# Hydrogen and its Storage for Mobility, a Challenge, not only for Materials Science and Technology

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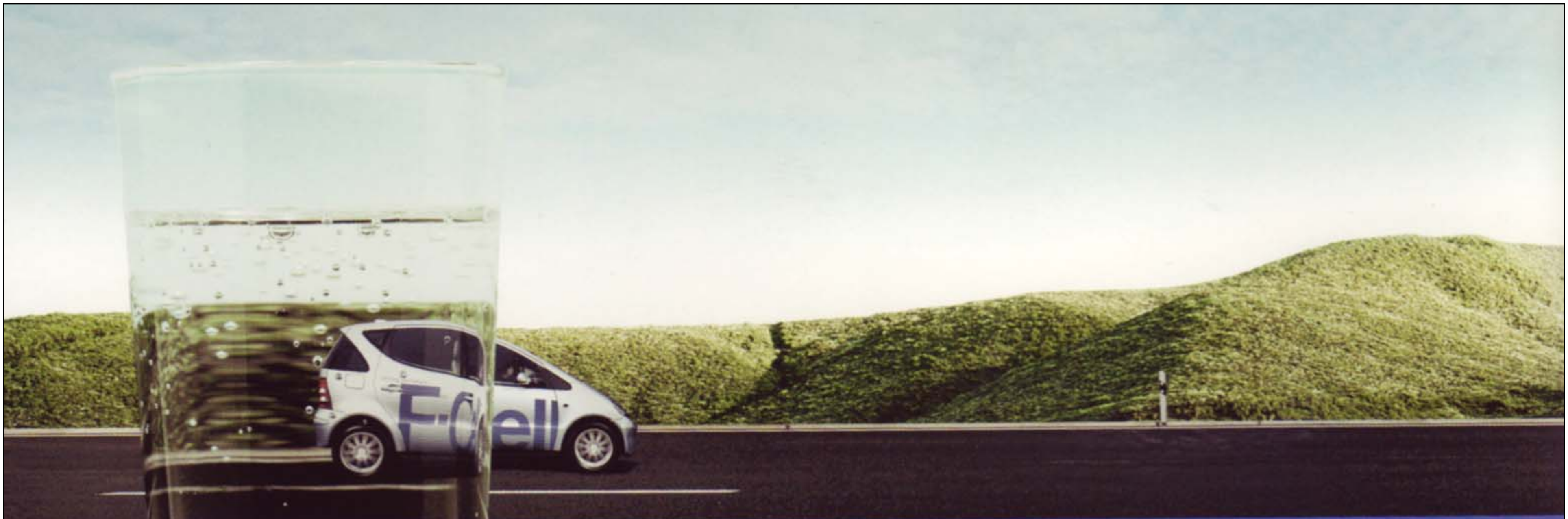
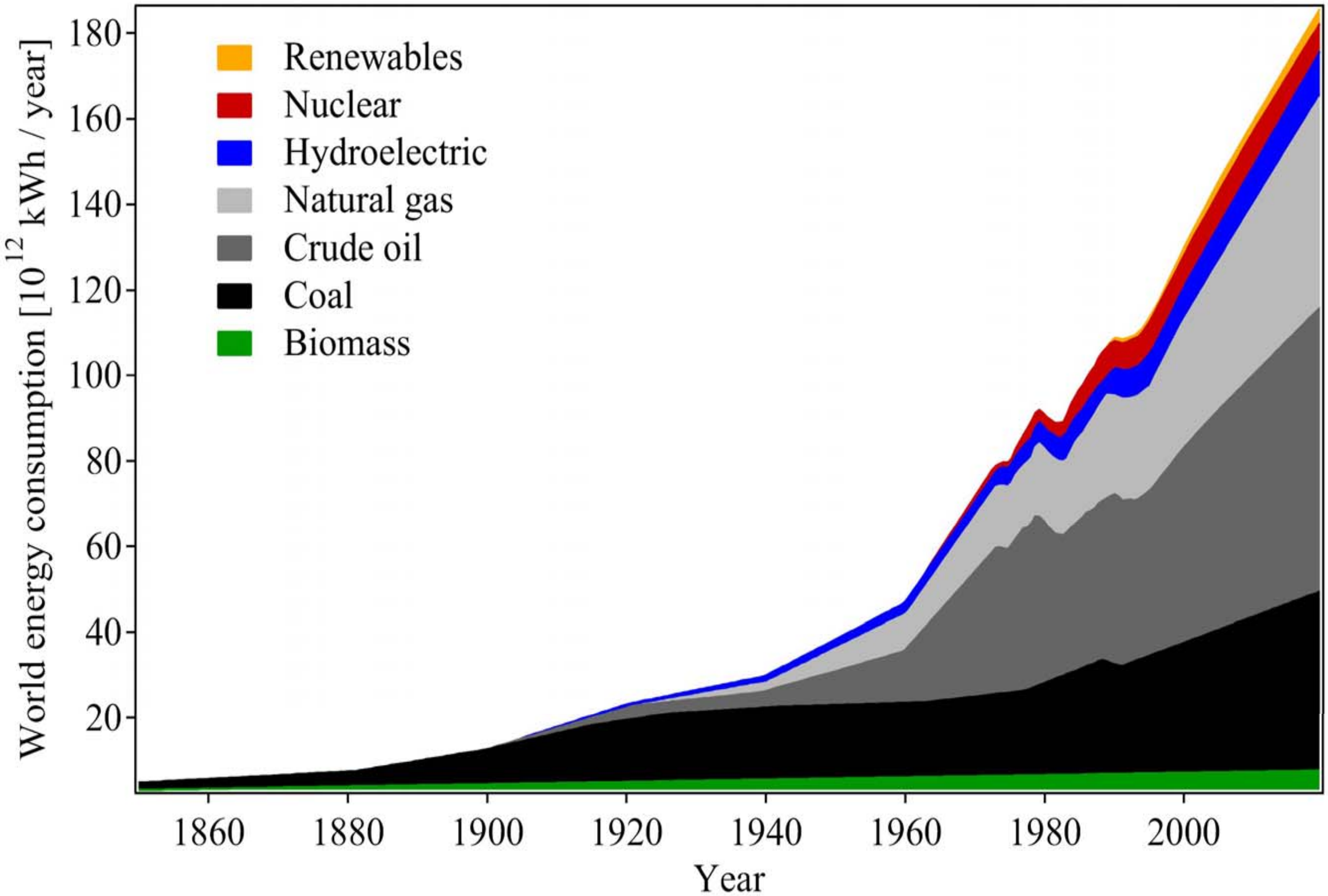


