Hydrogen Storage

A Concise Overview

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Dutch Armed Forces

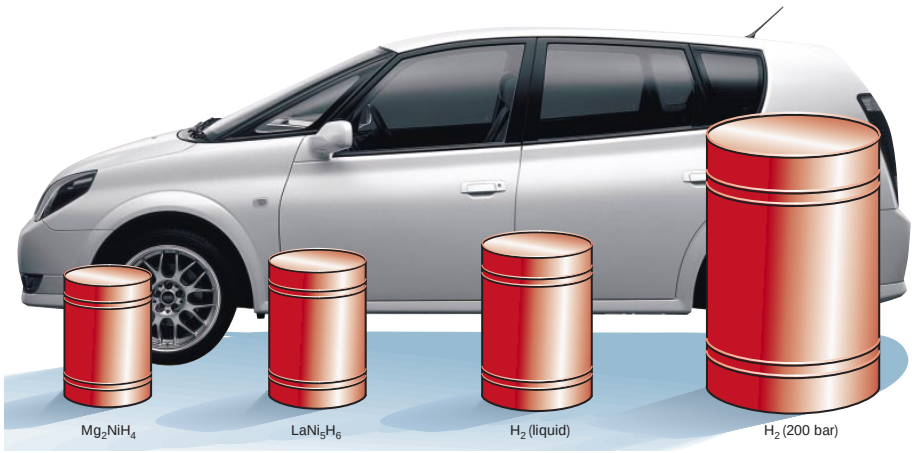


[a] **Knowledge Center for Explosion and Hydrogen Safety**
http://www.explosionsolutions.org/kcehs/
[b] **Royal Dutch Army**
https://en.wikipedia.org/wiki/Royal_Netherlands_Army
[c] **Royal Dutch Navy**
https://en.wikipedia.org/wiki/Royal_Netherlands_Navy

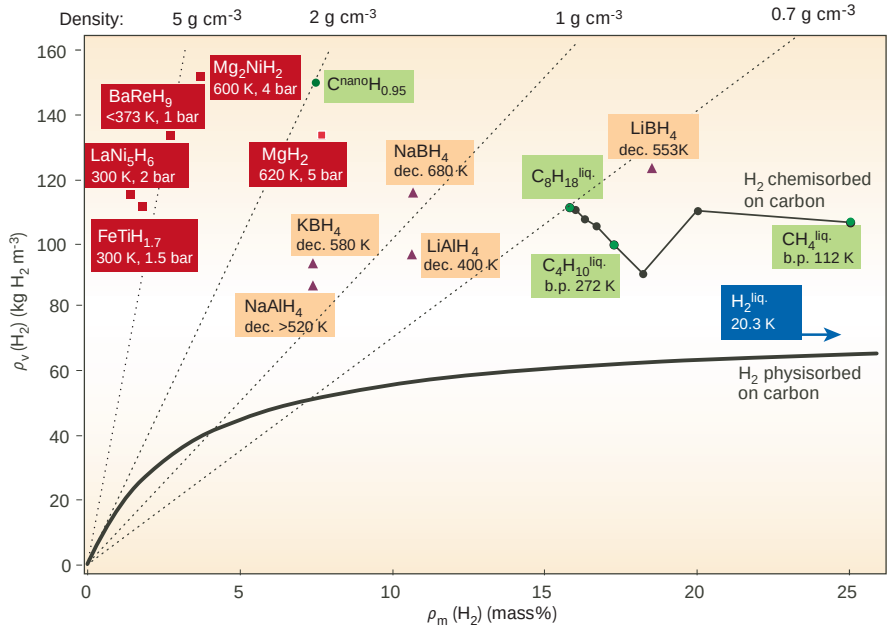
ACKNOWLEDGEMENTS

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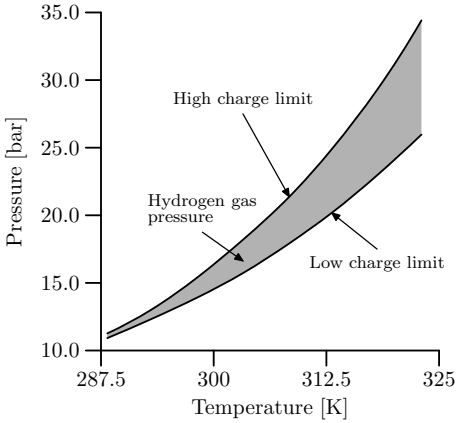
TU Delft
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Required volume for 4 kg hydrogen storage. From [1].



Gaseous Hydrogen	
At 1 bar and 25 °C:	$\rho = 0.09\ \mathrm{kg\,m^{-3}} = 0.00009\ \mathrm{g\,cm^{-3}}$
At 350 bar and 25 °C:	$\rho = 26.1\ \mathrm{kg\,m^{-3}} = 0.0261\ \mathrm{g\,cm^{-3}}$
At 700 bar and 25 °C:	$\rho = 42\ \mathrm{kg\,m^{-3}} = 0.042\ \mathrm{g\,cm^{-3}}$
Liquid Hydrogen	
At 1 bar and -253 °C:	$\rho = 71\ \mathrm{kg\,m^{-3}} = 0.071\ \mathrm{g\,cm^{-3}}$

