	0.20	0.73
San Jose, California	One-story single family house with attached 2-car garage	107	1 panel overhead retracting garage "car" door well sealed by vinyl flaps	Vent opening sealed off for most of test	3.1	7.4	0.03	0.13

Note: \* - Calculated using the ASHRAE average temperature and wind speed for the site

Table 5: Summarised Results Of TIAX Garage Ventilation Measurements [30 & 31]

The results of the TIAX garage ventilation calculations are shown in Figure 10. The calculations are based on information provided by the garage owners regarding dimensions, ventilation, gaps around doors, etc. It is important to note that the calculations are based on the average prevailing wind speed and temperatures for the closest city listed in the ASHRAE weather database. The calculations are based on average weather conditions and do not represent the lowest measured ventilation rates by significant margins as can be seen in Table 5.

#### Air Change Data



Figure 10: Available Real World Measurements (exc. Canadian studies) & TIAX Calculated Ventilation Rates

#### iv) TNO Study

The main report on the TIAX work refers to unreferenced values of 0.5-2.0ac/hr in much earlier work by TNO [31], though it has not been possible to locate further details of this work.

#### v) CEA Measurements (Experimental Facility)

Experiments in a well sealed experimental garage at CEA, gave a natural helium leak rate of 6NL/min, which is equivalent to 0.01ac/hr [32]. Additional measurements in the experimental garage are detailed in Section 10.2.2 and Table 2. The natural ventilation rate 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.

#### 10.5.3 Problems With The Available Data

The available garage natural ventilation data is summarised in Figure 10.

A number of problems exist with the available measurements of natural garage ventilation rates:

- Only a limited number of measurements (25 in total) are available of which 17 are taken from real world garages and 8 from an experimental facility.
- Of the 17 available real world measurements, 3 are less than or equal to the minimum value used in earlier justifications of the allowable permeation rates (0.18 ac/hr), and a further 4 measurements are relatively close, i.e. less than 0.4 ac/hr. <u>It is clear that a reasonable minimum natural ventilation rate for garages is less than 0.18ac/hr.</u>
- Of the 8 measurements in an experimental facility, 4 were less than 0.18ac/hr and a further 2 were less than or approximately equal to 0.4ac/hr.
- Large differences appear between the rates measured in the Canadian and the EPRI/TIAX studies for the limited number of measured values available. The Canadian measurements (17 47ac/hr) appear high for a reasonably weather

proofed garage given that the highest rates in the EPRI and TIAX studies were 2.6 and 0.28ac/hr respectively.

- It is clear from the EPRI data that climatic conditions can have a significant effect on the ventilation rates, however, it is the only study that provides full climatic data for the time of the measurements and a comparison of different conditions at the same location. The published TIAX data includes wind speed measurements but not temperatures.
- Both the LLNL and JARI justifications (see Section 9) refer to a statistical garage ventilation study, however, the study did not involve any actual measurements and was purely a statistical estimate based on an ASHRAE (American Society of Heating, Refrigerating and Air Conditioning Engineers) recommended design value. The statistical study was based on a Poisson distribution around a design value of 3.73ac/hr recommended in ASHRAE Standard 62-1989 "Ventilation For Acceptable Indoor Air Quality" which gives 0.18 ac/hr for one in 1 billion garages. Coincidentally this is the same value as the minimum measurement in the EPRI study, which was used as further justification for accepting the value as the assumed minimum natural garage ventilation rate. Additionally there are certainly significantly less than 1 billion domestic garages existing in the world, and taking the statistical result and comparing it with an identical value from 1 out of 18 actual measurements is not a valid method of identifying a reasonable minimum value. The statistical estimate should be discounted, since it is not based on measurements.

Due to the problems identified above with the available data and the assumptions made in earlier studies it was deemed necessary to make a new estimate of the reasonable minimum natural garage ventilation rate to allow for statistical variation around the measured values.

#### 10.5.4 Identification Of A Reasonable Minimum Natural Ventilation Rate

#### i) Base Value

The data identified in the previous section shows that 18% of the known measurements in real world garages are less than or equal to the minimum natural ventilation rate of 0.18 ac/hr adopted in the earlier LLNL and JARI studies (see Section 9). Additionally 41% of the measurements in real world garages are less than 0.4ac/hr. The measurements in the real world garages are of a similar order to the measurements in the CEA experimental facility, see Section 10.2.2. The 0.18 ac/hr value had previously been assumed to be a lower bound figure, however, it is clear from Figure 10 that it is not a reasonable minimum value.

The minimum CEA measurement [32] confirms that it is not realistic to assume that a domestic garage will be completely airtight regardless of weatherproofing, since a helium leak rate of 0.01ac/hr was recorded despite measures to completely seal the experimental facility, e.g. aluminium tape over joints. The CEA value can be taken to represent the absolute minimum natural ventilation rate.

The 4 Canadian measurements were neglected as being too high for a reasonably weather proofed garage.

From the above it can be concluded that a reasonable minimum natural ventilation rate lies between 0.01 and 0.18 ac/hr.

If the remaining 13 measurements from EPRI and TIAX (shown in Figure 10) are considered, 0.12ac/hr represents a reasonable minimum value, though this is a relatively arbitrary figure.

#### *ii)* Effect Of Climatic Conditions

The EPRI measurements for Site 8 [28] indicate that the climatic conditions can have a significant effect on the natural ventilation rate, see Table 6.

Site 8	i	ii	iii	iv	
Inside temperature °C	21.0	19.9	23.8	25.8	
Outside temperature °C	31.5	25.4	16.7	12.0	
Difference °C	-10.5	-5.5	7.1	13.8	
Wind* m/s (kph)	2.19 (7.9)	1.57 (5.7)	3.08 (11.1)	1.60	(5.8)
Ac/hr	0.18	0.29	0.28	0.38	
Note: * Wind direction not known.					Source: [28]

Table 6: Climatic Variations At Site 8 Of The EPRI Study

It is not clear from the EPRI Site 8 data what the actual mechanism is that affects the natural ventilation rate, e.g. internal/external temperature difference, the temperature gradient or an effect of the wind speed. The Site 8 data indicates that given probable climatic conditions it would be reasonable to expect the minimum natural ventilation value to be lower than 0.18ac/hr, say 0.10ac/hr as an extreme, i.e. approximately 25% of the Site 8 value that is comparable with the climatic conditions of the other EPRI measurements (0.38ac/hr).

Inspection of the detailed TIAX measurements and calculation data indicates that the wind speed has a significant effect on the natural ventilation rate, see Table 7. Table 7 suggests that the ventilation rate drops as the wind speed decreases, however, the related temperatures are not known. The measurements were taken at wind speeds that were 10-50% of the ASHRAE average for the areas, resulting in measured natural ventilation rates of 10-25% of the values calculated for the average wind speed.

