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MODELLING AND NUMERICAL SIMULATION OF HYDROGEN PERMEATION IN A GARAGE WITH ADIABATIC WALLS AND STILL AIR

Prepared during the InsHyde Project within HySafe

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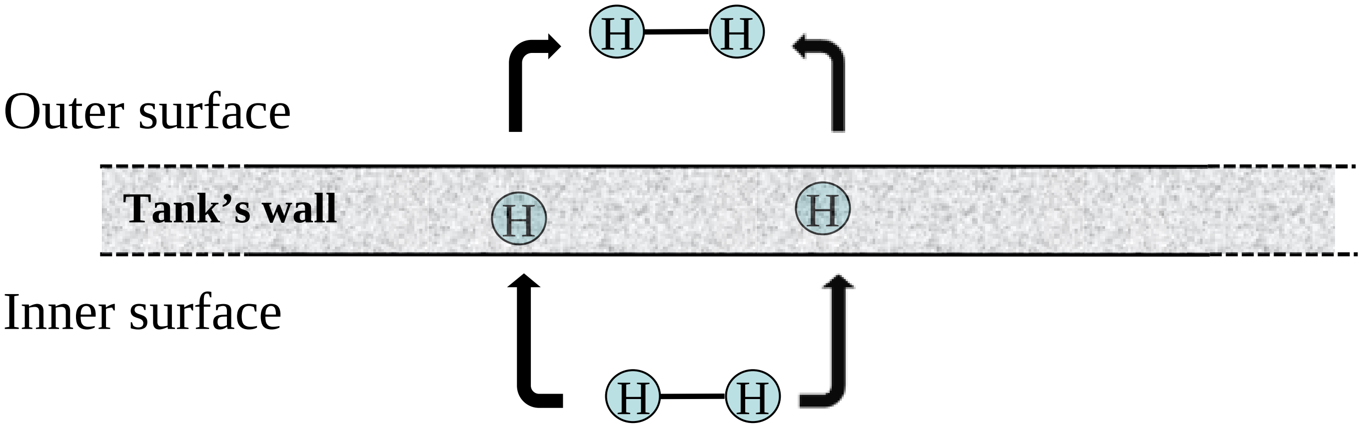
Phenomena of permeation

Permeation: overall process of a fluid crossing a membrane caused by a pressure difference.

Particularly relevant to hydrogen due to its:
- High diffusivity;
- Small molecular size;
- Small molecular weight;
- Low viscosity.



Engineering correlations

The permeability[1] ϕ is expressed in mol/s/m/Pa$^{1/2}$:

$$\phi = \phi_0 \cdot \exp(-E_\phi / R \cdot T)$$

ϕ - permeability (mol/s/m/Pa$^{1/2}$)

R - perfect gases universal constant (8.3144 J/mol/K)

T - external temperature (K)

ϕ_0 - pre-exponential factor (mol/s/m/Pa$^{1/2}$)

E_ϕ - activation energy (J/mol)

Material dependent

The rate of permeation[1] J is expressed in mol/s/m^2:

$$J = \phi \frac{\sqrt{p}}{L}$$

J - permeation rate of hydrogen (mol/s/m2)

ϕ - permeability of the material of the tank (mol/s/m/Pa$^{1/2}$)

p - tank pressure (Pa)

L - tank wall thickness (m)