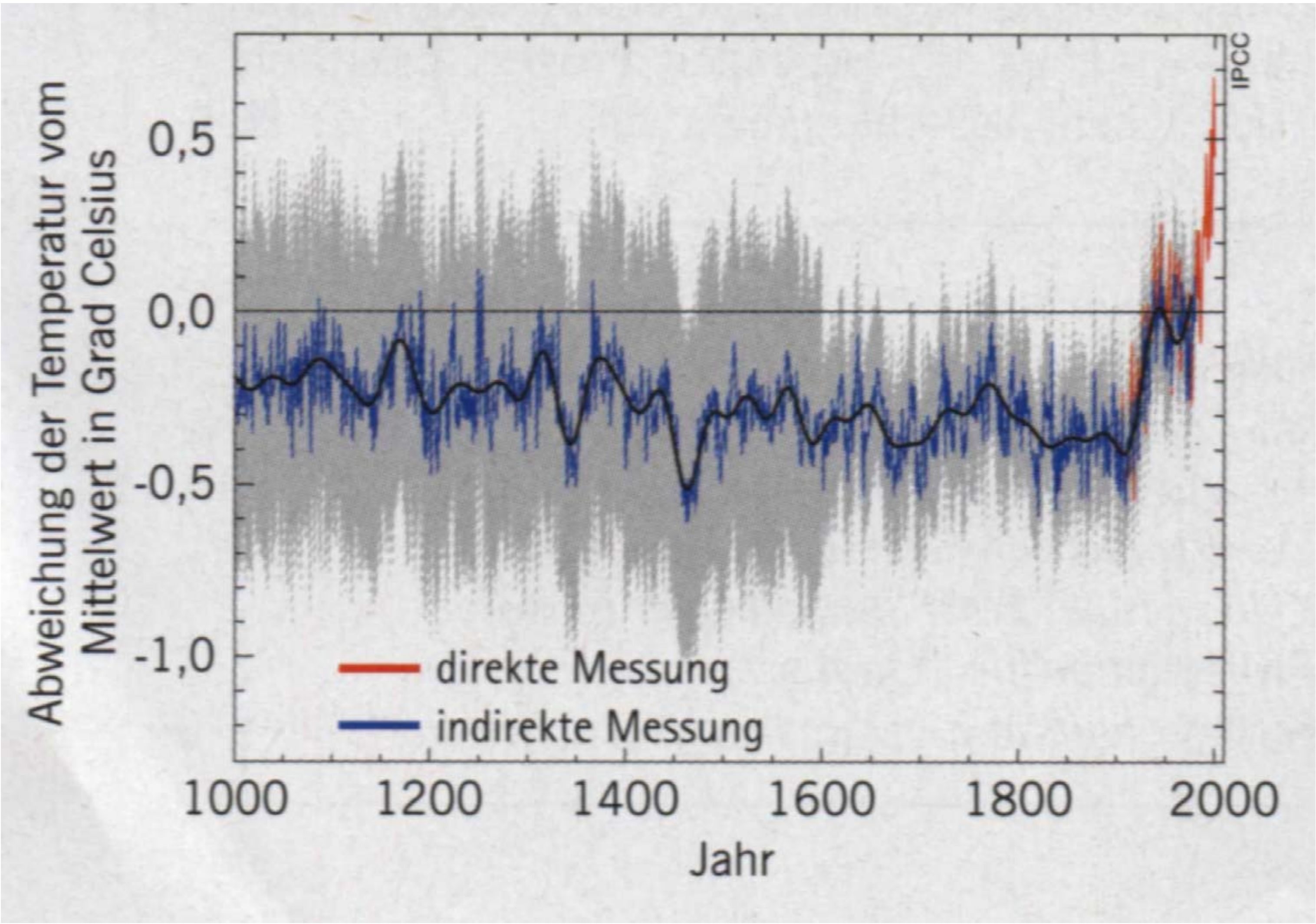
*"The primal element of all things is water*
*all things come from water and all things*
*return to water."*