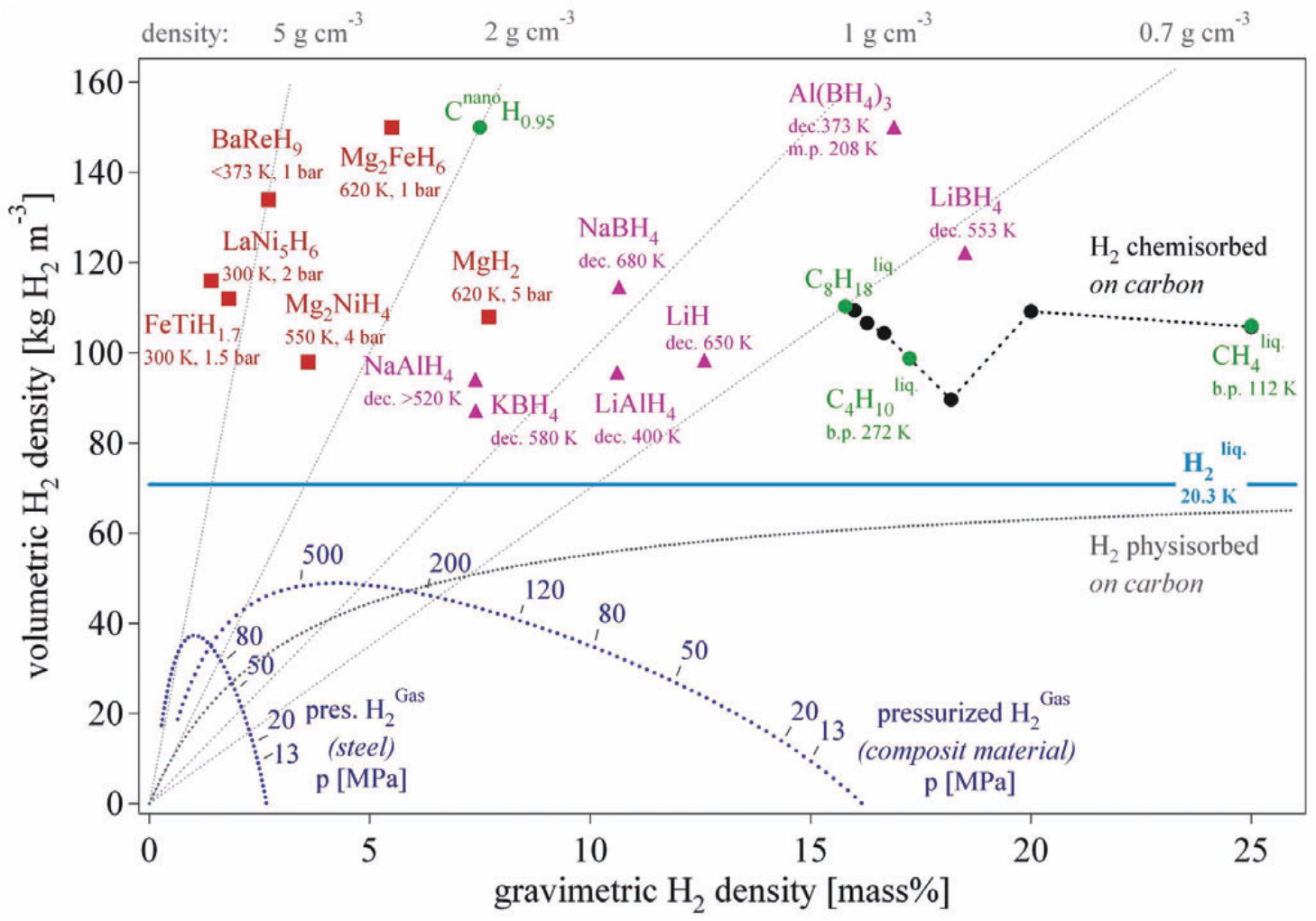


Solid-gas equilibrium curves: $MgH_2\,(s) \longleftrightarrow Mg\,(s) + H_2\,(g)$.



Metal Hydride Canister

[Dr A.E. Dahoe: *Principles of Hydrogen Safety*, Knowledge Center for Explosion and Hydrogen Safety, Dutch Armed Forces]



Volumetric and gravimetric hydrogen density of some selected hydrides. [2].

AMMONIA AS A HYDROGEN CARRIER?

Hydrogenation: $\mathrm{N_2(g) + 3\,H_2(g) \xrightarrow[2\,\Delta_f H_{NH_3}]{} 2\,NH_3(g) \xrightarrow[-2\,\Delta_v H_{NH_3}]{} 2\,NH_3(l)}$

Dehydrogenation: $\mathrm{2\,NH_3(g) \xrightarrow[-2\,\Delta_f H_{NH_3}]{} N_2(g) + 3\,H_2(g)}$

$\Delta_f H_{NH_3}$ = -46.1 kJ mol^{-1} = -2.71 MJ kg^{-1}
$\Delta_v H_{NH_3}$ = 23.5 kJ mol^{-1} = 1.38 MJ kg^{-1}
$\Delta_c H_{NH_3}$ = -382.8 kJ mol^{-1} = -22.477 MJ kg^{-1}

1 kg H_2 requires 5.7 kg NH_3

WATER AS A HYDROGEN CARRIER?

Hydrogenation: $\mathrm{O_2(g) + 2\,H_2(g) \xrightarrow[2\,\Delta_f H_{H_2O}]{} 2\,H_2O\,(l)}$

Dehydrogenation: $\mathrm{2\,H_2O\,(l) \xrightarrow[-2\,\Delta_f H_{H_2O}]{} O_2(g) + 2\,H_2(g)}$

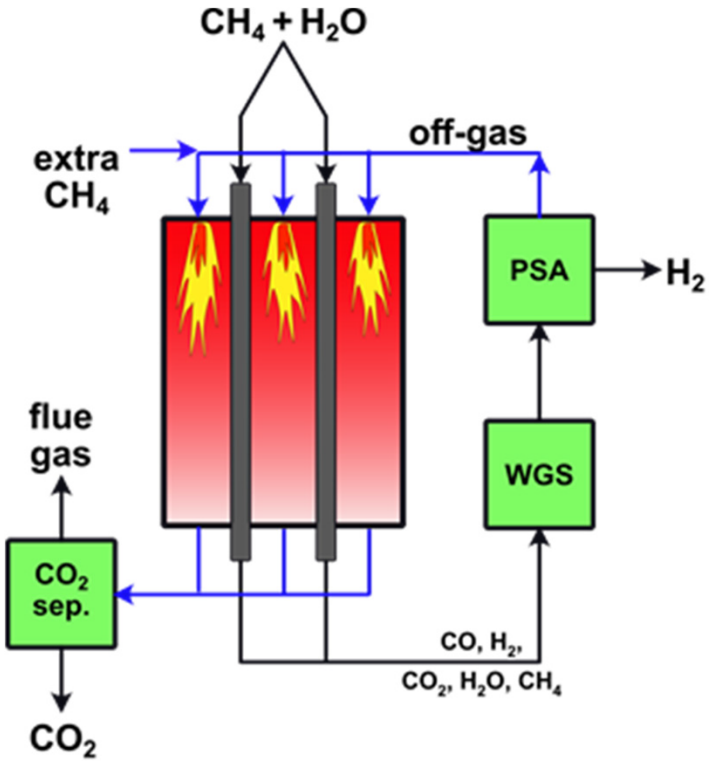
$\Delta_f H_{H_2O}$ = -286 kJ mol^{-1} = -15.9 MJ kg^{-1}
$\Delta_v H_{H_2O}$ = 40.63 kJ mol^{-1} = 2.26 MJ kg^{-1}

1 kg H_2 requires 9 kg H_2O

Chemical formula	Hydrogen (wt%)
CH_4	25
NH_3BH_3	19.6
$LiBH_4$	18.3
$(CH_3)_4NBH_4$	18
NH_3	17.7
$Al(BH_4)_3$	16.8
$Mg(BH_4)_2$	14.8
LiH	12.6
CH_3OH	12.5
H_2O	11.2
$LiAlH_4$	10.6
$NaBH_4$	10.6
AlH_3	10.0
MgH_2	7.6
$NaAlH_4$	7.4

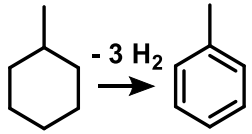
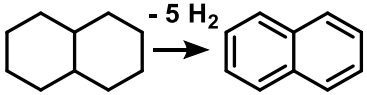
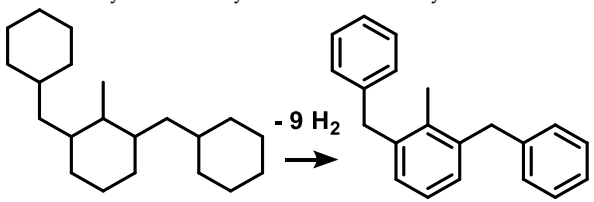
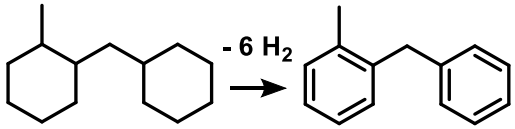
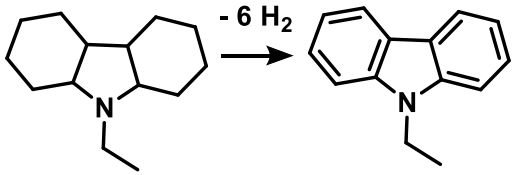
HYDROGEN FROM METHANE

$CH_4 + H_2O \rightleftharpoons CO + 3\,H_2$	(steam reforming)
$CO + H_2O \rightleftharpoons CO_2 + H_2$	(water-gas shift reaction)
$CH_4 + 2\,H_2O \rightleftharpoons CO_2 + 4\,H_2$	(overall reaction)



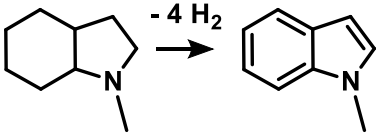
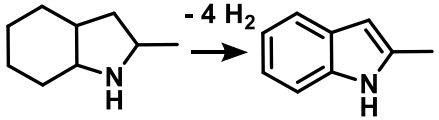
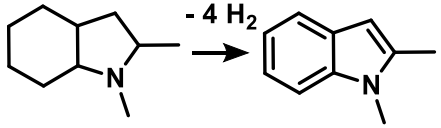
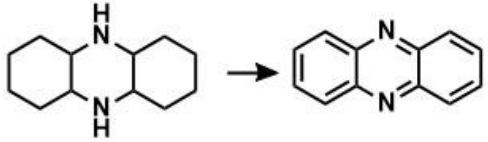
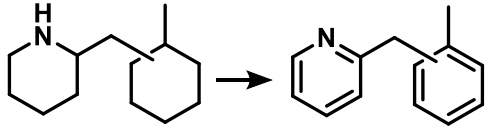
From: [3].