	ACTUAL		CALCULAT	ED		
Location	Measured Wind Speed (kph)	Minimum Measured Ventilation Rate (ac/hr)	Average Wind Speed from ASHRAE Data (kph)	Calc. Vent. Rate for Average Wind Speed (ac/hr)	Ratio Of Measured To Calculated Vent. Rate	Ratio Of Measured To Average Wind Speed
Spring, Texas	2.3 av	0.28	20.4	2.86	0.10	0.11
Freemont, California	3.7 av	0.20	7.4	0.73	0.27	0.50
San Jose, California	3.1 av	0.03	7.4	0.13	0.23	0.42
						Source: [31]

 Table 7:
 Wind Speed & TIAX Ventilation Measurements

The effects of the climatic variations between actual minimum measurements and measurements or calculations for typical or average conditions are summarised in Table 8. It can be seen that the measured ventilation rates average 27% of the values based on typical or average climatic conditions.

Measurement	Actual Measured	Ventilation Rate For	Ratio Of Actual To
Details &	Minimum	Typical/ Average	Typical Ventilation
Reference	Ventilation Rate	Climatic Conditions	Rate
	(ac/hr)	(ac/hr)	
EPRI, Site 8	0.18	0.38	0.47
[28]			
TIAX, Spring	0.28	2.86	0.10
[31]			
TIAX, Freemont	0.20	0.73	0.27
[31]			
TIAX, San Jose	0.03	0.13	0.23
[31]			
		Average Ratio	0.27
		-	

#### Table 8: Comparison Of Actual & Typical Ventilation Rates

In estimating a reasonable minimum natural ventilation rate, it is clear from the above that climatic factors have to be considered and it is not reasonable to take average climatic values when hoping to identify a reasonable minimum rate. Based on the available data a conservative value of 10% of the average or typical measured values could be arbitrarily assumed. However, it could also be argued that an average rate is more appropriate, since permeation is a long term affect and the ventilation rate will fluctuate above the minimum value over a period of time. On this basis it is proposed to take 25% of the average or typical measured values

#### iii) Summary

Prior to this study it had been assumed that domestic garages benefited from reasonable natural ventilation, however, the available measurements clearly demonstrate that many are relatively poorly ventilated.

The available real world measurements (excluding the Canadian study) and the TIAX calculated values are shown in Figure 10. The two data sets clearly show that the natural ventilation rate for approximately 70% of garages can be expected to be less than 1ac/hr. This range is supported by the CEA measurements in an experimental facility, see Section 10.2.2.

Based on the previous sections it is proposed that a reasonable minimum garage natural ventilation rate is 25% of 0.12ac/hr, i.e. the proposed reasonable minimum 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 that adopted by the SAE Fuel Cell Safety Work Group.

Reasonable minimum natural garage ventilation rate = 0.03ac/hr.

#### 10.6 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 [3]. 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:

• New container or simulated end of life container

As indicated previously it has been reported that 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 [6].

If the testing is under taken on a "new" container, the allowable permeation rate should be set such that the equivalent permeation rate at end of life results in a safe condition, i.e. the allowable end of life permeation rate should be reduced by a factor of 2 for a new container. This factor was investigated further after January 2009 and is reviewed in Section 11.2.1.

• Test pressure

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°C and 82°C [5]. No further published data was identified at the time the original work was carried out. New data made available after January 2009 is considered in Section 11.2.2. Additionally, such a curve will vary from material to material. As a result it has been conservatively assumed that the increase in permeation rate with temperature is linear. Figure 11 illustrates the effect of different testing temperatures. If the testing is under taken at ambient temperature, say 10-20°C, the allowable permeation rate should be set such that the equivalent permeation at the maximum prolonged material temperature, previously proposed as 55°C, results in a safe condition. The factor depends on the actual specified testing temperature and material behaviour.



Permeation

Figure 11: Effect Of Different Testing Temperatures On Allowable Permeation Rate

#### 10.7 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°C. 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 100°C. 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 [33]. 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, however, the assumptions used in this respect have been conservative. 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 and to take into account statistical variation. 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 [3].

#### 10.8 Calculation Of An Allowable Permeation Rate

#### 10.8.1 Assumptions

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, i.e. NmL/hr/L water capacity (also see Section 10.8.5).
- 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 =  $55^{\circ}$ C.
- New container.
- Section 11 contains a revised estimation of the following factors based on new testing data made available after the January presentations which was the basis of the presentation to the EC Hydrogen Working Group on 10<sup>th</sup> March 2009.
  - For a test conducted at a temperature of 20°C, a factor of 12 is used to convert from the calculated permeation rate at end of life (factor of 2) and maximum prolonged material temperature (factor of 6) to that of a new container at 20°C.
  - For a test conducted at a temperature of  $15^{\circ}$ C, a factor of 14 is used to convert from the calculated permeation rate at end of life (factor of 2) and maximum prolonged material temperature (factor of 7) to that of a new container at  $15^{\circ}$ C.
  - For a test conducted at a temperature of  $10^{\circ}$ C, a factor of 15.5 is used to convert from the calculated permeation rate at end of life (factor of 2) and maximum prolonged material temperature (factor of 7.75) to that of a new container at  $10^{\circ}$ C.

#### 10.8.2 Basic Method

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.  $20^{\circ}$ C. Values will also be derived for alternative test temperatures of 15 or  $10^{\circ}$ C, as the draft EU Regulation [11] defines ambient temperature (at which the permeation test is carried out) as  $20^{\circ}$ C ±  $10^{\circ}$ C.

The perfect mixing equation can be used to calculate the hydrogen release rate required to give a steady state hydrogen concentration [34]:

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

where:

 $C_{\%}$  = Steady state gas concentration (%)

 $Q_{\underline{a}} = \text{Air flow rate (m<sup>3</sup>/min)}$ 

 $Q_g$  = Gas leakage rate (m<sup>3</sup>/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 (NmL/hr/L water capacity) at a test temperature of x<sup>0</sup>C,

V = Water capacity of hydrogen storage (L),

 $f_a$  = Aging factor, taken to be 2,

 $f_t$  = Test temperature factor = 6.0 at a test temperature of 20<sup>o</sup>C, or 7.0 at 15<sup>o</sup>C, or 7.75 at 10<sup>o</sup>C.

In the draft EU Regulation (comitology document), Annex IV, Appendix 3, Section 4.2.5 [11] the allowable component leak rate is 10NmL/hr per component (or per metre of flexible fuel line) and is at worst case conditions for the component considering various combinations of maximum and minimum temperature and pressure. As the rate is a notional rate per type approved component and assuming 30 such components for a car system and 90 for a city bus system, the combined leak rate typically represents between 3 and 5% (1% or less for a city bus) of the total allowable hydrogen discharge into the HySafe garage scenarios and can be neglected. It is also negligible in comparison to the other assumptions made.