*Thales of Miletus (circa 625 – 547 BC.)*

# WORLD ENERGY CONSUMPTION



# WORLD CLIMATE CHANGE




Spektrum der Wissenschaft Mai 2001, pp. 90-91

**Energy for Mobility** (fuel and its storage)

globalisation: mobility of persons
transport of goods

emissions, greenhouse gases, $CO_2$

global warming, more breathing problems

We have a problem!

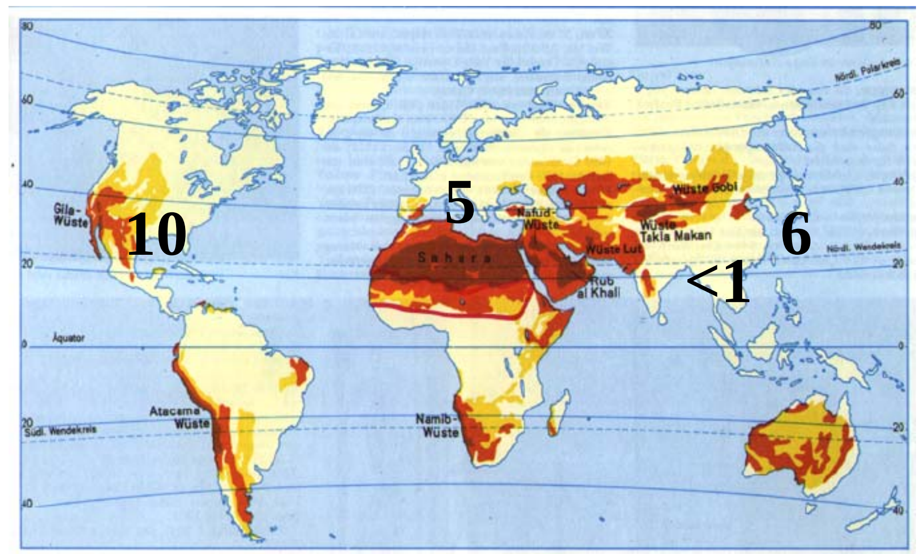
# Statistics related to energy consumption

# WORLD ENERGY ECONOMY

| Energy carrier | Demand | Reserve [years] |
|---|---|---|
| **Fossile** | | |
| Crude Oil | 32.7 % | 41 |
| Natural Gas | 19.5 % | 63 |
| Coal | 21.4 % | 218 |
| **Renewable** | | |
| Hydropower | 6.7 % | |
| Biomass | 11.6 % | |
| Others | 2.0 % | |
| **Nuclear** | 6.1 % | 100 |

20th Century

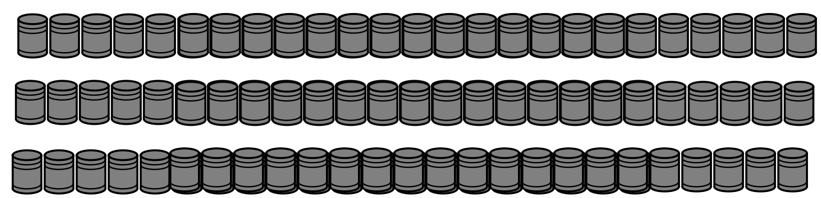
Average Power Consumption per Person **kW**