Liquid-Organic Hydrogen Carriers (LOHC)

LOHC System	H_2 Capacity (wt%)	Enthalpy (kJ/mol H_2)	Melting Point (°C)		Boiling Point (°C)		Flash Point (°C)	
			H_2-Rich	H_2-Lean	H_2-Rich	H_2-Lean	H_2-Rich	H_2-Lean
Methylcyclohexane → toluene 	6.2	68.3	−126	−95	101	111	−3	6
Decalin → Naphthalene 	7.3	63.9	−37	79	189	218	57	80
Perhydro-dibenzyltoluene → dibenzyl toluene 	6.2	65.4	NA	−34	NA	398	NA	190
Perhydro-benzyl toluene → benzyl toluene 	6.2	63.5	NA	−30	270	280	NA	NA
Dodecahydro-N-ethyl carbazole → N-ethyl carbazole 	5.8	50.6	−85	70	NA	166	NA	186

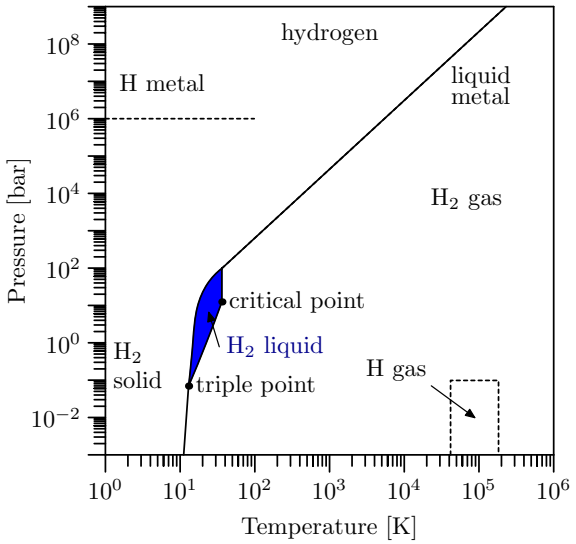
From: [4].

Liquid-Organic Hydrogen Carriers (LOHC)

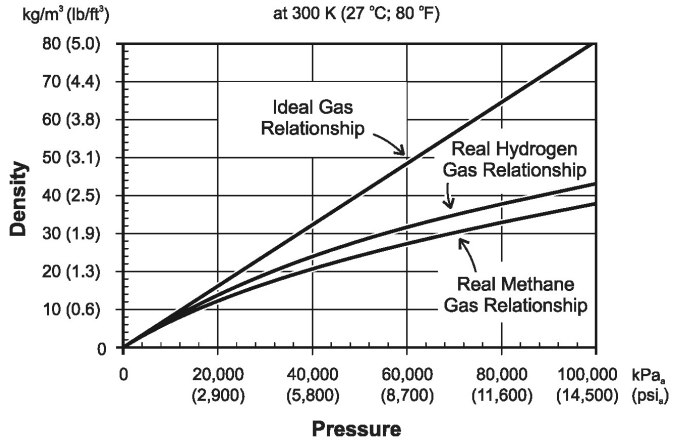
LOHC System	H_2 Capacity (wt%)	Enthalpy (kJ/mol H_2)	Melting Point (°C)		Boiling Point (°C)		Flash Point (°C)	
			H_2-Rich	H_2-Lean	H_2-Rich	H_2-Lean	H_2-Rich	H_2-Lean
1-methylperhydro indole → 1-methyl indole 	5.8	51.9	NA	95	180	238	60	110
2-methylperhydro indole → 2-methyl indole 	5.8	NA	NA	60	178	272	58	141
1,2-perhydrodimethyl indole → 1,2-dimethyl indole 	5.23	NA	<−15	55	≥260.5	260.5	≥230	≥230
Perhydro-phenazine → phenazine 	7.2	NA	NA	174–177	NA	357.2	NA	160.3
Perhydro-2-(n-methylbenzyl pyridine) → 2-(n-methylbenzyl pyridine) 	6.15	67.3	−19.3	−50.1	293	291	NA	NA

From: [4].

AGGREGATION STATES, PHASE DIAGRAM AND EQUATIONS OF STATE



Phase diagram [5].



H_2 and CH_4 density as a function of pressure [6].

EQUATIONS OF STATE

Idea Gas Law: $P(V/n) = \mathrm{R}T$

Nobel-Abel: $P\left[(V/n) - b\right] = \mathrm{R}T$

Van der Waals: $\left[P + \dfrac{a}{(V/n)^2}\right]\left[(V/n) - b\right] = \mathrm{R}T$

$$a = \frac{27\mathrm{R}^2T_c^2}{64P_c} \quad \text{and} \quad b = \frac{\mathrm{R}T_c}{8P_c}$$

Beattie-Bridgeman:

$$\left[P + \frac{A}{(V/n)^2}\right]\frac{(V/n)^3T^3}{\left[(V/n)T^3 - c\right]\left[(V/n) + B\right]} = \mathrm{R}T$$

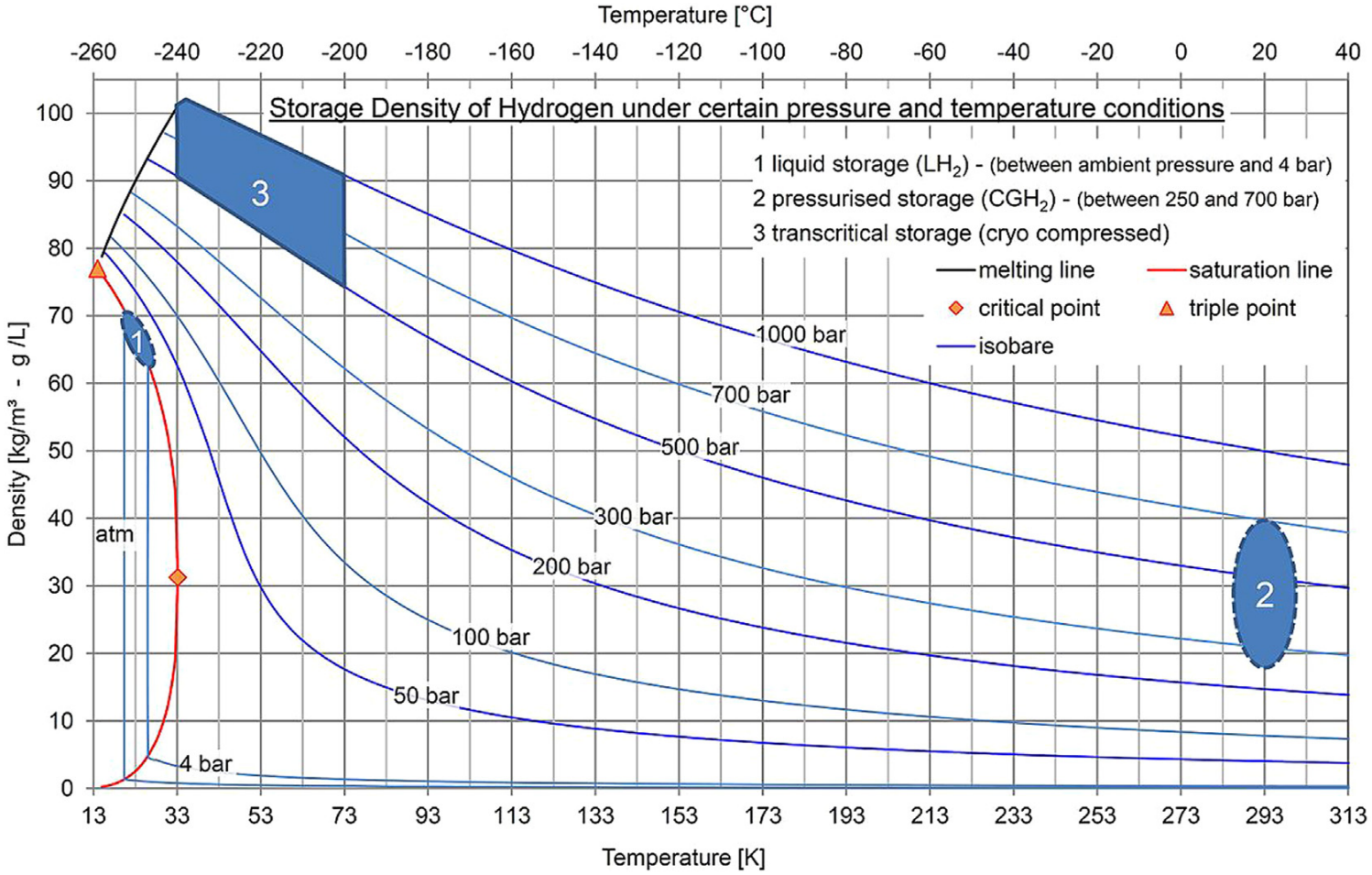
$$A = A_0\left[1 - \frac{a}{(V/n)}\right] \quad \text{and} \quad B = B_0\left[1 - \frac{b}{(V/n)}\right]$$