#### 10.8.3 Theoretical Permeation Rates

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 9.

Minimum Testing Temperature ( <sup>0</sup> C)	Maximum Allowable Permeation Rate (NmL/hr/L water capacity)			
	Passenger Car	City Bus		
10	2.7	2,3*		
15	3.1	2,5*		
20	3.5	2,9*		

Note: \* See Section 10.8.4

 Table 9:
 Theoretical Allowable Permeation Rates

#### 10.8.4 Proposed 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 10.

Table 10 is based on the HySafe methodology and the figures were published in presentations made to the UN ECE WP29 HFCV-IG SGS and the EC Hydrogen Working Group on 21<sup>st</sup> and 27<sup>th</sup> January 2009 respectively. Section 11 contains a revised estimation based on new testing data made available after the January presentations which was the basis of the presentation to the EC Hydrogen Working Group on 10<sup>th</sup> March 2009.

New Or	Minimum Testing	Maximum Allowable
Simulated	Temperature (°C)	Permeation Rate
End Of Life		(NmL/hr/L water
Container		capacity)
	10	2.7*
New	15	3.1*
	20	3.5*

Note: \* The value to be adopted depends on the definition of ambient temperature, i.e. with a definition of  $20\pm10^{\circ}$ C the allowable permeation rate should be 2.7NmL/hr/L, but if the test is specified at  $15^{\circ}$ C the allowable permeation rate would be 3.1NmL/hr/L.

 Table 10:
 Originally Proposed Allowable Permeation Rate (See Section 11)

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.

#### 10.8.5 Units

"NmL/hr/L water capacity", is this the best specification of the allowable permeation rate? The alternative approach adopted by SAE, and in many respects a better approach, is to consider an absolute rate representing a total vehicle permeation rate covering all vehicles in a particular category, e.g. passenger cars.

In the context of the draft EU Regulation, the focus is on component level approval of the container rather than vehicle level tests. Additionally a container manufacturer may develop containers that are used in a variety of vehicles by different vehicle manufacturers and in this context a rate more directly linked to the size of the container is more practical, e.g. NmL/hr/L water capacity. However, using the water capacity, i.e. volume of the container, is not necessarily the best approach. A more appropriate measure of the size of the container would be surface area, i.e. per square metre of container internal surface area. However, historically the measure used has been per unit volume of the container as a simple number.

#### 11. UPDATED ESTIMATION OF THE HYSAFE ALLOWABLE PERMEATION RATE

This section describes the revised HySafe estimation which was published in a presentation to the EC Hydrogen Working Group on 10<sup>th</sup> March 2009 and is based on new testing data made public after the January presentations. It also incorporates the changes made in Corrigendum 1.

#### 11.1 Introduction

Following the HySafe presentations made to the UN ECE WP29 HFCV-IG SGS on 21 January 2009 and the EC Hydrogen Working Group on 27 January 2009, GM Powertrain Germany published new material temperature/permeation rate data [35] for the subsequent meeting of the EC Hydrogen Working Group on 10 March 2009. In addition further research

has been undertaken regarding the affect of aging. As a result, the rates originally proposed by HySafe have been optimised using the original methodology.

The work is also covered in a paper submitted to the 3rd International Conference on Hydrogen Safety, 16-18 September 2009, Corsica, France, see Section 17.

#### 11.2 New Testing Data

#### 11.2.1 Aging

As explained in Sections 10.6 and 10.8, the allowable end of life permeation rate has been reduced by a factor of 2 for a new container based on early research by LLNL [6]. 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, and weak points, e.g. the interface with the metal end boss [36]. Other organisations suggest that the phenomena has been observed [37] and also that a factor of 2 may not be adequate [38]. 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 [39] 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

#### 11.2.2 Test Temperature

As explained in Sections 10.6 and 10.8, 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. GM Powertrain Germany published new material temperature/permeation rate data for three different materials from GM and JARI experimental studies for the meeting of the EC Hydrogen Working Group on 10 March 2009 [35], see Figure 12.



Figure 12: Material Temperature/Permeation Characteristics

Based on the new data, the following factors were estimated:

- For a test conducted at a temperature of 20°C, a factor of 3.5 is used to convert from the maximum prolonged material temperature to the test temperature.
- For a test conducted at a temperature of 15°C, a factor of 4.7 is used to convert from the maximum prolonged material temperature to the test temperature.
- For a test conducted at a temperature of 10<sup>o</sup>C, a factor of 6.7 is used to convert from the maximum prolonged material temperature to the test temperature.

In addition to the proposal for the maximum allowable permeation rate, it is also proposed that the test temperature is more closely defined, since the actual permeation rate is temperature dependant. At present in the draft EU Regulation [11] the test temperature is stated in Section 4.1.12.2 as "ambient temperature", which under "Definitions" is stated as: "Ambient temperature" means a temperature range within  $20 \ C \pm [10] \ C$ .". It is proposed that the test temperature is defined more tightly, with a specific value and a small tolerance for normal laboratory temperature variations, i.e.  $20 \ C \pm 2 \ C$ .

#### **11.3 Updated Allowable Permeation Rate**

The calculation of the updated allowable permeation rate is based on the same methodology described in Section 10.8, except that factors used to derive a rate based on a specific test temperature have been revised as indicated in Table 11. Very few scientific results have been published, so the factor for temperature is based on the data published by GM (see Section 11.2.2) and the factor for aging is arbitrary. These factors should be reviewed as and when further results become available.

Test Temperature (ºC)	Material Temperature Factor (End of life to New)	Aging Factor	Combined Factor
10	6.7	2.0	13.4
15	4.7	2.0	9.4
20	3.5	2.0	7.0

 Table 11:
 Updated Temperature & Age Factors

Based on the above, the updated proposed permeation rates are given in Table 12. <u>Table 12</u> is based on the HySafe methodology and new material temperature/permeation data and was the basis of a presentation for the EC Hydrogen Working Group on 10<sup>th</sup> March 2009. It also incorporates the changes made in Corrigendum 1.

New Or	Minimum	Presentation Dated 10/03/09	Corrigendum 1
Simulated	Testing	Updated Allowable Permeation	Dated 15/06/09
End Of Life	Temperature	Rate	Allowable Permeation Rate
Container	$(O^{0})$	(NmL/hr/L water capacity)	
	10	3.1*	4.2* NmL/hr/L water capacity
New	15	4.6*	6.0* NmL/hr/L water capacity
	20	6.0*	8.0* NmL/hr/L water capacity
Simulated	55 .	00	90 NmL/min per standard
End of Life	55+	90	passenger vehicle

Note: \* The value to be adopted depends on the definition of ambient temperature, i.e. with a definition of  $20\pm10^{\circ}$ C the allowable permeation rate should be 4.2NmL/hr/L, but if the test is specified at  $20^{\circ}$ C the allowable permeation rate would be 8.0NmL/hr/L.