World = **2 kW/person** for 2 Bil. = **0 kW**

6

80



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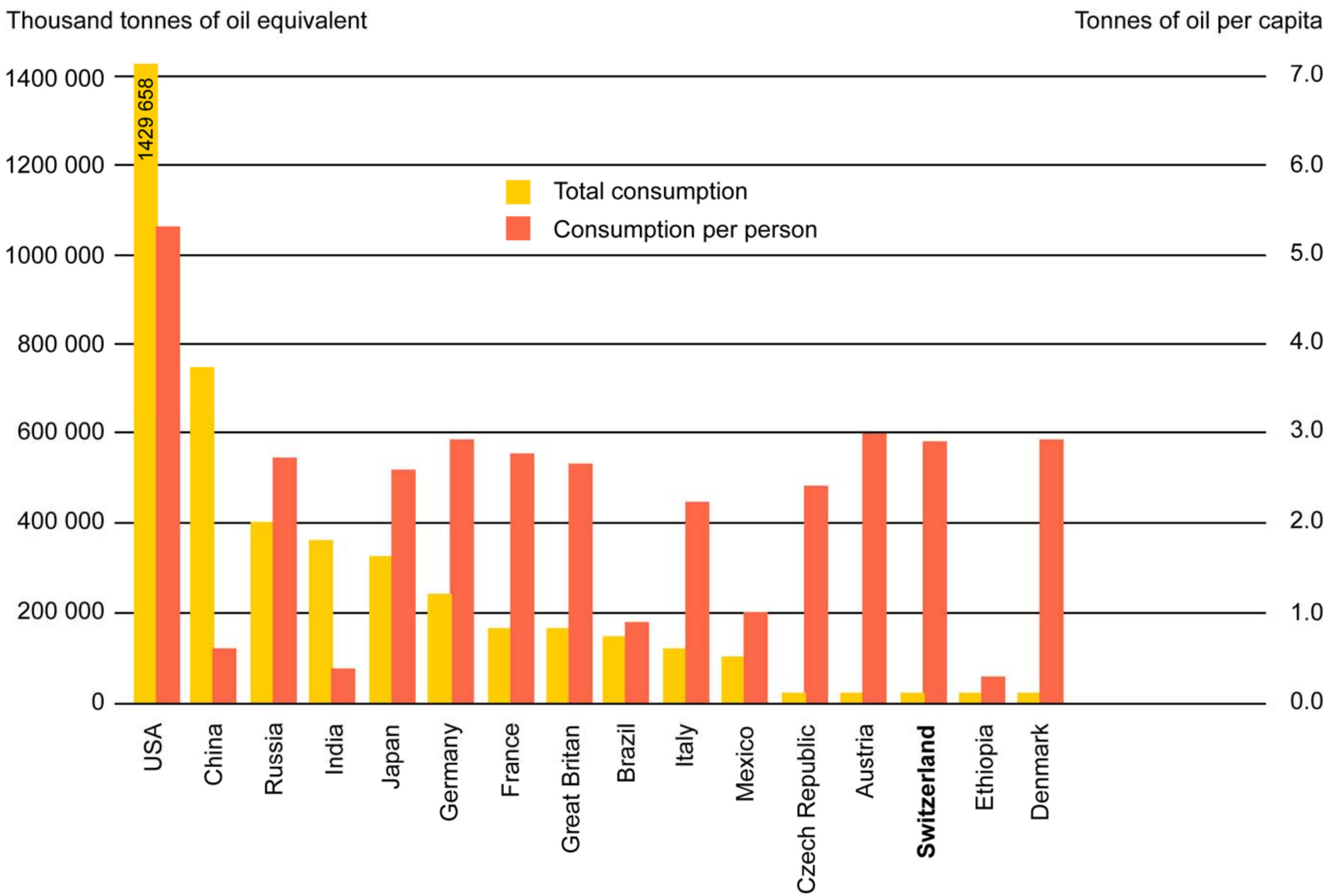
# International comparison of energy consumption 1998




Source: International Energy Agency 2001

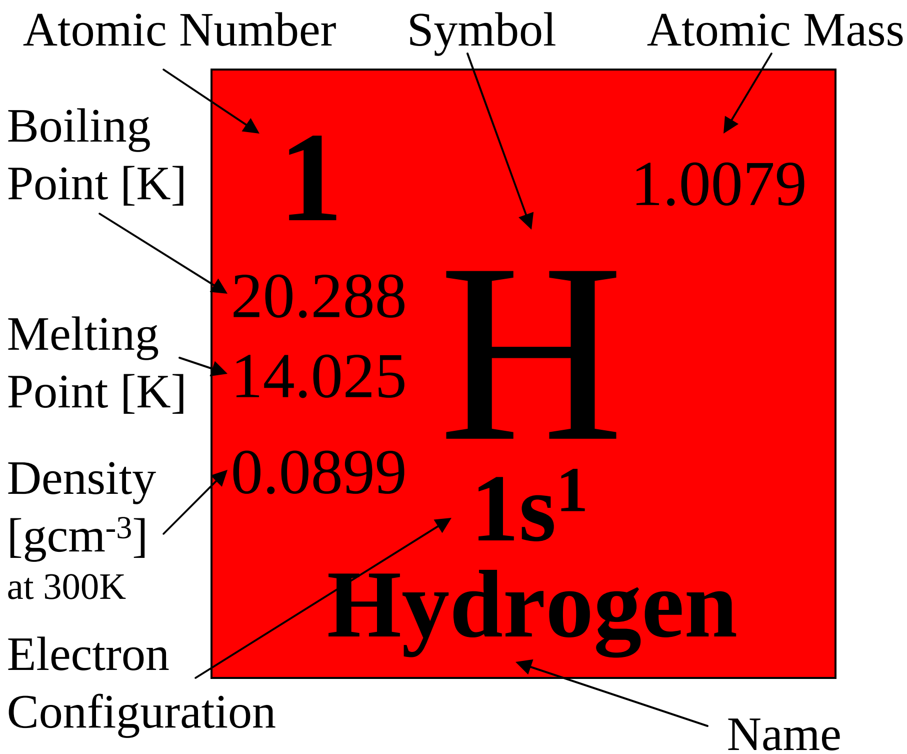
# Why hydrogen?