Container dependent

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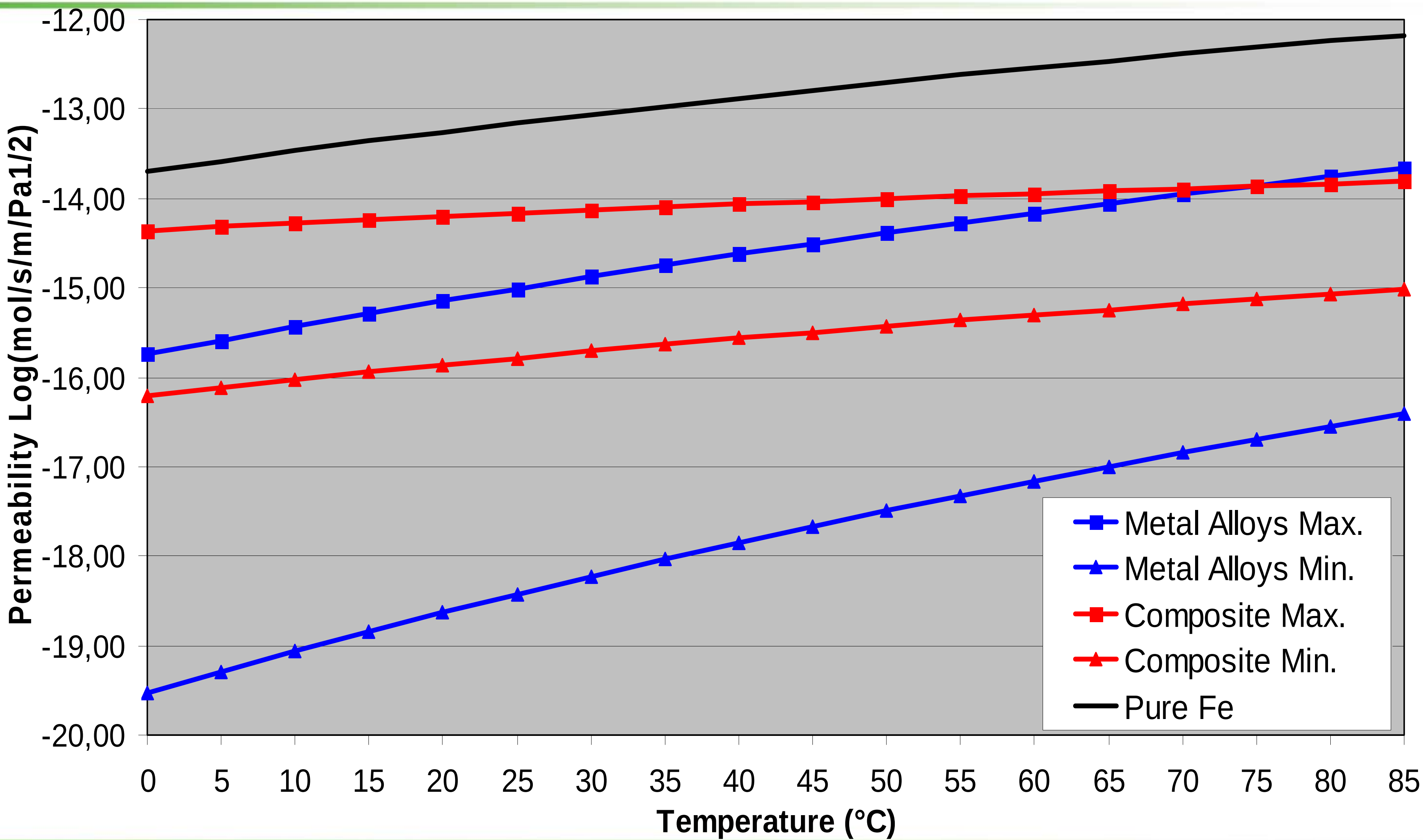
[1] Schefer et al., IJHE, 2006, Vol.31, pp.1247-1260

Comparison of permeabilities



Goals of this study

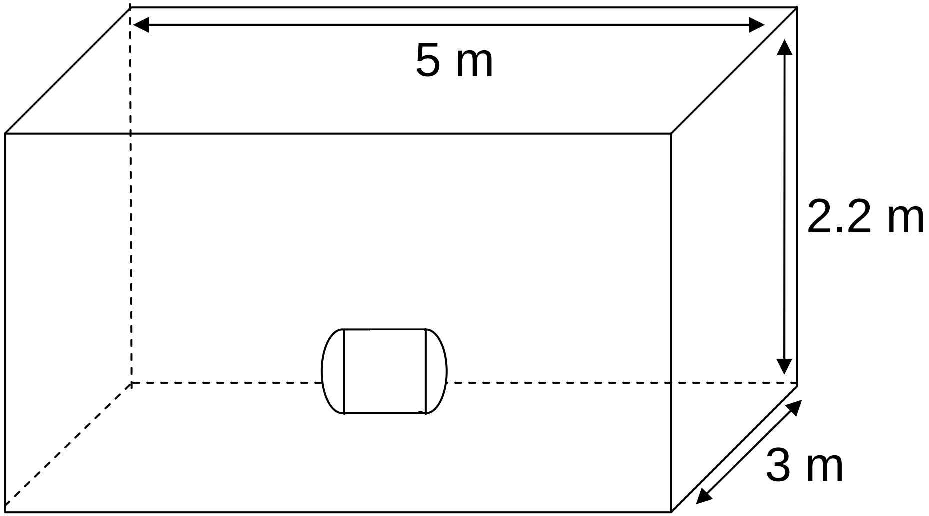
- **Safety concern with hydrogen permeation:** The formation of a flammable hydrogen-air mixture in closed space (e.g. a car in a garage with type IV compressed hydrogen tank).

- **HySAFER performed a simplified analysis to estimate:**
  - Hydrogen concentration on a tank surface as a function of time;
  - Hydrogen average concentration in an enclosure in assumptions of fully sealed garage and uniform hydrogen distribution.
- **HySAFER performed a numerical study to clarify:**
  - The interplay between hydrogen diffusion and buoyancy;
  - The distribution of permeated hydrogen with still air.

Case study

We choose a conservative approach for a tank in an assumed perfectly sealed garage.