Table 12: Proposed Allowable Permeation Rate

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

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 as appropriate, in the relevant vehicle regulations or standards are necessary to avoid the potential development of flammable hydrogen/air mixtures.

#### **11.4 Proposed Text For The EU Regulation**

It is proposed that the text in the EU Regulation [11] is amended as follows:

Annex IV, Appendix 2,

#### 4.2.12. PERMEATION TEST

#### 4.2.12.1. Sampling

The test applies to Type 4 containers only. Type approval testing - number of finished containers to be tested: 1

#### 4.2.12.2. Procedure

Special consideration shall be given to safety when conducting this test.
The container shall be tested in the following sequence:
a) Pressurize with hydrogen gas to nominal working pressure,
b) Place in an enclosed sealed chamber at ambient temperature 15 °C ± 2 °C and monitor for permeation for ≥ 500 hours.

#### 4.2.12.3. Requirement

The steady state permeation rate shall be less than <del>1.0</del> 6.0 Ncm3 per hour of hydrogen per litre internal volume of the container.

#### 4.2.12.4. Results

The steady state permeation rate shall be presented in a test summary, e.g. [reference to table on 'container specifications and test data' in info. doc. to be inserted] of appendix 1 to Annex II.

The above proposal is based on the HySafe methodology, new material temperature/permeation data and the changes in Corrigendum 1, presented to the EC Hydrogen Working Group on 16 June 2009.

Part III

### COMPARISON OF HYSAFE AND OTHER PROPOSALS

#### 12. ALTERNATIVE PROPOSALS FOR AN ALLOWABLE PERMEATION RATE

The HySafe proposal is primarily intended to replace the figure currently quoted in the draft European Regulation [11 & 40], formerly the draft UN ECE regulation (in turn often referred to as the EIHP proposal) for the storage of compressed gaseous hydrogen on-board road vehicles [7 & 8] and versions of draft ISO/DIS15869: Gaseous Hydrogen And Hydrogen Blends – Land Vehicle Fuel Tanks [13 & 14] recently superseded by ISO/TS15869:2009 [12]. In addition the value may be adopted in the container requirements for the GTR. The appropriate section of the draft EU Regulation (Annex IV, Appendix 2, 4.2.12) is as follows:

#### 4.2.12. PERMEATION TEST

*4.2.12.1. Sampling The test applies to Type 4 containers only. Type approval testing - number of finished containers to be tested: 1* 

4.2.12.2. Procedure

Special consideration shall be given to safety when conducting this test.
The container shall be tested in the following sequence:
a) Pressurize with hydrogen gas to nominal working pressure,
b) Place in an enclosed sealed chamber at ambient temperature and monitor for permeation for ≥ 500 hours.

4.2.12.3. Requirement

The steady state permeation rate shall be less than 1.0 Ncm3 per hour of hydrogen per litre internal volume of the container.

4.2.12.4. Results

The steady state permeation rate shall be presented in a test summary, e.g. [reference to table on 'container specifications and test data' in info. doc. to be inserted] of appendix 1 to Annex II.

Note: In the current draft of the EU Regulation ambient temperature is defined as  $20^{\circ}C \pm 10^{\circ}C$ .

The various alternative proposals for an allowable permeation rate are shown in Table 13 together with the test conditions at which they are specified.

Section 13 provides a comparison of the three primary justifications behind the rates listed under reference numbers 1, 2, 3, 4 & 7 in Table 13. Section 14 provides a comparison between the HySafe proposal and the SAE J2579: January 2009 and ISO/TS15869: 2009 Option ii) rates.

Ref.	Source	Justification Reference	New Or Simulated End Of Life Container	Minimum Testing Temperature ( <sup>0</sup> C)	Maximum Allowable Permeation Rate (NmL/hr/L water capacity – except where indicated)
1	HySafe Proposal	See Part II		10*	4.2*
		of this	New	15*	6.0*
		report		20*	8.0*
	Alternative	See Pt III of this report	Sim. End of life	Min.55	90 NmL/min per standard passenger vehicle
2	Early ISO15869 & draft ECE	LLNL [6]	New	Ambient	1.0
3	Draft EU Reg	LLNL [6]	New	20±10	1.0
4	ISO/DIS15869.2&.3 & ISO/TS15869:2009 Option i) Test B16	JARI (2004) [9]	New	Ambient	2.0@35MPa & 2.8@70MPa
5	ISO/TS15869:2009 Option ii) Test E5	-	Simulated end of life	20	75 NmL/min per container
6	JARI for GTR	-	?	15	5
7	Initial ACEA proposal for EU Regulation	LLNL [6]	New	20±10	10
8	SAE J2579: Jan. 2009	-	Simulated end of life	Min. 55	150 NmL/min per standard vehicle

Notes:

\*

Value to be adopted depends on the definition of ambient temperature, i.e. with a definition of 20±10<sup>0</sup>C the allowable permeation rate should be 4.2NmL/hr/L, but if the test is specified at 20<sup>0</sup>C the allowable permeation rate would be 8.0NmL/hr/L.

Table 13: Alternative proposals For An Allowable Permeation Rate

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### 13. COMPARISON OF HYSAFE, LLNL & JARI(2004) SCENARIOS

Table 14 provides a direct comparison of the three different justifications used for the various rates referred to in Table 13.

	Table 14 Comparison Of HySafe, LLNL & JABI(2004) Scenarios							
	Factor	LLNL	JARI (2004)	HySafe	Comments			
1	Dispersion behaviour of permeated hydrogen	Homogeneous	Homogeneous	Homogeneous	Based on experimental and numerical work by HySafe, hydrogen can be assumed to disperse homogeneously at the ventilation rates and hydrogen flow rates considered in the permeation scenarios.			
2	Enclosure Scenarios							
2.1	Garage dimensions	1 domestic garage	1 domestic garage	<ul> <li>3 domestic garages:</li> <li>For a large car</li> <li>For a small car</li> <li>Smallest prefabricated garage</li> <li>2 city bus facilities</li> <li>Maintenance</li> <li>Minimum storage space</li> </ul>	See Table 15 for details			
2.2	Reasonable minimum ventilation rate	0.18ac/hr	0.18ac/hr	0.03ac/hr (Harmonised with SAE FC Safety WG)	<ul> <li>Some European markets <u>do not</u> <u>have any minimum natural</u> <u>ventilation requirements</u> for garages</li> <li>17 measurements of the natural ventilation rate in <u>real world</u> domestic garages have been identified including: <ul> <li>3 ≤0.18ac/hr</li> <li>7 ≤0.4ac/hr</li> </ul> </li> </ul>			