# Why not already today?

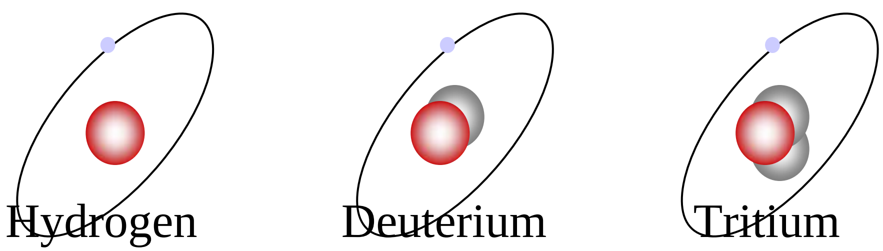
# Energy is linked to forces

| Natural forces | Ratio | Example | Store technique |
|---|---|---|---|
| Gravitation | $10^{0}$ | Mechanical | Hydropower |
| Weak Nuclear | $10^{33}$ | | |
| Electromagnetic | $10^{38}$ | Chemical | Hydrogen synthetic fuel<br>Electric battery |
| Strong Nuclear | $10^{40}$ | Nuclear fission, fusion | Nuclear fuel |

# Hydrogen




## Isotopes




## HYDROGEN on EARTH

$$2H_2O \rightleftharpoons 2H_2 + O_2$$

# Properties of hydrogen

non toxic, C-free gas, unlimited available as $H_2O$

simplest element of periodic table

best ratio of valence electrons to nucleons:
$1e^-$ per 1 proton, high binding energy 13.5 eV

isotopes D deuterium, T tritium for nuclear fission and fusion reaction

molecular gas $H_2$, liquid T< 21 K, solid T< 14K

transforms @ high pressure from molecular insulating solid into atomic metallic solid

high temperature superconductor?

# Use of Hydrogen

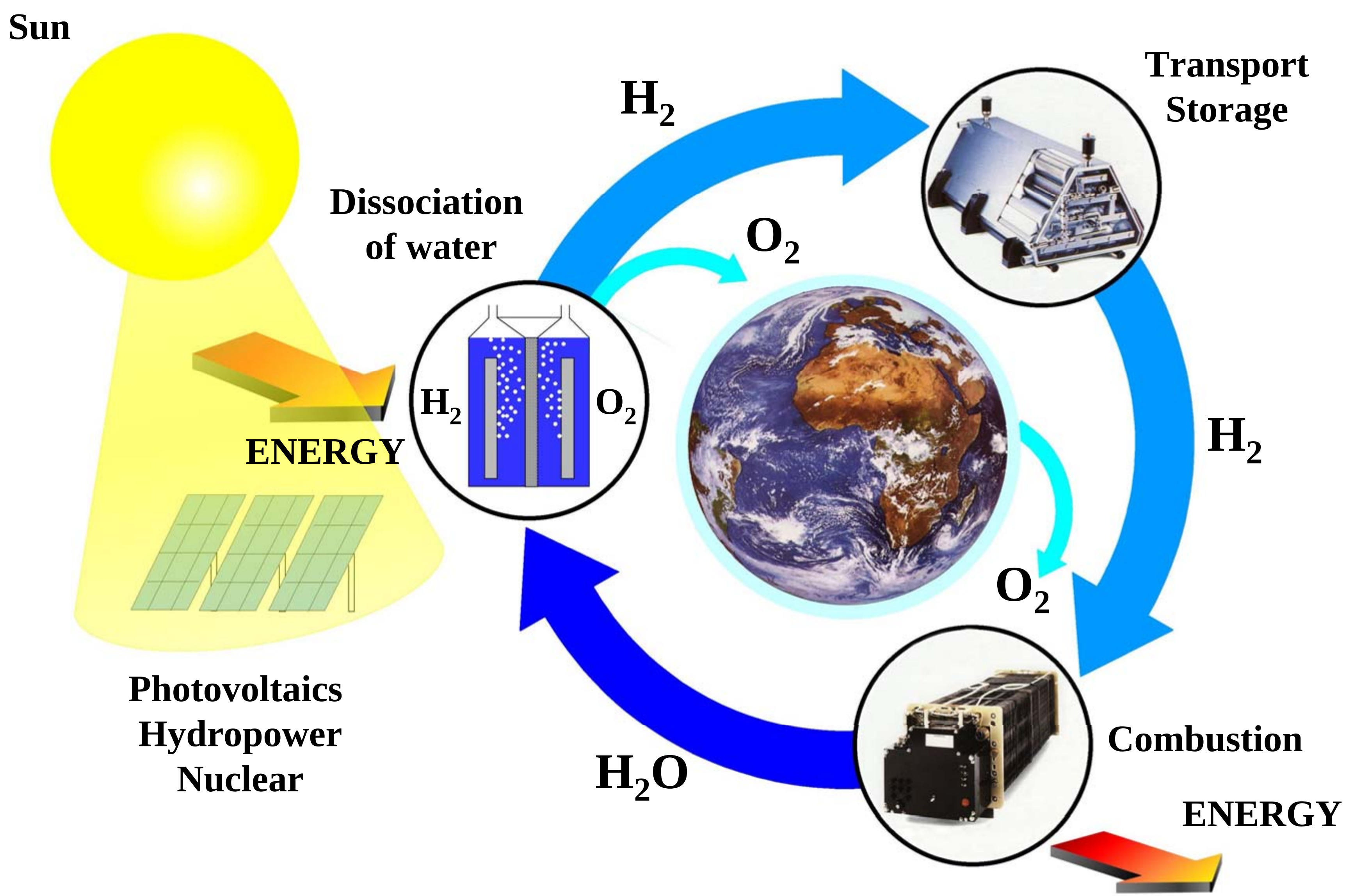
Store hydrogen, convert it $CO_2$ free into heat, electric or mechanical power