- The garage : 5 m long, 3 m wide, and 2.2 m high.
- The tank[2]: 0.672 m long, 0.505 m diameter with two hemispherical ends with diameter of 0.505 m, 0.5m above ground. (Area=A_r, volume =V_r)
- Rate of permeation: J=1.40×10^{-6} mol·s^{-1}·m^{-2} or 1.14 NmL·hr^{-1}·L^{-1}, close to the value of the draft of the UN ECE Regulation for type IV containers (i.e 1.0 NmL·hr^{-1}·L^{-1}).



[2] A. Sarkar, R. Banerjee, IJHE, 2005, Vol. 30, pp.867–877

Initiation of leak

We use the Brownian Motion described by Einstein's law[3] to calculate the "*displacement of particles by diffusion in direction of the X-axis*" :

$$\lambda_x = \sqrt{\overline{\Delta x^2}} = \sqrt{2 \cdot D \cdot t}$$

D is the diffusion coefficient of H_2 in air ($m^2 \cdot s^{-1}$)
t is time (s)

It was hence possible to calculate the hydrogen concentration in a volume close to the tank's surface as a function of time, considering only diffusion.

Assuming uniform distribution of hydrogen molecules, the hydrogen concentration $[H_2]_t$ after time t, is the ratio of the volume of hydrogen over the total volume:

$$[H_2]_t = 100 \frac{J t A_r V_m}{\sqrt{2Dt}\, A_r} = 100 \frac{J V_m}{\sqrt{2D}} \times \frac{t}{\sqrt{t}}$$

The concentration on the surface increase with time as $[H_2]_t \propto \sqrt{t}$ until the buoyancy will overcome diffusion transport of hydrogen.

How to define this characteristic time?

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[3] Einstein, A. 1905, *Annalen der Physik,* vol. 17, pp. 549-560

Time to buoyancy

The idea is to define a characteristic time at which the displacement by buoyancy overcomes the displacement by diffusion. The second Newton's Law for buoyant motion of hydrogen-air mixture of density ρ_{mixt} in air of density ρ_{air} can be written as:

$$F = ma = (\rho_{air} - \rho_{mixt})g = \rho_{mixt}\frac{2L}{t^2}$$ Where $$\rho_{mixt} = \frac{[H_2]_t}{100}\cdot\left(\rho_{H_2} - \rho_{air}\right) + \rho_{air}$$

The displacement by buoyancy is equal to $$L = \left(\frac{\rho_{air}}{\frac{J\cdot t\cdot V_m}{\sqrt{2\cdot D\cdot t}}\left(\rho_{H2} - \rho_{air}\right) + \rho_{air}} - 1\right)\cdot\frac{g\cdot t^2}{2}$$

We can then calculate a time *t,* when the displacement of hydrogen by buoyancy equals the displacement by diffusion $\lambda_x = L$:

$$\sqrt{2\cdot D\cdot t} = \left(\frac{\rho_{air}}{\frac{J\cdot t\cdot V_m}{\sqrt{2\cdot D\cdot t}}\left(\rho_{H2} - \rho_{air}\right) + \rho_{air}} - 1\right)\cdot\frac{g\cdot t^2}{2}$$

At about 35 seconds, the displacement by buoyancy equals the displacement by diffusion. The hydrogen concentration on the surface for that characteristic time is $2\text{x}10^{-3}$% vol.

Modelling permeation leak (1/7)