Constants a, b, c, A_0, and B_0 assume different values for each substance.

Benedict-Webb-Rubin:

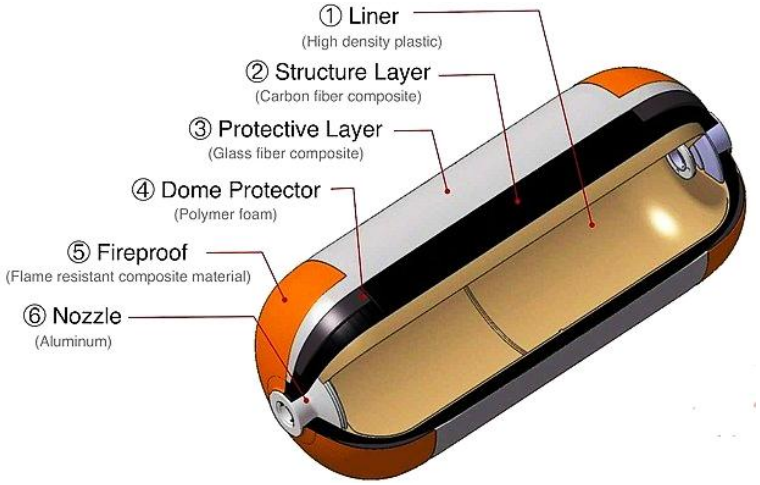
$$P - \left[B_0\mathrm{R}T - A_0 - \frac{C_0}{T^2}\right]\frac{1}{(V/n)} - \frac{b\mathrm{R}T}{(V/n)^2} - \frac{a\alpha}{(V/n)^5} - \frac{c}{(V/n)^2T^2}\left[1 + \frac{\gamma}{(V/n)^2}\right]\exp\left(-\frac{\gamma}{(V/n)^2}\right) = \mathrm{R}T$$

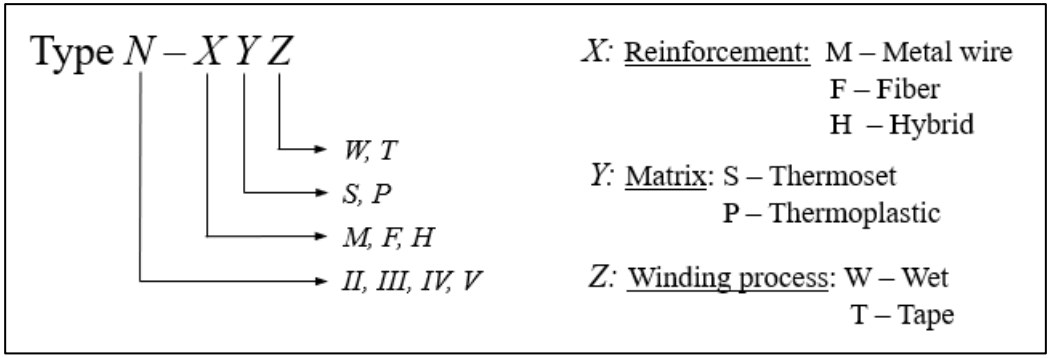
Substance specific constants a, b, c, A_0, B_0, C_0, α, and γ.



Density of hydrogen as a function of temperature at different pressure levels. [7].

GASEOUS HYDROGEN – VEHICLE STORAGE TANKS



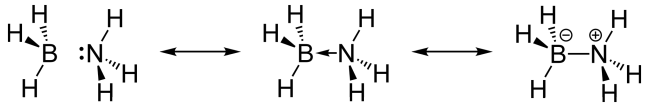


[95]	Sub-Type Classification	Liner	Wrap Extent	Winding Method	Resin Type	Fiber Type	Low Cost	Recyclability	Light Weight
	Type II—FSW [1]	Metal	Cylinder	Wet	TS	CF	+++	++++	+
	Type II—MSW	Metal	Cylinder	Wet	TS	SW	+++++	+++++	+
	Type III—FSW [1]	Metal	Full	Wet	TS	CF	+++	+++	++
	Type III—MSW	Metal	Full	Wet	TS	SW	++++	++++	+
	Type IV—FSW [1]	Plastic	Full	Wet	TS	CF	+	++	+++++
	Type IV—FST	Plastic	Full	Tape	TS	CF	+	++	+++++
	Type IV—FPT	Plastic	Full	Tape	TP	CF	+	+++	+++++
	Type IV—FPW	Plastic	Full	Wet	TP	CF	+	+++	+++++
	Type IV—MSW	Plastic	Full	Wet	TS	SW	++++	++++	+++
	Type V [2]	Liner-less	Full	Tape	TS/TP	CF	++	++	+++++

TS: thermoset; TP: thermoplastic; CF: carbon fiber; SW: steel wire. [1] Most frequent configuration. [2] Early stage of development. "+", "++", "+++", "++++" and "+++++" denotes a comparative score, from very low to very high.

From Ref. [8].

NH_3BH_3 (AMMONIA BORANE)



DEHYDROGENATION

Thermolytic:

$$H_3BNH_3\,(s) \xrightarrow{90\,^\circ C} H_2BNH_2\,(s) + H_2\,(g)$$

$$\xi\,H_2BNH_2\,(s) \longrightarrow [H_2BNH_2]_\xi\,(s)$$

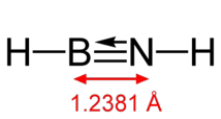
$$[H_2BNH_2]_\xi\,(s) \xrightarrow{155\,^\circ C} [HBNH]_\xi\,(s) + \xi\,H_2\,(g)$$

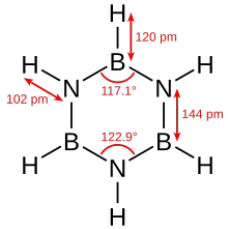
$$[HBNH]_\xi\,(s) \xrightarrow{>500\,^\circ C} \xi\,BN\,(s) + \xi\,H_2\,(g)$$

Catalytic:

$$3\,H_3BNH_3\,(\text{non-aq}) \xrightarrow[\text{tetrahydrofuran}]{\text{catalyst}} B_3N_3H_6\,(\text{non-aq}) + 6\,H_2\,(g)$$

NH_3BH_3 = ammonia borane.
H_2BNH_2 = aminoborane.
$[H_2BNH_2]_\xi$ = polyaminoborane.
HBNH = iminoborane (lower-left).
$[HBNH]_\xi$ = polyiminoborane.
$B_3N_3H_6$ = borazine (lower-right).
BN = boron nitride.