$H_2 + {}^1/_2\, O_2$ $H_2O$ + heat (combustion)
$H_2 + {}^1/_2\, O_2$ $H_2O$ + electricity (fuel cell)

# PROPERTIES OF FUELS

| Properties | | Hydrogen ($H_2$) | Methane ($CH_4$) | Gasoline ($-CH_2-$) |
|---|---|---|---|---|
| lower **heating value** | [$kWh \cdot kg^{-1}$] | **33.33** | 13.9 | 12.4 |
| self ignition temperature | [°C] | 585 | 540 | 228-501 |
| flame temperature | [°C] | 2045 | 1875 | 2200 |
| **ignition limits in air** | [Vol%] | **4 - 75** | 5.3 - 15 | 1.0 - 7.6 |
| minimal ignition energy | [mWs] | 0.02 | 0.29 | 0.24 |
| flame propagation in air | [$m \cdot s^{-1}$] | 2.65 | 0.4 | 0.4 |
| detonation limits | [Vol%] | 13 - 65 | 6.3 - 13.5 | 1.1 - 3.3 |
| detonation velocity | [$km \cdot s^{-1}$] | 1.48 - 2.15 | 1.39 - 1.64 | 1.4 - 1.7 |
| explosion energy | [$kg\ TNT \cdot m^{-3}$] | 2.02 | 7.03 | 44.22 |
| **diffusion coefficient** in air | [$cm^2 \cdot s^{-1}$] | **0.61** | 0.16 | 0.05 |

# HYDROGEN CYCLE



# Is Hydrogen a Safe Energy Carrier ?

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# LZ 129 “Hindenburg”

New York / Lakehurst, May 6th 1937, 6 pm

**Accident:**

While the airship was landing she has got on fire about 80 meters above ground level and crashed.

Fatalities:

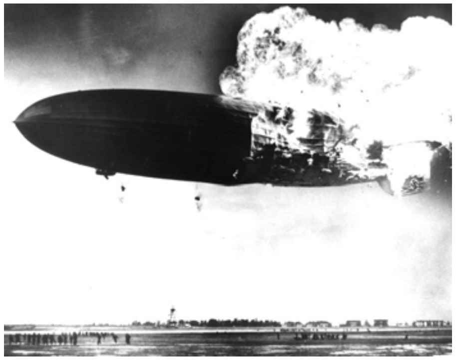
13 of 36 passengers,

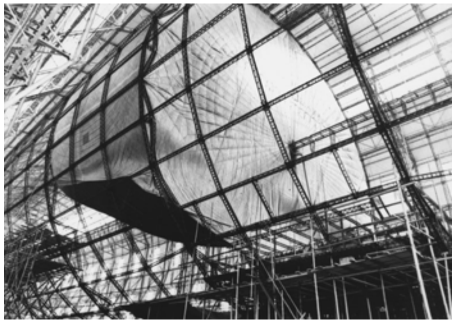
22 of 60 crew members

1 member of 228 ground staff holding the ship.

# Cause of fire

**New investigation: The inflammable skin of the Hindenburg was ignited by an electric discharge arc between the electrostatic charged skin and the grounded metallic frame.**