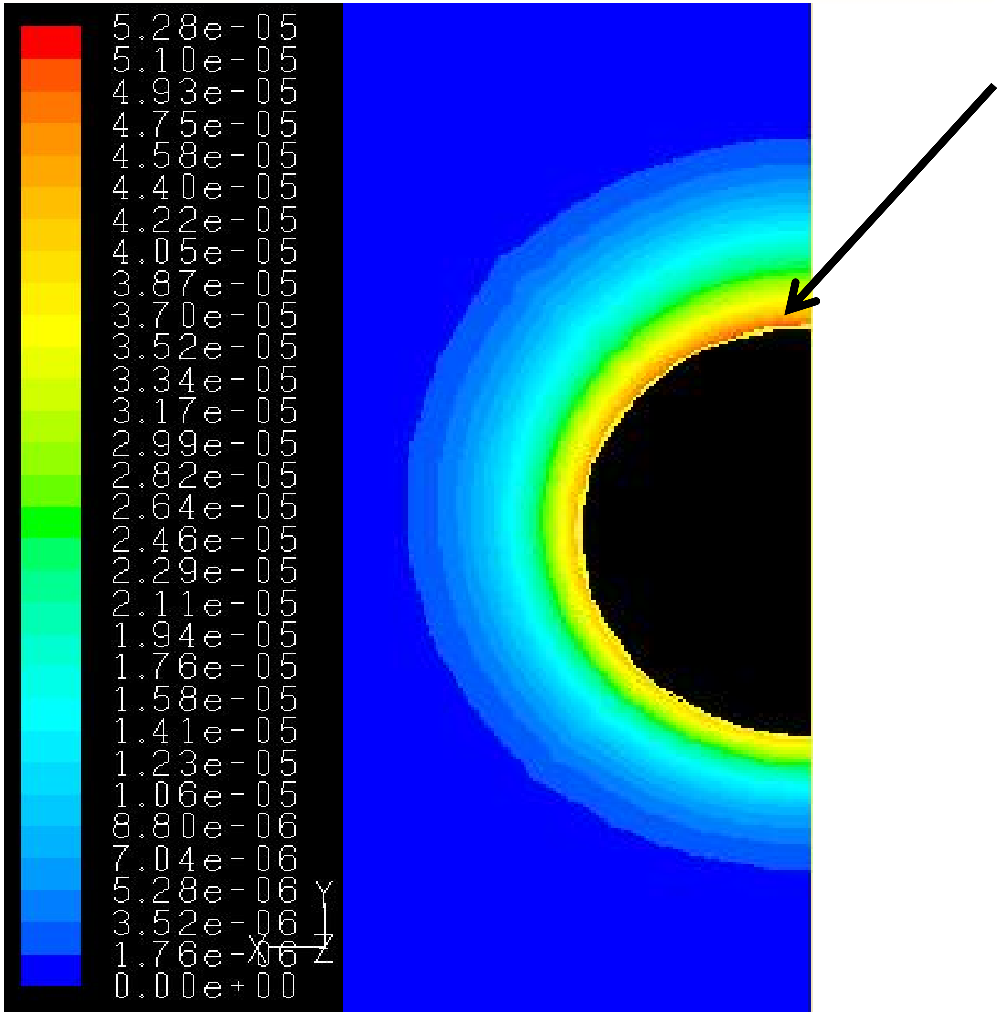
The hydrogen release was modelled using a tiny volumetric source of hydrogen in a thin layer (two computational cell of 0.5 mm thickness) around the whole surface of tank. This is different from modelling of permeation by artificial plumes/jets with a mass fraction Y_{H2}=1 at “release orifice” (our numerical experiments confirmed that there is no layer Y_{H2}=1 on the tank’s surface).
To match the specified permeation rate, the volumetric source term for hydrogen mass was $S_{H2}=2.61\times10^{-8}$ kg·m^{-3}·s^{-1}.

•3D unsteady laminar flow

•SIMPLE algorithm, 3rd order MUSCL discretisation scheme for convective terms, central difference for diffusion terms, 2nd order implicit time stepping

•Time step: D*t*=0.05s (max *V*=0.0215m/s, max Courant number *CFL*=0.06, max cell Reynolds number Re~100)

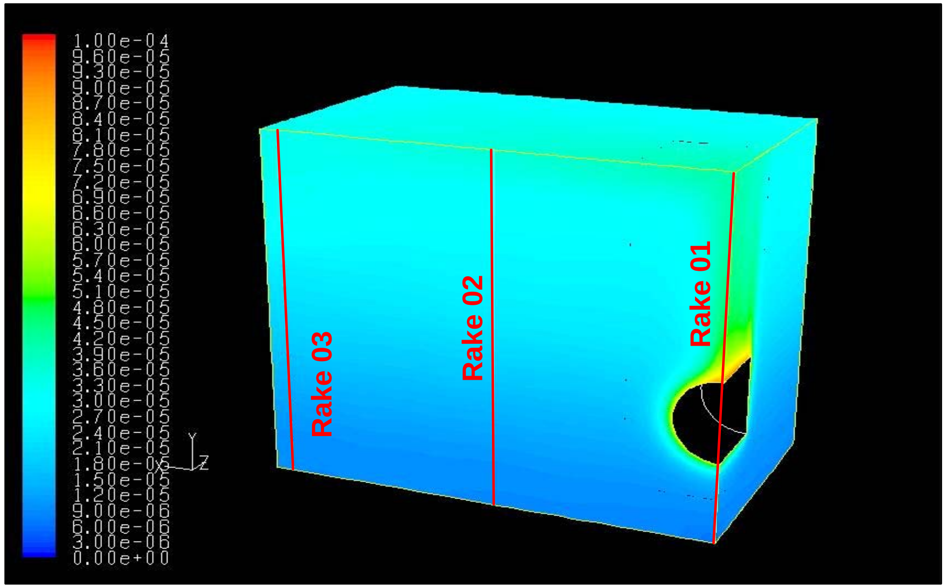
Modelling permeation leak (2/7)