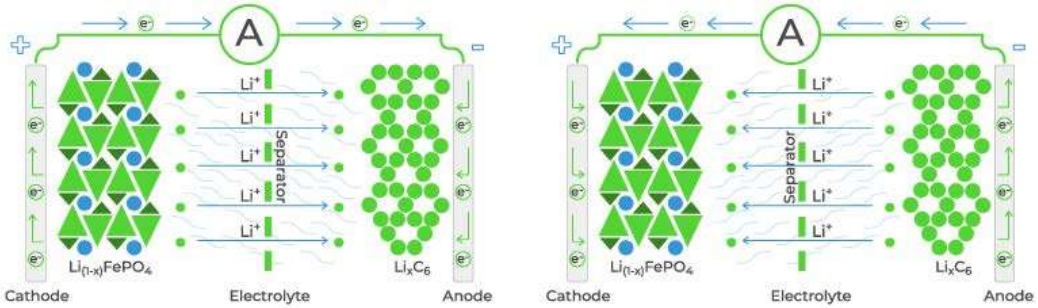




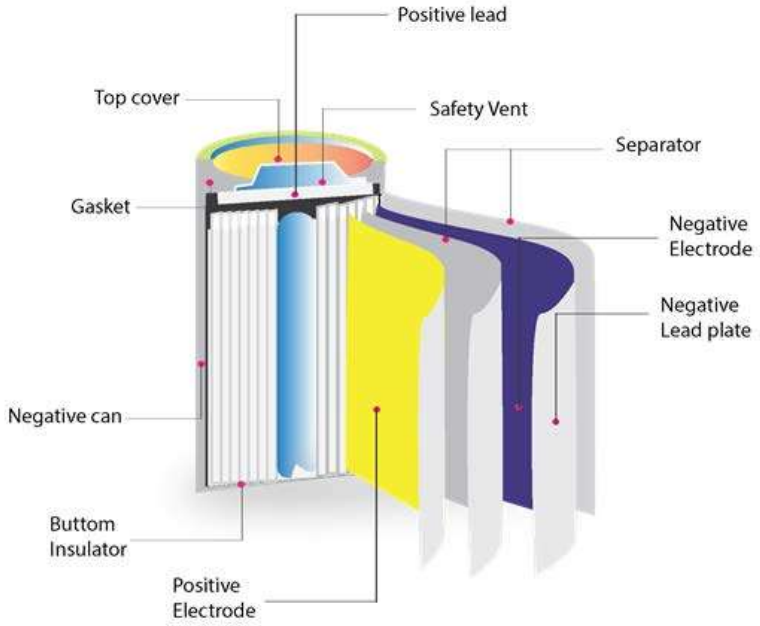


From [9].

MILITARY GRADE LITHIUM ION BATTERIES



CHARGING: $Fe^{2+} \longrightarrow Fe^{3+}$ **DISCHARGING:** $Fe^{3+} \longrightarrow Fe^{2+}$



Electrolyte: $LiPF_6$ and $H{-}[CH_3F]_n{-}H$

Solid Electrolyte Interface: (cathode) $Li_{(1-\xi)}FePO_4 \longleftrightarrow Li_\xi C_6$ (anode)

Charging progress variable, ξ:
[$\xi = 0$: fully discharged; $\xi = 1$: fully charged]

Anode: $C_6 + \xi Li^+ + \xi e^- \longrightarrow Li_\xi C_6$

Cathode: $LiFePO_4 - \xi Li^+ - \xi e^- \longrightarrow Li_{(1-\xi)}FePO_4$

Overall: $C_6 + LiFePO_4 \longrightarrow Li_\xi C_6 + Li_{(1-\xi)}FePO_4$

$\xi = 0$: $C_6 + LiFePO_4$ (fully discharged)

$\xi = 1$: $LiC_6 + FePO_4$ (fully charged)

Discharge progress variable, $\eta = 1 - \xi$:
[$\eta = 0$: fully charged; $\eta = 1$: fully discharged]

Anode: $Li_{1-\eta}C_6 - (1\text{-}\eta)\, Li^+ - (1\text{-}\eta)\, e^- \longrightarrow C_6$

Cathode: $Li_\eta FePO_4 + (1-\eta)\, Li^+ + (1\text{-}\eta)\, e^- \longrightarrow LiFePO_4$

Overall: $Li_{(1-\eta)}C_6 + Li_\eta FePO_4 \longrightarrow C_6 + LiFePO_4$

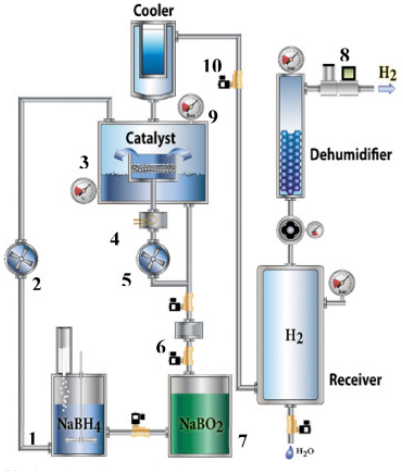
$\eta = 0$: $LiC_6 + FePO_4$ (fully charged)

$\eta = 1$: $C_6 + LiFePO_4$ (fully discharged)

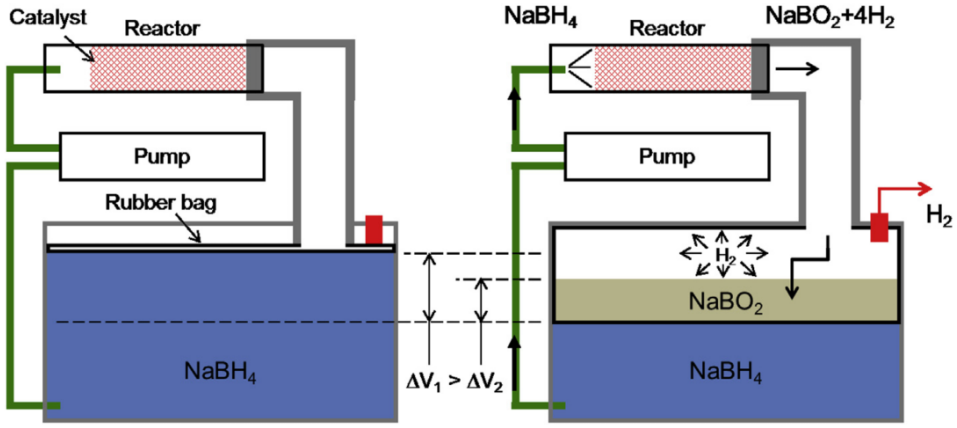
[Dr A.E. Dahoe: *Principles of Hydrogen Safety*, Knowledge Center for Explosion and Hydrogen Safety, Dutch Armed Forces]

$NaBH_4$ (SODIUM BOROHYDRIDE, SODIUM TETRAHYDRIDOBORATE)

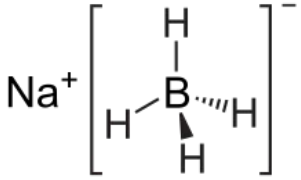




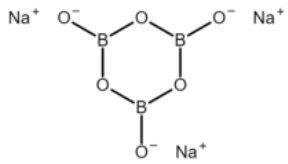
Laboratory prototype [10].



Operational concept [11].