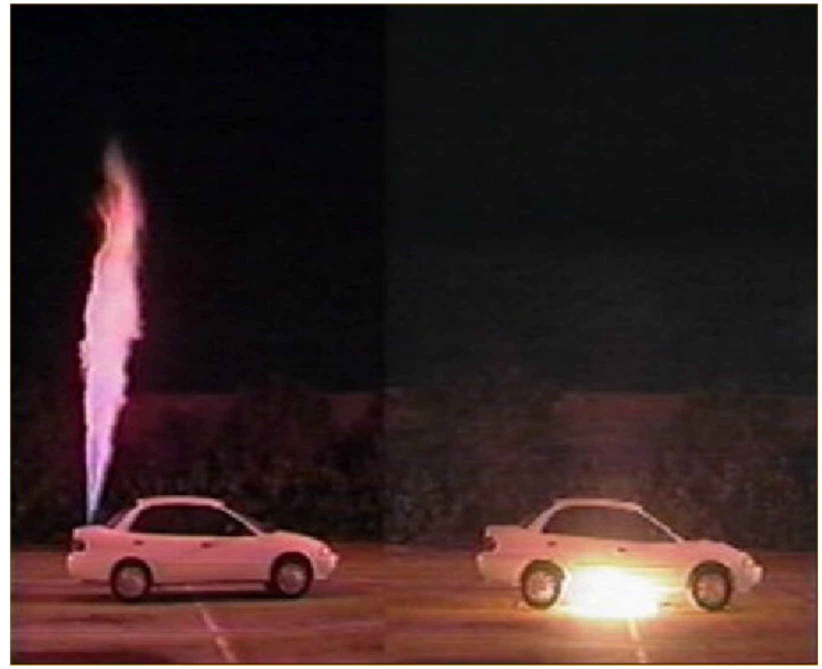
Ref.: Addison Bain, Wm. D. Van Vorst, "The Hindenburg tragedy revisited: the fatal flaw found", Int. Journal of Hydrogen Energy 24 (1999), pp. 399-403

# FUEL LEAK SIMULATION

## Before ignition t = 0 s

Hydrogen powered vehicle on the left.
Gasoline powered vehicle on the right.

## Ignition t = 3 s



Ignition of both fuels occur.
Hydrogen flow rate 2100 SCFM (0.18 $m^3$/min.)
Gasoline flow rate 680 $cm^3$/min.

Ref.: Michael R. Swain, University of Miami, Coral Cables, FL 33124, USA

# FUEL LEAK SIMULATION

## t = 60 s

Hydrogen flow is subsiding, view of gasoline vehicle begins to enlarge

## t = 90 s

Hydrogen flow almost finished. View of gasoline powered vehicle has been expanded to nearly full screen.

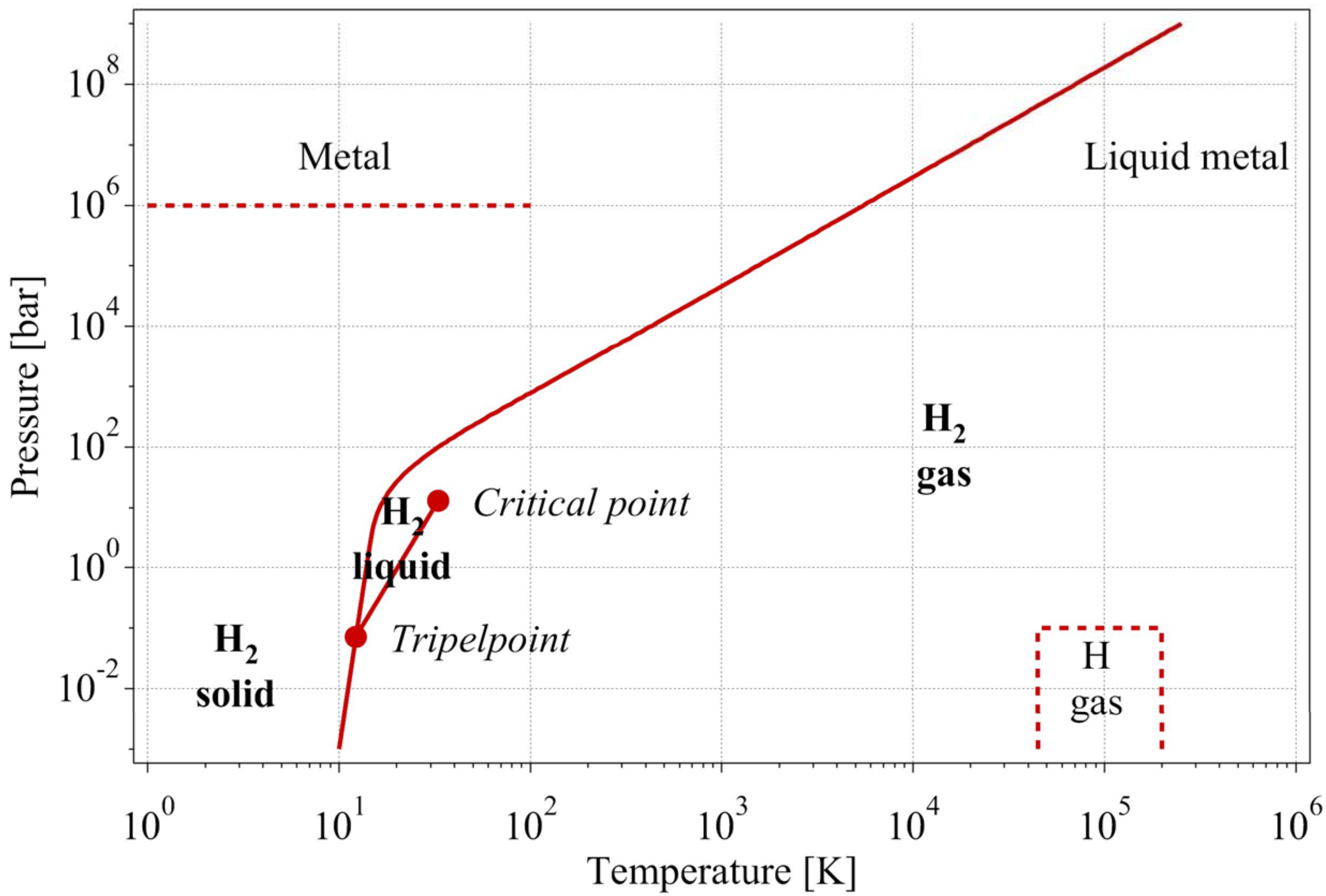
Ref.: Michael R. Swain, University of Miami, Coral Cables, FL 33124, USA