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	Table 14						
	Factor		JARI (2004)	HySafe	Comments		
					<ul> <li>1 additional measurement was taken in a well sealed experimental facility (0.009ac/hr)</li> <li>In normal circumstances it is assumed that the city bus facilities would have forced ventilation. The scenarios are included to consider "what if" concerns.</li> </ul>		
2.3	Maximum prolonged material temperature (°C)	82ºC	Not stated	55 <sup>°</sup> C (Harmonised with SAE FC Safety WG)	-		
3	Vehicle Scenarios				See Table 15 for details		
3.1	Туре	SUV	Small car	<ul> <li>Large car, e.g. Volvo S80</li> <li>Small car, e.g. Ford Fiesta</li> <li>Micro-car</li> <li>City Bus (full size, single decker)</li> </ul>	-		
3.2	Hydrogen storage pressure (MPa)	35MPa	Assumed 35 & 70 MPa	Cars: 70MPa City buses: 35 & 70MPa	-		

	Table 14							
	Factor		JARI (2004)	HySafe	Comments			
3.3	Hydrogen mass (kg)	13kg	1.4 & 2.4kg	Cars: 10*, 6* &3 <sup>#</sup> kg Buses: 50**kg	Sources: * StorHy [17] ** HyFleet/CUTE [18] # Estimated Note: GM HydroGen4 SUV has a 70MPa system with a capacity of 4.2kg giving a range of 320km [16]. On this basis 10kg would give a range of 760km comparable to conventional vehicles.			
3.4	Hydrogen capacity (L)	540L	60L	Cars: 75, 149, 249L Buses: 1244 & 2082L	See 3.3 above			
4	Test Procedures							
4.1	New or end of life container	New	Not stated, assumed new	New	Assume that permeation from an end of life container is twice that of a new container [6].			
4.2	Test pressure	Nominal working pressure	Nominal working pressure	Nominal working pressure	-			
4.3	Test temperature	Room temperature	Ambient temperature	20 or 15 or 10 <sup>0</sup> C	Test temperature depends on definition of ambient temperature, e.g. for the EC regulation it should be assumed to be at $10^{\circ}$ C if ambient is to be defined as $20 \pm 10^{\circ}$ C			

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	Table 14 Comparison Of HvSafe, LLNL & JARI(2004) Scenarios								
	Factor	LLNL	JARI (2004)		Comments				
5	Level Of Safety								
5.1	Philosophy	Similar to HySafe	Not indicated	If test is undertaken on a new container at ambient temperature, then the permeation rate must be set such that the permeation rate is safe at the maximum prolonged temperature for an end of life container	-				
5.1	Max. hydrogen concentration	2%	Not explicitly stated	1%*	* Harmonised with SAE FC Safety WG and general explosive atmosphere standards, e.g. IEC. The value of 1% is conservative but allows for variations in the other parameters. In addition permeation is a long term build up and is different to specific localised short term releases at up to 4% concentration as proposed by OICA for the GTR.				
5.3	Material temperature/ permeation relationship	Factor of 10 between 24 and 82°C	Not indicated	Original proposal: Extrapolation/ interpolation of a straight line based on the LLNL data. Updated proposal: From Figure 12	The original HySafe assumption is conservative but not excessively so, the extent depending on the material, the test temperature and the maximum prolonged material temperature. The value must allow for the behaviour of different liner materials as a specific material is not stated in the draft requirements.				

Source	LLNL	JARI			HyS	Safe		
		(2004)	Ca	ar Scenari	OS	Βι	Bus Scenarios	
	SUV	Small Car	Large Car	Small Car	Min. Garage	35MPa Bus Maint. Garage	70MPa Bus Maint. Garage	Min. Bus Garage
Enclosure Length (m)	5.0*	6.00	6.5	5.0	3.7	16.00	16.00	12.60
Enclosure Width (m)	3.0*	2.43	3.5	3.0	2.4	6.55	6.55	3.55
Enclosure Height (m)	2.0*	2.40	2.2	2.2	2.1	6.50	6.50	5.50
Enclosure Volume (m <sup>3</sup> )	30	36	50	33	19	681	681	246
Solid Material Vol.** (m <sup>3</sup> )	-	-	4	2	1	5	5	5
Free Vol. *** (m <sup>3</sup> )	30	36	46	31	18	676	676	241
Storage pressure (MPa)	35	35 & 70	70	70	70	35	70	35 (worst)
Hydrogen Stored (kg)	13	1.4* & 2.4*	10	6	3	50	50	50
Storage Volume (L)	540	60	249	149	75	2082	1244	2082 (worst)

Notes:

Hydrogen density at  $35MPa/15^{\circ}C = 24.02kg/m^{3}$  [20] Hydrogen density at  $70MPa/15^{\circ}C = 40.18kg/m^{3}$  [20] i)

ii)

\* Estimated from volume iii)

\*\* Volume of impermeable materials, tyres, etc. includes the volumes of those parts of the vehicle iv) without the possibility of air movement (assuming that hydrogen can enter the passenger and other compartments). Assume 4m<sup>3</sup>, 2m<sup>3</sup> and 1m<sup>3</sup> for large, small and micro cars respectively, and 5m<sup>3</sup> for a city bus.

\*\*\* Free volume in facility = Facility Volume - Volume of impermeable materials, tyres, etc V)

vi) European minimum garage based on smallest garage that is easily available

Table 15: Summary Of Scenarios

#### 14. COMPARISON OF HYSAFE, SAE J2579: 01/09 & ISO/TS15869 OPTION ii) RATES

#### 14.1 Requirements

#### 14.1.1 HySafe

The HySafe proposals for the allowable permeation rates are given in Section 11.4 and are based on the draft EU regulation (see Section 12 for the requirements in the current draft [11]).

#### 14.1.2 SAE J2579 (January 2009)

The requirements of SAE J2579, Jan. 2009 "Technical Information Report For Fuel Systems In Fuel Cell And Other Hydrogen Vehicles", section 5.2.2.1.3 [3] are:

"The fully filled storage system shall be held at a temperature of at least 55 °C to stabilize and measure the total discharge rate due to leakage and permeation according to procedures given in Appendix C.7. This test may be conducted coincidentally with the last half of testing in 5.2.2.1.2 (at 85 °C) or after testing in 5.2.2.1.2 is completed with the system temperature held at least 55 °C for the measurement. The maximum allowable discharge from the compressed hydrogen storage system is 150 Ncc/min for standard passenger vehicles. The maximum allowable discharge for systems in larger vehicles is R\*150 Ncc/min where R = (Vwidth+1)\*(Vheight+0.5)\*(Vlength+1)/30.4 and Vwidth, Vheight, Vlength are the vehicle width, height, length (m), respectively."