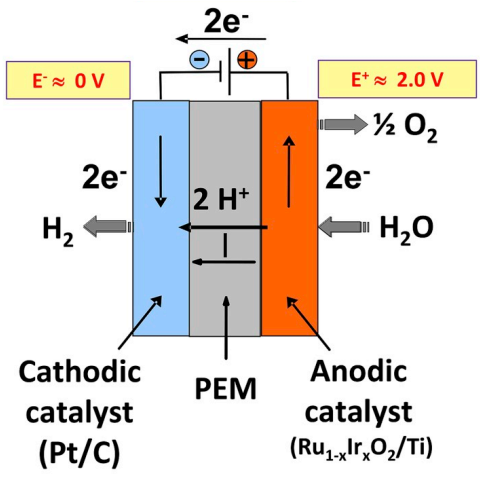
Sodium borohydride



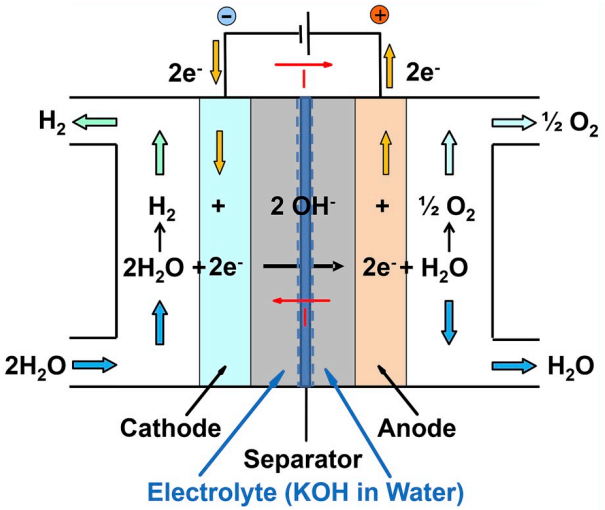
Sodium metaborate

DEHYDROGENATION	
Hydrolytic:	**Catalytic:**
$NaBH_4 + 2\,H_2O \longrightarrow NaBO_2 + 4\,H_2$	$NaBH_4 + 2\,H_2O \xrightarrow[HCl,\,H_2SO_4]{\text{acid accelerator}} NaBO_2 + 4\,H_2$
$NaBH_4 + (2+\xi)\,H_2O \longrightarrow NaBO_2 + 4\,H_2 + \xi H_2O$	$NaBH_4 + 2\,H_2O \xrightarrow[\text{Co-Ru-Ni}]{\text{catalyst}} NaBO_2 + 4\,H_2$

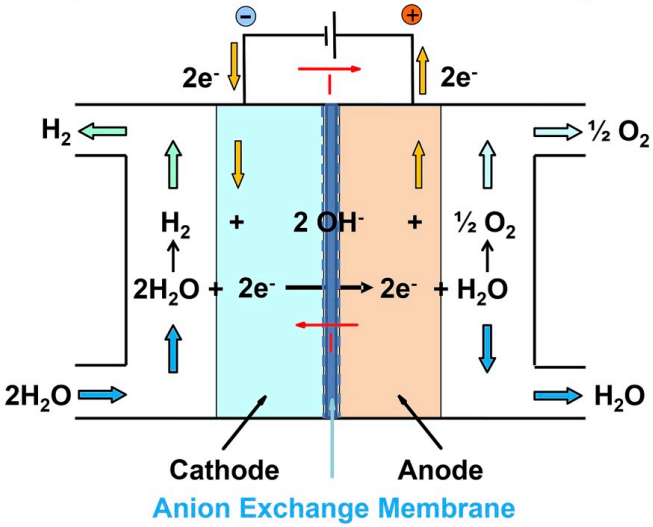
ELECTROLYZERS



Proton Exchange Membrane Electrolyser



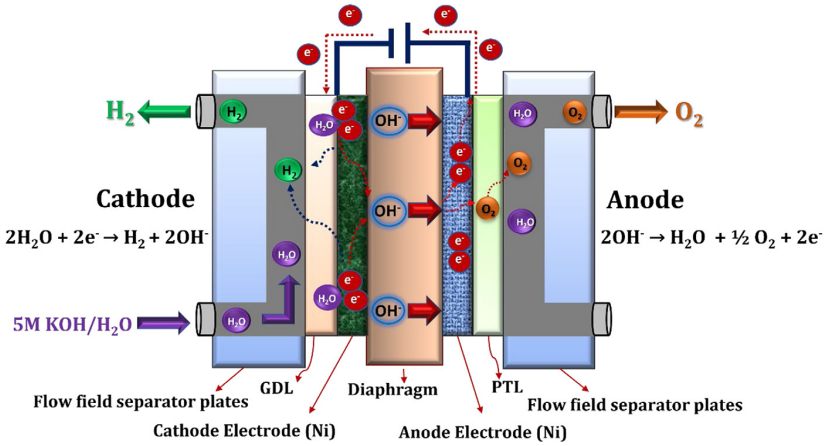
Alkaline Electrolyser



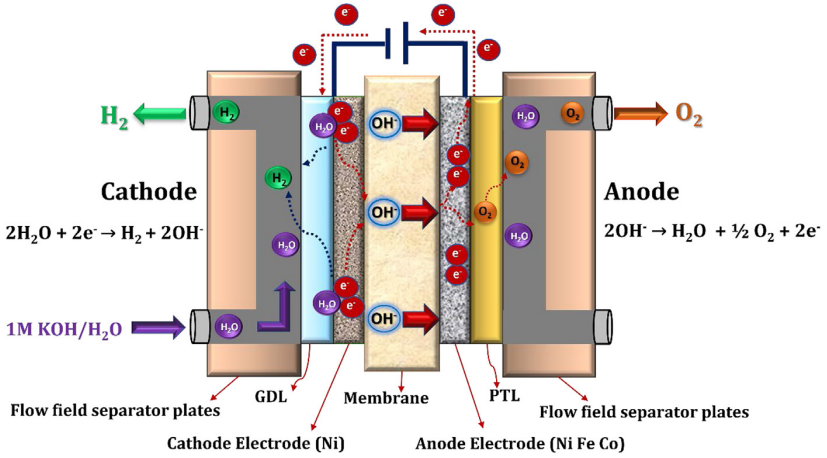
Anion Exchange Membrane Electrolyzer

Source: Ref. [12].

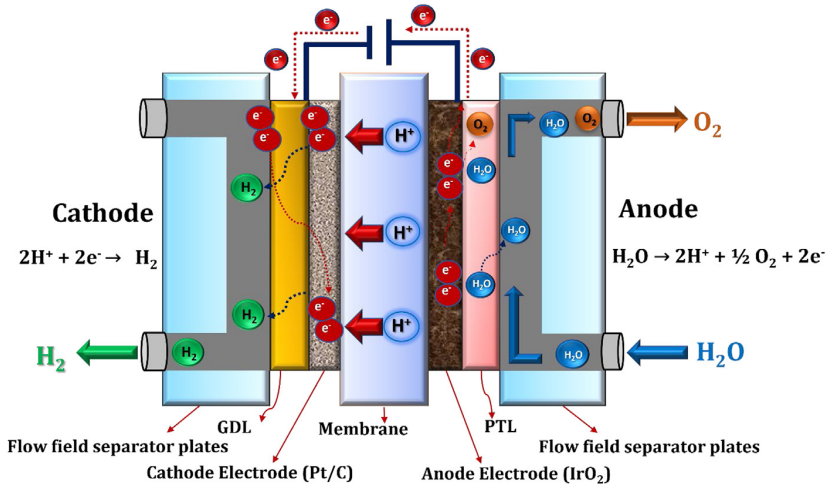
[Dr A.E. Dahoe: *Principles of Hydrogen Safety*, Knowledge Center for Explosion and Hydrogen Safety, Dutch Armed Forces]



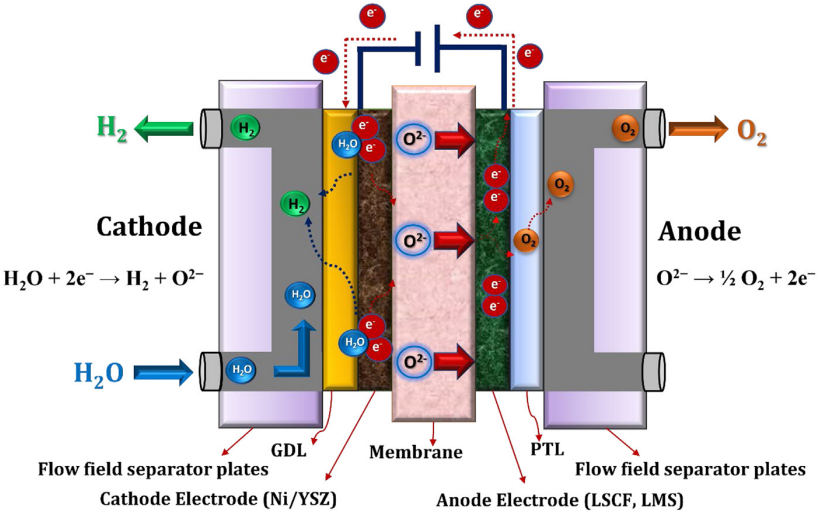
Alkaline water electrolysis [13].