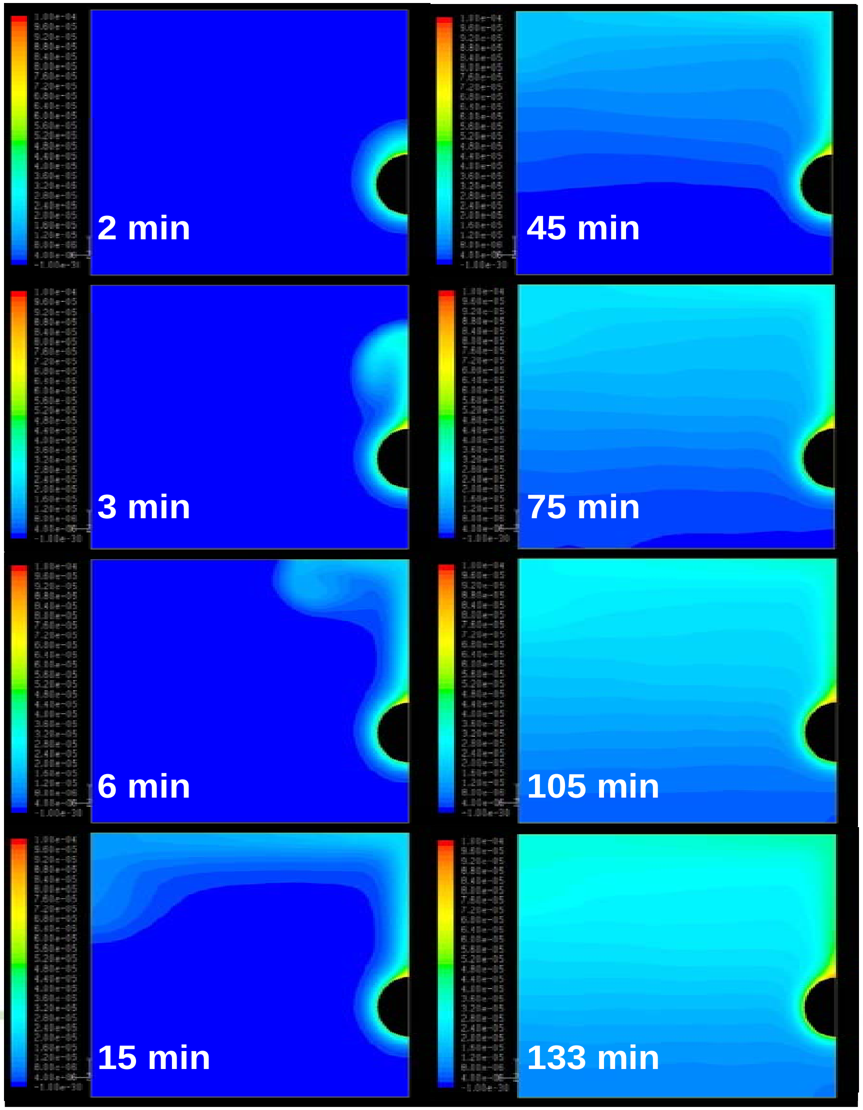


A visible distortion of the symmetrical hydrogen layer on the surface at the top of the tank, at 80 s, indicates the buoyancy starts acting on the hydrogen-air mixture.

Modelling permeation leak (3/7)

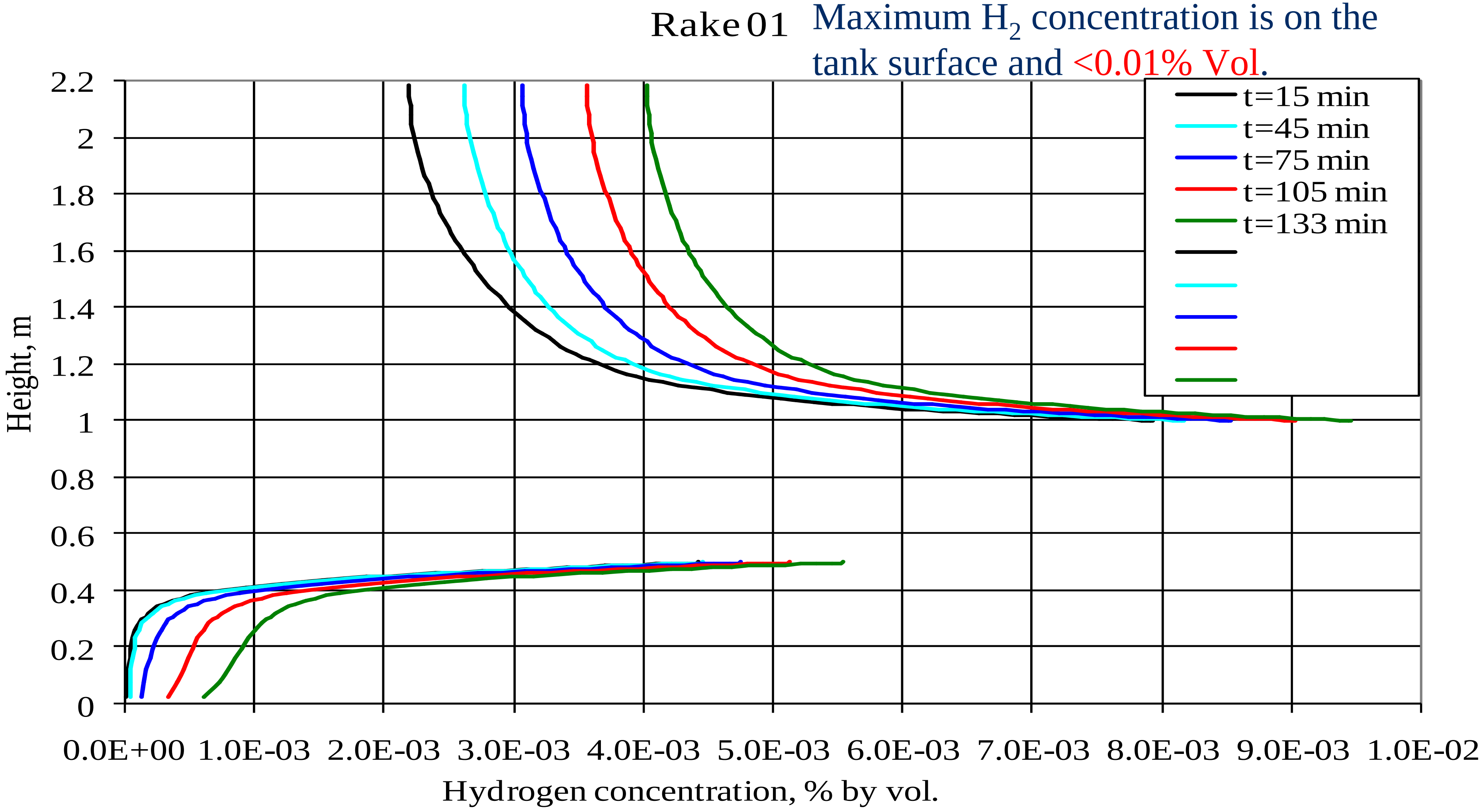
Hydrogen concentration distribution along three rakes