The SAE test is representative of an end of life container.

#### 14.1.3 ISO/TS15869 OPTION ii)

The requirements of ISO/TS15869: 2009 "Gaseous hydrogen and hydrogen blends — Land vehicle fuel tanks" Annex E, E.5 [12] are:

"The fuel tank shall be pressurized with hydrogen gas to at least working pressure in an enclosure to determine that the steady state hydrogen discharge rate due to leakage and permeation does not exceed 75 cm3/min (at 20 °C and 101,325 kPa) for use in standard passenger cars. For fuel tanks to be used in larger vehicles, such as buses, the allowable leakage may be increased in proportion to the enclosure volume for the vehicle. This test may be conducted coincidentally with the last half of the accelerated static stress test in E.4."

The ISO Option ii) test is representative of an end of life container.

#### 14.2 Fundamental Differences Between The Rates

There are a number of fundamental specification differences between the rates in the draft EU Regulation compared with SAEJ2579 and the ISO/TS15869 Option ii) test, including:

- i) In the draft EU Regulation the allowable rate is linked to the size of the hydrogen storage system as determined by its water capacity. Whereas in the SAE standard the rate is given for a vehicle regardless of the size of the hydrogen storage, however, larger vehicles may be catered for by increasing the size of the test enclosure in relation to the size of the vehicle. In ISO/TS15869 Option ii) the rate is per container regardless of its size and regardless of the number of containers fitted to a vehicle.
- ii) In the draft EU Regulation the allowable rate is for a new container at "ambient temperature", i.e. 20±10°C in the current draft. In SAE J2579 the permeation rate is for a maximum prolonged material temperature of at least 55°C and near the end of an expected service performance test sequence, i.e. the container should be in a condition

similar to that of an end of service life container. In ISO/TS15869 Option ii) the permeation rates is for a container at  $20^{\circ}C$ 

iii) In the EU regulation the allowable rate is quoted per hour, whereas in the SAE and ISO standards it is quoted per minute.

#### 14.3 Comparison Of The Allowable Permeation Rates

Table 16 shows the allowable permeation rates for the three HySafe car scenarios calculated using the HySafe methodology at the SAE, ISO and draft EU Regulation test conditions and rate specification. It can be seen from the table that the critical scenario using the EU Regulation test conditions and rate specification is the large car scenario whereas for the SAE and ISO conditions the critical scenario is the "minimum" garage. This is because the approach in the EU regulation is more critical on a per litre water capacity basis for large capacity systems, while the SAE approach uses a single rate irrespective of the storage system size which is more critical the smaller the garage size.

		HySafe Scenario		Relationship To	
		Large Car	Small Car	Min. Garage	Storage
Free Volume in Enclosure (m <sup>3</sup> )		46 (Corr.1)	31	18	Capacity
Hydrogen Storage Volume (L)		249	149	75	
Maximum Hydrogen Concentration (%)		1	1	1	
Natural Ventila	tion Rate (ach/hr)	0.03	0.03	0.03	
Allowable Permeation Rate Using HySafe Methodology & Scenarios With The Indicated Test Conditions & Rate Specification	SAE J2579 Test Conditions & Rate Spec. [EoL @ MPMT*] (NmL/min)	232	157	91	Per standard passenger vehicle
	ISO Option ii) Test conditions & Rate Spec. [EoL* @ 20 <sup>0</sup> C] (NmL/min)	66	45	26	Image: Image and the image a
	EU Reg. Test Conditions & Rate Spec. [New @ 10 <sup>0</sup> C] (NmL/hr/L)	4.2	4.7	5.4	
	EU Reg. Test Conditions & Rate Spec. [New @ 15 <sup>o</sup> C **] (NmL/hr/L)	6.0	6.8	7.9	Per L of storage capacity
	EU Reg. Test Conditions & Rate Spec. [New @ 20 <sup>0</sup> C] (NmL/hr/L)	8.0	9.1	10.5	

Notes: \* EoL = Simulated end of life, MPMT = Maximum prolonged material temperature \*\* Similar to ISO/TS15869 Option i)

Table 16: Allowable Permeation Rates Using The HySafe Methodology And Scenarios At The SAE, ISO And Draft EU Regulation Test Conditions/Rate Specification Why may 150NmL/min be considered more appropriate for SAE J2579 than 90NmL/min? The question may be answered by considering the realistic scenarios for each market. In Europe, garages of the size considered under the "minimum garage" scenario are readily available as prefabricated self-standing units. It would also be reasonable to assume that small prefabricated units have a relatively poor level of construction in terms of weather sealing, i.e. airtightness, compared to those directly attached to houses. In other words the minimum level of ventilation could reasonably be expected to be greater than 0.03ac/hr. In contrast in a North American context, the small car garage (30.4m<sup>3</sup> is quoted in J2579 compared with the 31m<sup>3</sup> HySafe "small" garage) may be considered to be more appropriate as the smallest reasonable garage and it may not be appropriate to set North American specific requirements in terms of European scenarios or vice versa. The rate should be considered further by SAE.

A more critical issue relates to the ISO Option ii) rate. It was understood that ISO/TS15869 Option ii) [12] was based on SAE J2579 (January 2008) [15], however, with respect to the permeation test there is a fundamental and significant difference between the rates specified in the ISO and SAE standards, see Table 17. A comparison between the relevant standards shows two major differences between the SAE test and the ISO Option ii) test:

- i) The test temperature is reduced from 85/55 °C in the SAE document to 20°C in the ISO document.
- ii) The SAE rates are per vehicle whereas the ISO rate is per container and it is likely that a typical passenger car would use more than one container, probably two but say four as a reasonable upper limit.