Anion Exchange Membrane (AEM) water electrolysis [13].



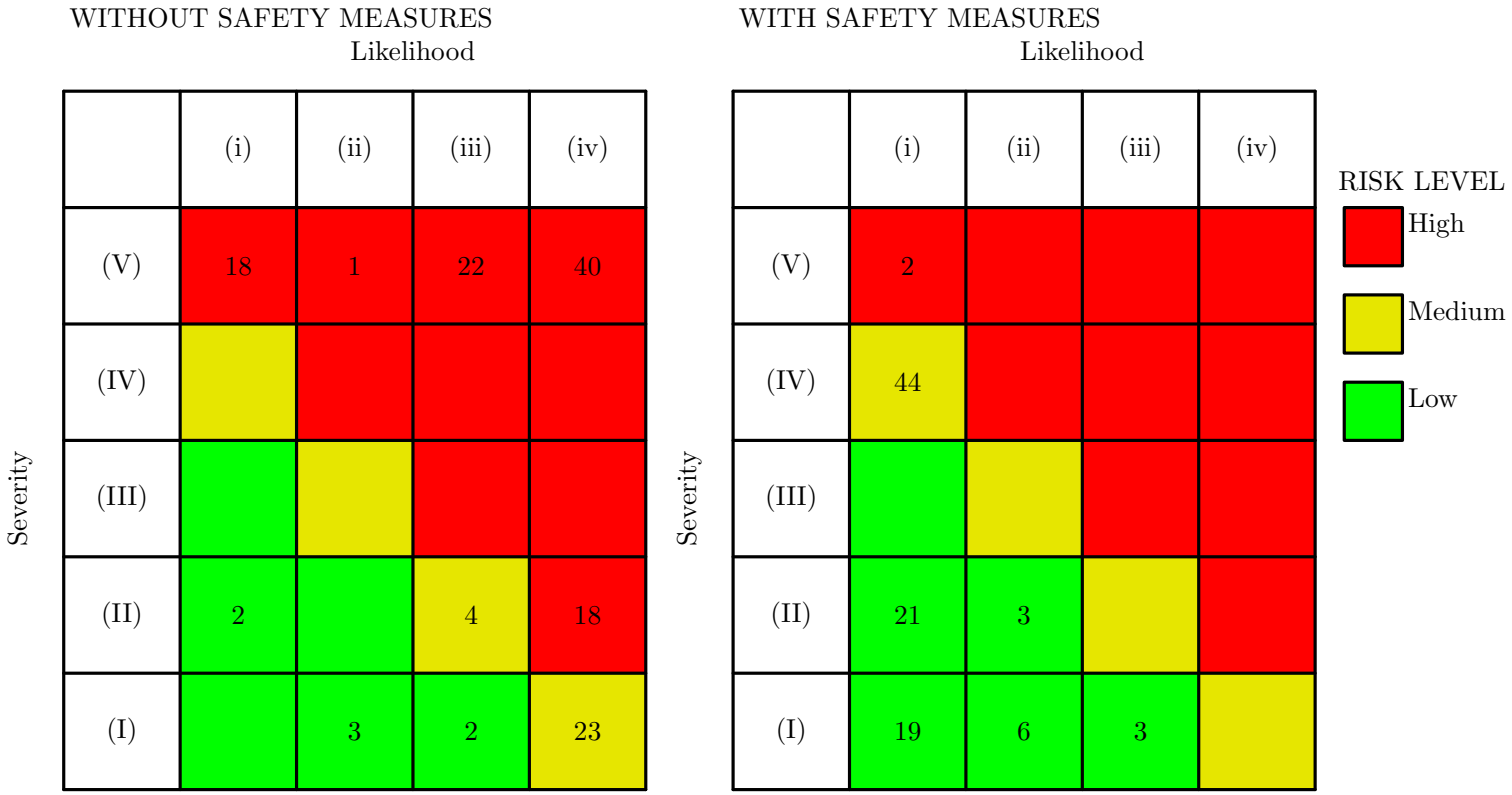
Proton Exchange Membrane (PEM) water electrolysis [13].



Solid oxide water electrolysis [13].

GDL = Gas Diffusion Layer. PTL = Porous Transport Layer.
LMS = Lanthanum Manganite Strontium. YSZ = Yttrium Stabilized Zirconia.
LSCF = Lanthanum Strontium Cobalt Ferrite.

ELECTROLYZERS – RISK ANALYSIS (HAZID, RISK MATRIX)



Modification of the Risk Matrix according to Ref. [14].

ELECTROLYZERS – RISK ANALYSIS (HAZID, RISK MATRIX)

Severity and likelihood criteria according to Ref. [14].

Severity ranking criteria		Blast pressure (kPa)	Jet flame length (m)
V	Severe damage to building and facilities outside the site	> 34	> 6
	Fatalities outside the site	> 100	> 6
IV	Significant damage to building and facilities outside the site	6.9 – 34	–
	Fatalities inside the site	> 100	2 – 6
III	Moderate damage to building and facilities outside the site	3.4 – 6.9	–
	Injuries outside/inside the site	16 – 100	–
II	Minor damage to building and facilities outside the site	1 – 3.4	1 – 2
	Minor injuries outside/inside the site	2 – 16	0.3 – 1
I	Negligible damage to building and facilities outside the site	< 1	< 6
	Negligible injuries outside/inside the site	< 2	< 0.3

Likelihood criteria	
i	Once per 1000 years
ii	Once per 100 years
iii	Once per 10 years
iv	Once per year

FMEA failure mode
Hydrogen embrittlement
Hydrogen assisted cracking
Corrosion
Stress corrosion cracking
Fatigue
Loosening of threaded joints
Weld defect
Sealing deterioration
Seat/gland leakage
Polymer electrolyte damage

[Dr A.E. Dahoe: *Principles of Hydrogen Safety*, Knowledge Center for Explosion and Hydrogen Safety, Dutch Armed Forces]

GASEOUS HYDROGEN STORAGE – EXPLOSION HAZARDS

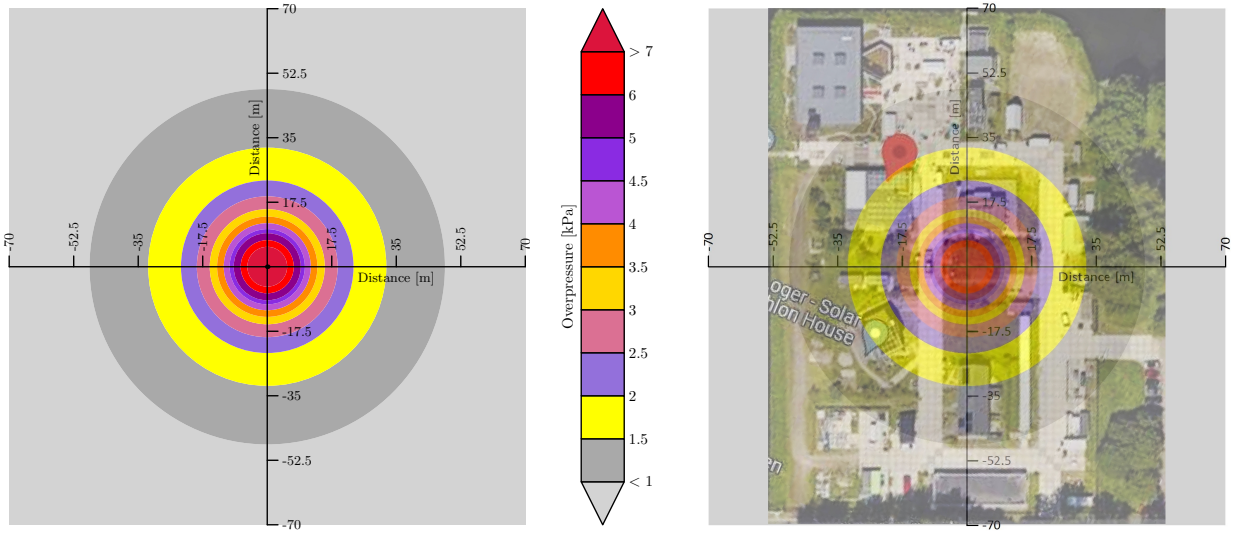


Kjorbo, Norway
Monday 10 June 2019 at 17:30 h
Hydrogen Explosion (3.0 – 3.5 kg H_2)
Glass damage at 60 – 70 m distance
Office at 60 m distance
Houses at > 225 m distance