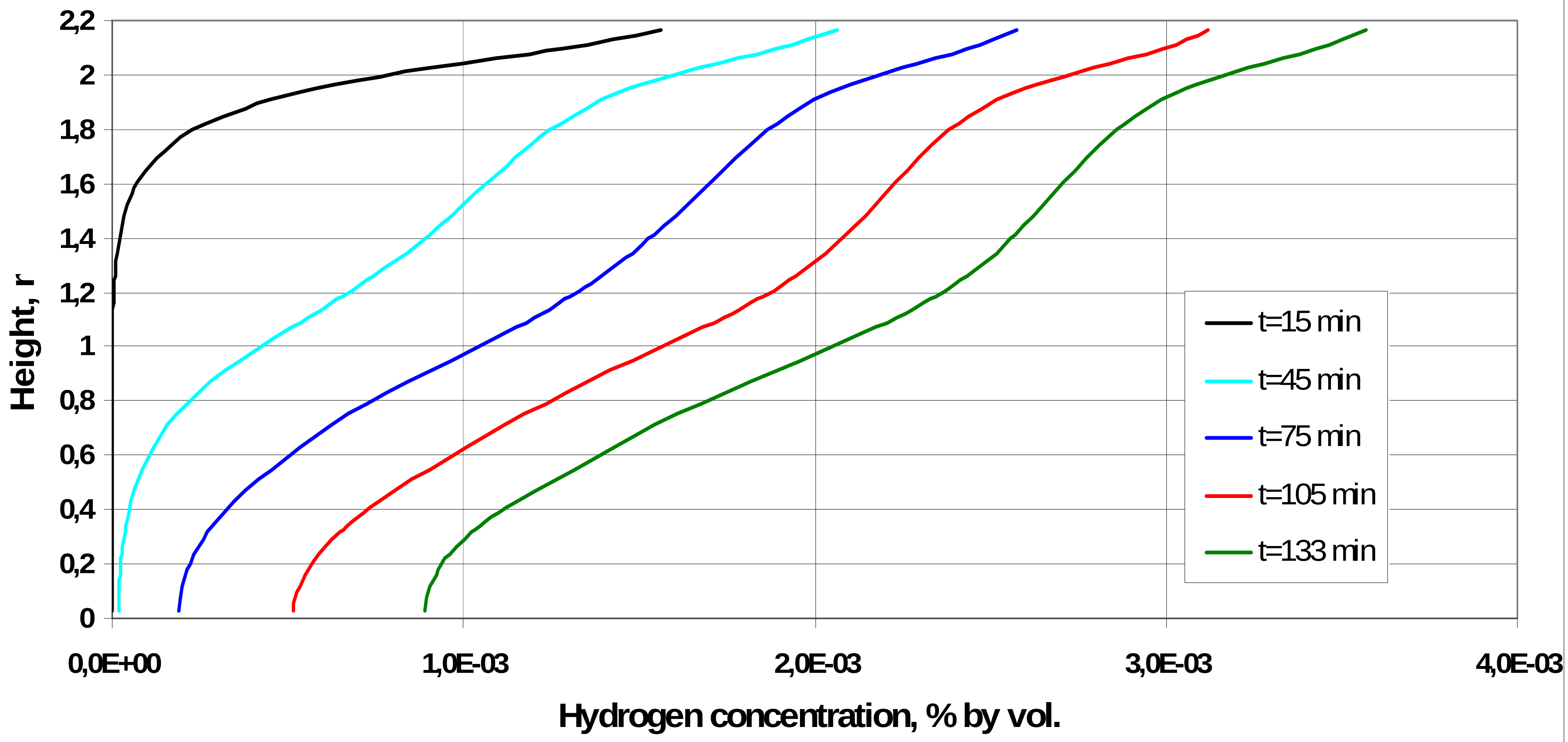
Modelling permeation leak (4/7)

Maximum H_2 concentration is on the tank surface and <0.01% Vol.