Taken together the changes introduced into the ISO Option ii) rate imply a significant relaxation of the hydrogen permeation rate in comparison to the SAE standard on which it is understood that it was based. If the ISO Option ii) rate is considered in relation to the HySafe Scenarios it represents an increase in the allowable permeation in comparison to the HySafe proposal by a factor of 1.1 to 3.0 if only one container is fitted per vehicle but an increase of 3 to12 times if four containers are fitted per vehicle. In addition there is a significant inconsistency between the rates given in Options i) and ii). <u>On this basis it is proposed that the test specified in ISO/TS15869: 2009 "Gaseous hydrogen and hydrogen blends — Land vehicle fuel tanks" Annex E, E.5 [12] is reviewed as a matter of priority.</u>

Standard	Status	Allowable Permeation Rate (NmL/min)	Test Temperature (°C)	New Or Simulated End Of Life (New or EoL)
SAE J2579 (Jan. 2009) [3]	Current	150 per vehicle	55	EoL
SAE J2579 (Jan. 2008) [15]	Superseded	75 per vehicle	85	EoL
ISO/TS 15869 Option ii) [12]	Current	75 per container	20	EoL

Table 17: Comparison Of SAE & ISO Allowable Permeation Rates & Test Conditions

Part IV

## CONCLUSIONS AND RECOMMENDATIONS

#### 15 CONCLUSIONS AND RECOMMENDATIONS

The work undertaken in HySafe IP1.3 has:

- Addressed a number of important issues in relation to hydrogen permeation in general and automotive applications in particular,
- 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 will be presented to ISO TC197 WG6 and the SAE Fuel Cell Safety Working Group,
- Identified a significant issue with the allowable rate stated in ISO/TS15869: 2009 Option ii),
- Established global discussions involving the vehicle manufacturers, SAE and key regulatory bodies.

Any allowable discharge rate for hydrogen has to be based on a number of key factors:

- 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 such that 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 it. 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. Additionally for hydrogen, vehicle regulations will be determined at a European or global level, while building regulations are usually determined at national level or even at a local level depending on the iurisdiction.

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.

System leaks have not been considered in this work, however, the permitted component leak rates for type approval in the draft EU Regulation were found to be negligible in comparison to the allowable release rate. The SAE J2579 proposal is for a combined permeation and leak rate for a vehicle which is more useful.

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 real world measurements and research conducted for this report, a reasonable minimum natural ventilation rate has now been agreed with the SAE as 0.03ac/hr. Similarly a reasonable maximum prolonged material temperature has been researched and agreed with the SAE as 55°C.

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 differing reports from different sources and further research may be necessary in addition to the French national project ENDEMAT. As a result it is considered necessary to retain an arbitrary factor of two reduction between end of life and new containers which would provides some allowance allow for:

- Unknown aging effects,
- Use of new materials,
- 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 related to the reasonable minimum ventilation rate in garages and maximum prolonged material temperature have been harmonised in discussions with the SAE Fuel Cell Safety Working Group. Following presentation of early parts of the work, a reduction was made in SAE standards from a 1400NmL/min total discharge rate to 150NmL/min take into account smaller garages and cars, which is particularly important in a global context and increasingly in the USA as a result of the increasing oil price and general economic crisis. The rate should be considered further by SAE.

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.
- The allowable permeation rate will be specified in the same manner as the rate in the draft EC regulation and ISO/DIS15869.2, i.e. NmL/hr/L water capacity.
- Permeated hydrogen can be considered to disperse homogeneously after experimental and modelling work by the HySafe partners.
- 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 =  $55^{\circ}$ C.

Based on the above assumptions the allowable permeation rates given in Table 18 were **<u>originally</u>** proposed during presentations to the UN ECE WP29 HFCV-IG SGS and the EC Hydrogen Working Group in **January** 2009.

Minimum Testing	Original	
Temperature ( <sup>0</sup> C)	Maximum Allowable	
	Permeation Rate	
	(NmL/hr/L water capacity)	
10	2.7*	Superseded by
15	3.1*	values in the
20	3.5*	following table.

Note: \* The value to be adopted depends on the definition of ambient temperature, i.e. with a definition of  $20\pm10^{\circ}$ C the allowable permeation rate should be 2.7NmL/hr/L, but if the test is specified at  $15^{\circ}$ C the allowable permeation rate would be 3.1NmL/hr/L.

#### Table 18: Original HySafe Proposal For The Allowable Permeation Rate

Following those presentations, material temperature/permeation data provided by GM Powertrain Germany has allowed the original proposal to be optimised using the same methodology. As very few scientific results have been published, the factors for temperature and aging should be reviewed as and when further results become available. Based on the new data, revised allowable permeation rates were proposed and were presented to the EC Hydrogen Working Group on 10 March 2009 with the support of ACEA. The proposals were also presented to UN ECE WP29 HFCV-IG SGS on 29 May 2009, and provided to ISOTC197 WG6 and SAE Fuel Cell Safety Work Group. The values were subsequently updated by Corrigendum 1 to this report as shown in Table 19 and presented to the EC group on 16 June 2009.

New or Simulated End of Life Container	Minimum Testing Temperature ( <sup>0</sup> C)	Updated Corr.1 Maximum Allowable Permeation Rate
New	10	4.2* NmL/hr/L water capacity
New	15	6.0* NmL/hr/L water capacity
New	20	8.0* NmL/hr/L water capacity
Simulated end of life	55+	<b>90</b> NmL/min per standard passenger vehicle

Note: \* The value to be adopted depends on the definition of ambient temperature, i.e. with a definition of  $20\pm10^{\circ}$ C the allowable permeation rate should be 4.21NmL/hr/L, but if the test is specified at  $20^{\circ}$ C the allowable permeation rate would be 8.0NmL/hr/L.

 Table 19: Updated HySafe Proposal For The Allowable Permeation rate

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

The HySafe proposals for allowable hydrogen permeation rates are intended only for use in appropriate road 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 legal requirements and standards is given in Table 20.

Source	Justification Reference	New Or Simulated End Of Life Container	Minimum Testing Temperature (°C)	Maximum Allowable Permeation Rate (NmL/hr/L water capacity – except where indicated)
HySafe Proposal	See Part II		10	3.1
	of this	New	15	4.6
	report		20	6.0
Alternative	See Pt III of this report	Sim. End of life	Min.55	90 NmL/min per standard passenger vehicle
Early ISO15869 & draft ECE	LLNL [6]	New	Ambient	1.0
Draft EU Reg	LLNL [6]	New	20±10	1.0
ISO/DIS15869.2&.3 & ISO/TS15869:2009 Option i) Test B16	JARI (2004) [9]	New	Ambient	2.0@35MPa & 2.8@70MPa
ISO/TS15869:2009 Option ii) Test E5	-	Simulated end of life	20	75 NmL/min per container
JARI for GTR		?	15	5
Initial ACEA proposal for EU Regulation	LLNL [6]	New	20±10	10
SAE J2579: Jan. 2009	-	Simulated end of life	Min. 55	150 NmL/min per standard passenger vehicle

 Table 20: Alternative Proposals For An Allowable Permeation rate

A critical issue has been identified that relates to the allowable permeation rate in ISO/TS15869: 2009 Test E5. It was understood that the rate was based on SAE J2579 (January 2008), however, with respect to the permeation test there is a fundamental and significant difference between the rate specified in the ISO and SAE standards. A comparison of the two standards shows two major differences between the SAE test and the ISO Option ii) test:

- i) The test temperature is reduced from 85°C (now 55°C) in the SAE document to 20°C in the ISO document.
- ii) The SAE rate is per vehicle, whereas the ISO rate is per container and it is likely that a typical passenger car would use more than one container, possibly two but say four as a reasonable upper limit.