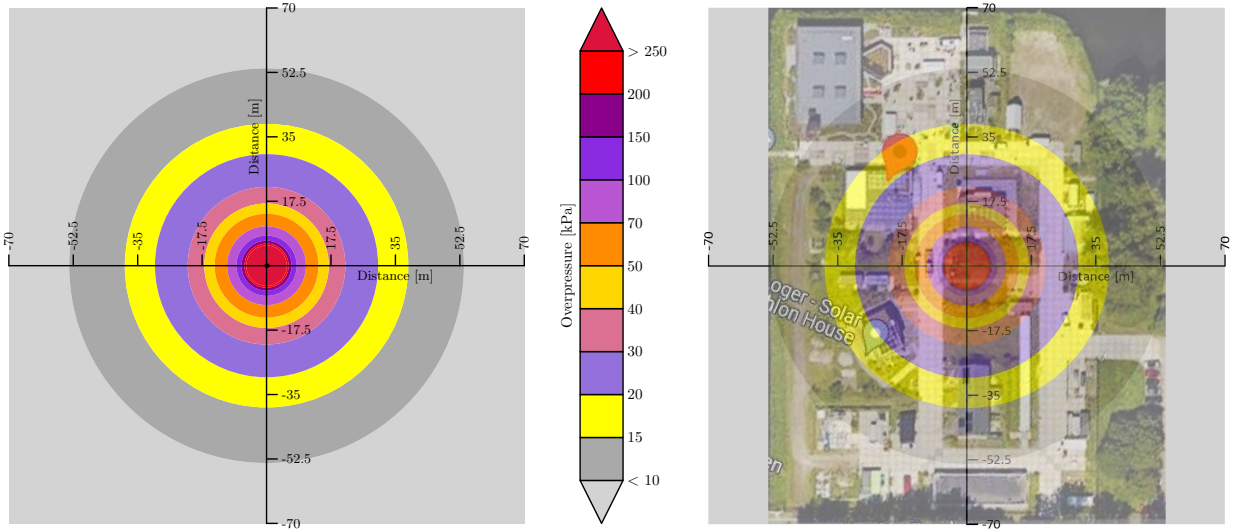


DELFT UNIVERSITY OF TECHNOLOGY – GREEN VILLAGE

Explosion involving 3.5 kg hydrogen Blast calculation according to Ref. [15].



Deflagration



Detonation

PRESSURE EFFECTS ON PEOPLE, STRUCTURES AND EQUIPMENT

Entry	Overpressure (kPa)	Damage/injury
1	190	Occupants of unstrengthened building will be killed by direct action of blast, by being struck by building debris, or by impact against hard surfaces [16].
2	69–76	Occupants of unstrengthened building are expected to suffer severe injuries or death from direct blast, building collapse, or translation [16].
3	70	Probable total destruction of buildings; heavy machine tools moved and badly damaged [17].
4	60	Loaded train boxcars completely demolished [17].
5	55	Occupants of unstrengthened building will be killed or incur serious injuries by blast to eardrums and lungs, by being blown down or struck by fragments and building debris [16].
6	50–55	Unreinforced brick panels, 25–35 cm thick, fail by shearing or flexure [17].
7	50	Loaded tank cars/train wagons overturned; brick walls, 20–30 cm thick, collapse [17].
8	40–45	Collapse of pipe bridge [17].
9	35–40	Displacement of pipe bridge, failure of piping [17].
10	20–40	Large trees fall down [17].
11	35	Breakage of wooden telephone poles; most buildings destroyed, except for concrete reinforced shear wall buildings; *platting* of cars and trucks pressed inwards [17].
12	30–35	Near-complete destruction of houses [17].
13	30	Cladding of light industrial buildings ruptured. Panelling torn-off [17].
14	20–28	Frameless, self-framing steel panel building demolished; rupture of oil storage tanks [17].
15	24	Personnel injuries of a serious nature or possible death are likely from fragments, debris, firebrands or other objects. There is a 10% chance of eardrum rupture [16].
16	15–20	Unreinforced concrete or cinderblock walls shattered [17].
17	20	Heavy machines in industrial buildings suffer little damage; steel frame building distorted and pulled away from foundations [17].
18	18	Lower limit of serious structural damage; 50% destruction of brickwork of houses [17].

Table 1: Continued.

19	16	Personnel may suffer temporary hearing loss or injury from secondary blast effects such as building debris and the tertiary effect of displacement. No fatalities or serious injuries from direct blast are expected [16].
20	15	Partial collapse of walls and roofs of houses [17].
21	7–15	Corrugated asbestos shattered. Corrugated steel or aluminum panels fastenings fail, followed by buckling; wood panel fastenings fail, panels blown in [17].
22	10	Steel frame of clad buildings slightly distorted [17].
23	5.9–8.3	Personnel are provided a high degree of protection from death or serious injury with injuries that do occur being principally caused by glass breakage and building debris [1].
24	8	Partial demolition of houses, made uninhabitable [17].
25	7	Collapse of roof of a tank [17].
26	3.5–7	Windows usually shattered; occasional damage to window frames [17].
27	5	Minor damage to house structures [17].
28	3	Limited minor structural damage [17].
29	2	Probability of 0.95 of no serious damage beyond this value; some damage to house ceilings; 50% of window glass broken [17].
30	1	Typical threshold for glass breakage [17].
31	0.7	Breakage of small windows under strain [17].
32	0.3	Loud noise similar to sonic boom; occasional glass failure [17].
33	0.2	No structural damage; occasional breaking of large window panes already under strain [17].
34	0.15	Annoying noise [17].

HYDROGEN PURITY FOR FUEL CELLS

	Hydrogen Availability			Fuel Cell Requirements	
Class	**3.0**	**5.0**	**6.0**	**ISO 14687-2 (2012)**	**Manufacturer**
H_2 mole fraction	≥ 99.9 (%)	≥ 99.999 (%)	≥ 99.9999 (%)	≥ 99.997	
N_2 + Ar (ppm)		≤ 3	≤ 0.5	≤ 100	≤ 100
H_2O (ppm)	≤30	≤ 5	≤ 0.5	≤ 5	≤ 5
O_2 (ppm)		≤ 2	≤ 0.5	≤ 5	≤ 5
C_xH_y[a] (ppm)		≤ 0.5	≤ 0.1	≤ 2	≤ 2
CO_2 (ppm)			≤ 0.1	≤ 2	≤ 1
CO (ppm)			≤ 0.1	≤ 0.2	≤ 0.1
S[b] (ppb)				≤ 4	≤ 1
NH_3 (ppb)				≤ 0.1	≤ 1
Halogens (ppm)				≤ 0.05	≤ 0.05
Formaldehyde (ppm)				≤ 10	
CH_4 + N_2 + Ar (ppm)				≤ 100	
Particles (mg/kg)				≤ 1	
Total contami-nants (ppm)				≤ 300	
Applications	Metallurgy	Gaschromato-graphy	Semiconductors Calibration gases	PEMFC	PEMFC
Source	Linde	Linde	Linde		Ballard
Notes: (a) Excluding methane. (b) Sum of sulfur containing compounds in natural gas.					

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