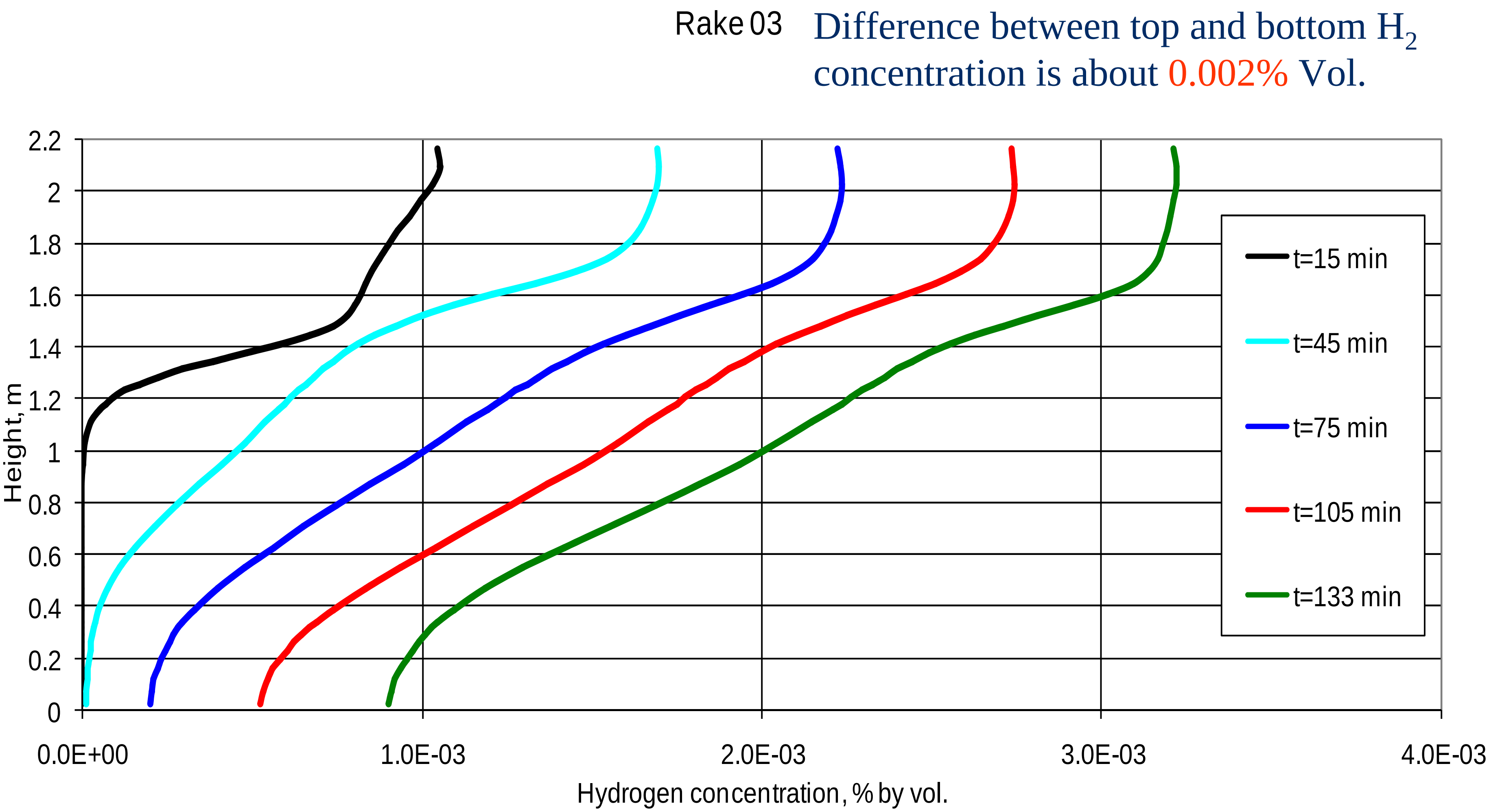
Modelling permeation leak (5/7)

Rake 02 Difference between top and bottom H_2 concentration is about 0.002% Vol.