Taken together the changes introduced into the ISO Option ii) rate imply a significant relaxation of the hydrogen permeation rate in comparison to the SAE standard on which it is understood that it was based. If the ISO Option ii) rate is considered in relation to the HySafe Scenarios it represents an increase in the allowable permeation rate in comparison to the HySafe proposal by a factor of 1.1 to 3.0 if only one container is fitted per vehicle but an

increase of 3 to12 times if four containers are fitted per vehicle. In addition there is a significant inconsistency between the rates given in Options i) and ii). <u>On this basis it is proposed that the test specified in ISO/TS15869: 2009 "Gaseous hydrogen and hydrogen blends — Land vehicle fuel tanks" Annex E, E.5 is reviewed as a matter of priority.</u>

Another issue that should be addressed in the context of general hydrogen releases, but is beyond the scope of this study, is movement of released hydrogen from one part of a structure to another, e.g. from a garage to an attached dwelling.

#### 16 FURTHER RESEARCH

The following issues were identified that may warrant further research:

- Further research could consider whether there is justification for raising the commonly accepted 25% LFL threshold for allowable hydrogen concentrations given the specific behaviour of hydrogen.
- The effects of aging on the permeation behaviour of complete containers appear uncertain with differing reports from different sources and further research may be necessary in addition to the French national project ENDEMAT.
- A wider infrastructure issue, in the context of general hydrogen releases, is further research on the movement of released hydrogen from one part of a structure to another, e.g. from a garage to an attached dwelling and any specific building requirements that may be required in future.

### 17 RELATED PUBLICATIONS

The HySafe permeation work is also covered in a series of four papers that have been submitted to the 3rd International Conference on Hydrogen Safety, 16-18 September 2009, Corsica, France:

- Adams, P., et al, "Allowable Hydrogen Permeation Rate From Road Vehicle Compressed Gaseous Storage Systems In Garages; Part 1 – Introduction, Scenarios, And Estimation Of An Allowable Permeation Rate".
- Venetsanos A.G., et al, "Estimation of an Allowable Hydrogen Permeation Rate From Road Vehicle Compressed Gaseous Hydrogen Storage Systems In Typical Garages; Part 2 – CFD dispersion calculations using the ADREA-HF code and experimental validation using helium tests at the GARAGE facility".
- Saffers J-B., et al, "Estimation of an Allowable Hydrogen Permeation Rate from Road Vehicle Compressed Gaseous Hydrogen Storage Systems in Typical Garages; Part 3: Modelling and Numerical Simulation of Permeation in a Garage with Adiabatic Walls and Still Air".
- Cariteau B., et al, "Experiments on the distribution of concentration due to buoyant gas low flow rate release in an enclosure".

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- Commissariat à l'Énergie Atomique (France): Alain Bengaouer, Isabelle Tkatschenko and Benjamin Cariteau
- National Center for Scientific Research Demokritos (Greece): Alexandros Venetsanos and Efi Papanikolaou

• University of Ulster (UK): Vladimir Molkov, Dmitriy Makarov and Jean-Bernard Saffers In addition the work has involved collaboration with the SAE Fuel Cell Safety Work Group and in particular Glenn Scheffler allowing harmonisation of some of the key assumptions, e.g. garage ventilation rate, maximum prolonged material temperature, etc.

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#### APPENDIX 1 PRESENTATION TO THE EC HYDROGEN WORKING GROUP ON 27 JANUARY 2009

Note: The presentation to the UN ECE WP29 HFCV-IG SGS on 21 January 2009 was similar.

EC Hydrogen Regulation Working Group					
Allowable Hydrogen Permeation Rate For Automotive Applications					
	By: Paul Adams, Volvo Technology, Sweden				
	In cooperation with the HySafe partners:				
	ceci			of R	
	&	SAE Fuel Cell Safety Wo	orkgroup		
Volvo Technology Dept 06120 1 27 Jan 2009		Hydrogen Permeation Paul Adams		VOLVO	





















The following assumptions have been made:

- The permitted permeation rate will be specified in the same manner as the rate in the draft EC proposal, 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°C

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#### APPENDIX 2 KEY EXTRACTS FROM PRESENTATIONS FOR THE EC HYDROGEN WORKING GROUP ON 10 MARCH 2009

Extracts from the presentation provided by P Adams, Volvo Technology on behalf of HySafe:

## **Revised Allowable Permeation Rate**

Based on the HySafe methodology & GM/JARI temperature/permeation data

Minimum Testing Temperature (ºC)	Max. Allowable Permeation Rate (NmL/hr/L water cap.)
10	3.1
15	4.6
20	6.0

Retains a factor of 2 reduction between end of life and new containers which would allow for:

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

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# **Revised HySafe Proposal**

4.2.12. PERMEATION TEST

4.2.12.1. Sampling The test applies to Type 4 containers only. Type approval testing - number of finished containers to be tested: 1

4.2.12.2. Procedure Special consideration shall be given to safety when conducting this test. The container shall be tested in the following sequence: a) Pressurize with hydrogen gas to nonjuer working pressure, b) Place in an enclosed sealed chamber at ambient temperature  $20^{\circ}C \pm 2^{\circ}C$  and monitor for permeation for  $\geq$  500 hours. 4.2.12.3. Requirement

The steady state permeation rate shall be less than <u>1.06.0</u> Ncm3 per hour of hydrogen per litre internal volume of the container.

4.2.12.4. Results The steady state permeation rate shall be presented in a test summary, e.g. [reference to table on 'container specifications and test data' in info. doc. to be inserted] of appendix 1 to Annex II.

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#### Extract from the presentation provided by V Rothe, GM Powertrain Germany:

#### Estimation of permeation requirements for 20 °C test temperature

Measured permeation data from GM and JARI · Tanks show different permeation at the same Permeation [ml/h/L] temperature levels, all below 36 ml/h/L. 35 · Different tanks show different relations between 30 25 temperature and permeation (exponential Arrhenius 20 function, depending on material properties and 15 10 activation energy). 5 0 Normalised permeation curves [6 ml/h/L @ 20°C] 80 100 Temperature [°C] 0 40 60 20 show that non of the tested liner materials would Normalised Permeation [6 ml/h/L @ 20°C] 60 exceed the 36 ml/h/L @ 55°C. 50 40 Conclusion: The criteria of 6 ml/h/L permeation rate 30 @ 20°C] is a very reasonable number to fulfill the 20 superior requirement of 1% maximum H2 10 concentration in the parking garage ! 0 0 20 40 60 80 100 Temperature [°C]

GM