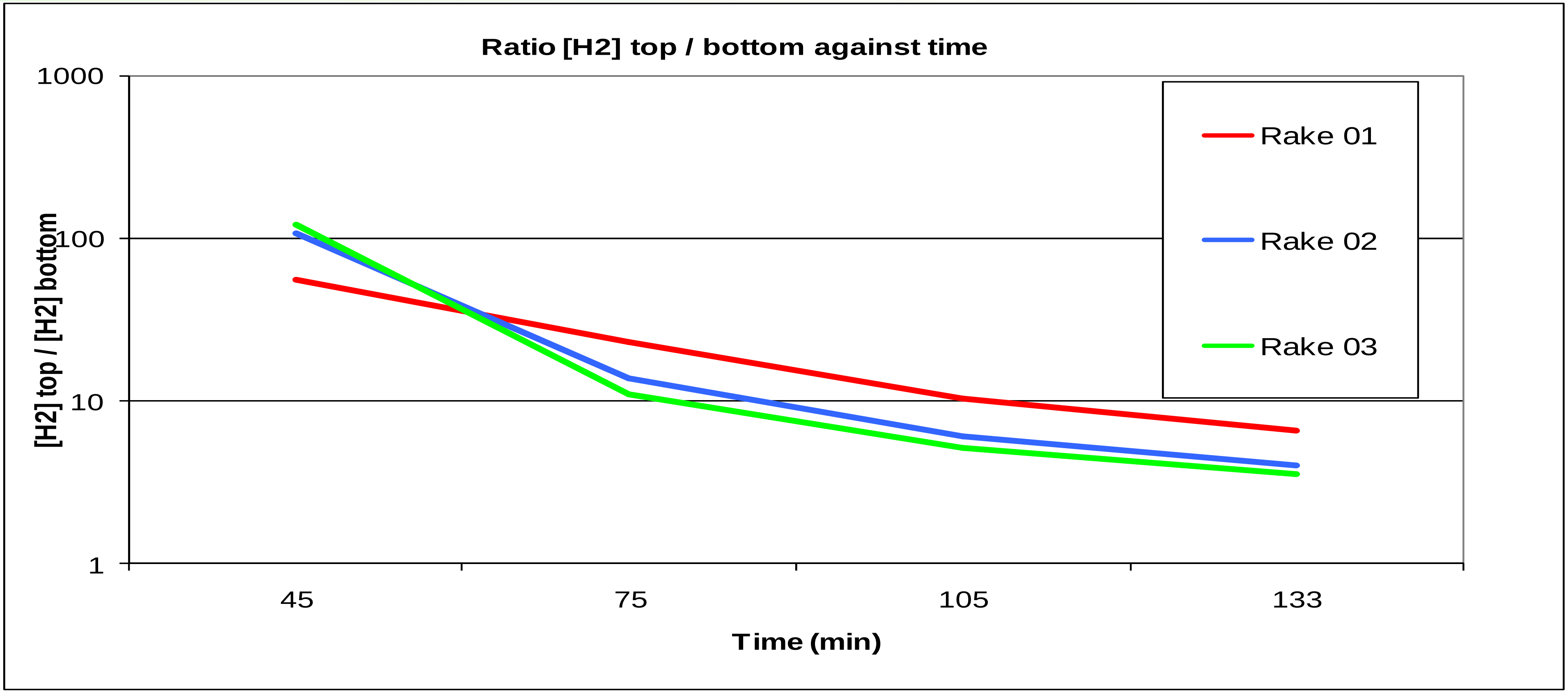


Modelling permeation leak (6/7)

Difference between top and bottom H_2 concentration is about 0.002% Vol.



Modelling permeation leak (7/7)



Indicates the formation of a practically homogenous hydrogen-air mixture within the enclosure over a long period of time. Identical observation made with experiments in CEA garage facility with 1.8 NL/hr leak rate (compared with 0.2 NL/hr in our case)

Conclusion

- The used rate of permeation in our scenario does not seem to represent a safety issue:
  - Low concentration on surface and in garage, and quasi-uniform distribution,
  - Assuming perfectly closed volume hydrogen concentration reaches 4% per Vol. after 240 days,
  - Assuming worst credible minimum air change per hour of 0,03 [4]→ 0.02% per Vol. maintained in the garage [5] and,
  - Assuming the presence of vents designed for natural ventilation to maintain 25% LFL → two vents of 2 cm by 2 cm are sufficient [6].

- Draft of the UN ECE Regulation is over-conservative.

[4] Deliverable 74, InsHyde Project, HySAFE

[5] Lees, F.P., Loss Prevention in the Process Industry, 1996.

[6] Barley et al., 2005, 1st ICHS

Further work

Further work would include

- Investigate safety issues of maximum allowable permeation rates for other RC&S (SAE J2579:01 2009, ISO/TS15869:2009),
- Assess more realistic scenario such as a tank in a whole car in a garage,
- Investigate the influence of atmospheric conditions (temperature, wind, etc.) on the distribution of hydrogen in the garage and on the efficiency of ventilation and,
- Investigate the necessity of implementing mitigation technologies in various types of private or public garages

Thank you for your attention

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