# Why do we use hydrocarbons instead of hydrogen?

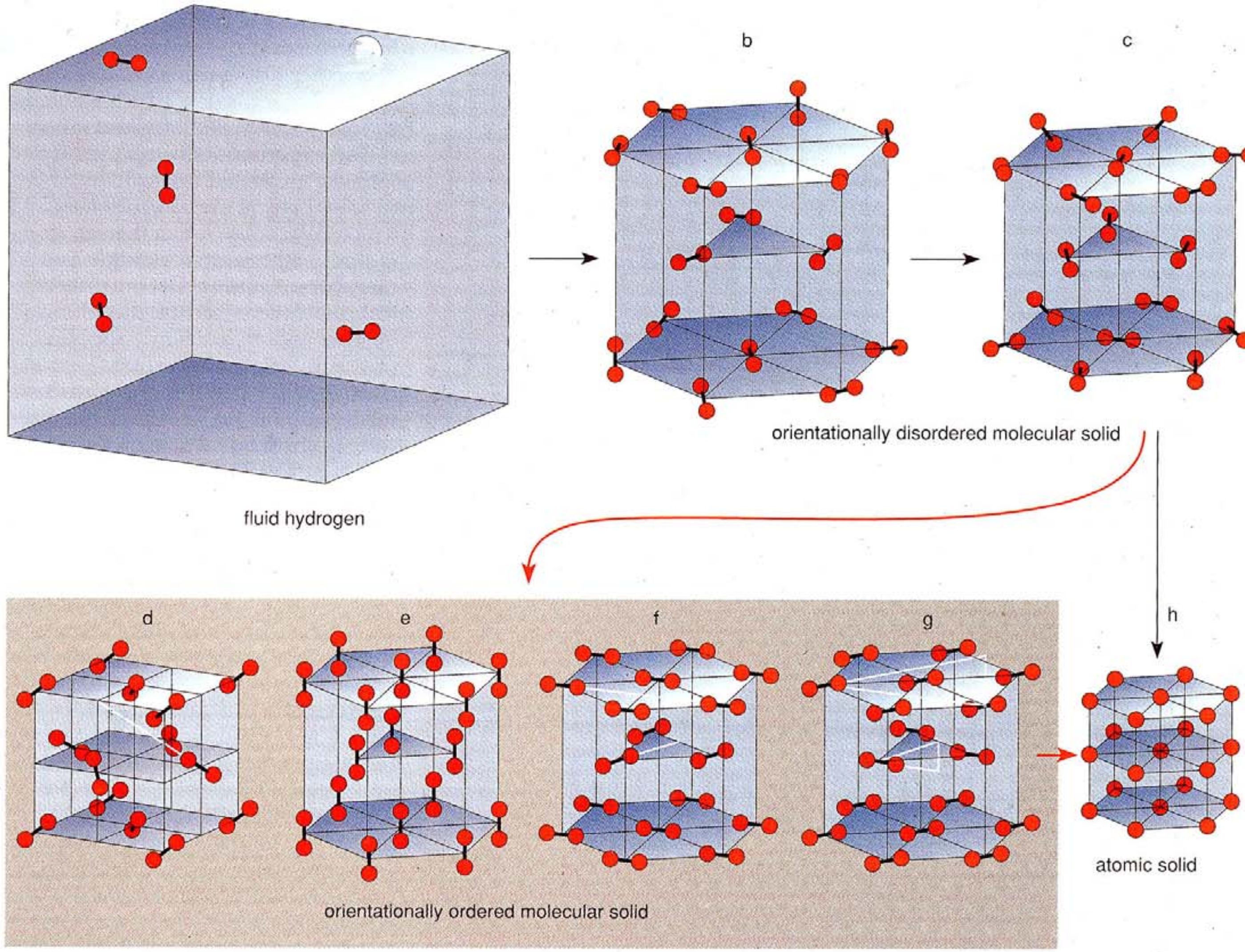
hydrogen is an ideal gas @ ambient conditions
1 mol $H_2$ = 2g = 22,4l = 284 kJ = 80 Wh

compacting by 1000 needed

# PRIMITIVE PHASEDIAGRAM OF HYDROGEN




Ref:: W. B. Leung, N. H. March and H. Motz, Physics Letters 56A (6) (1976), pp. 425-426



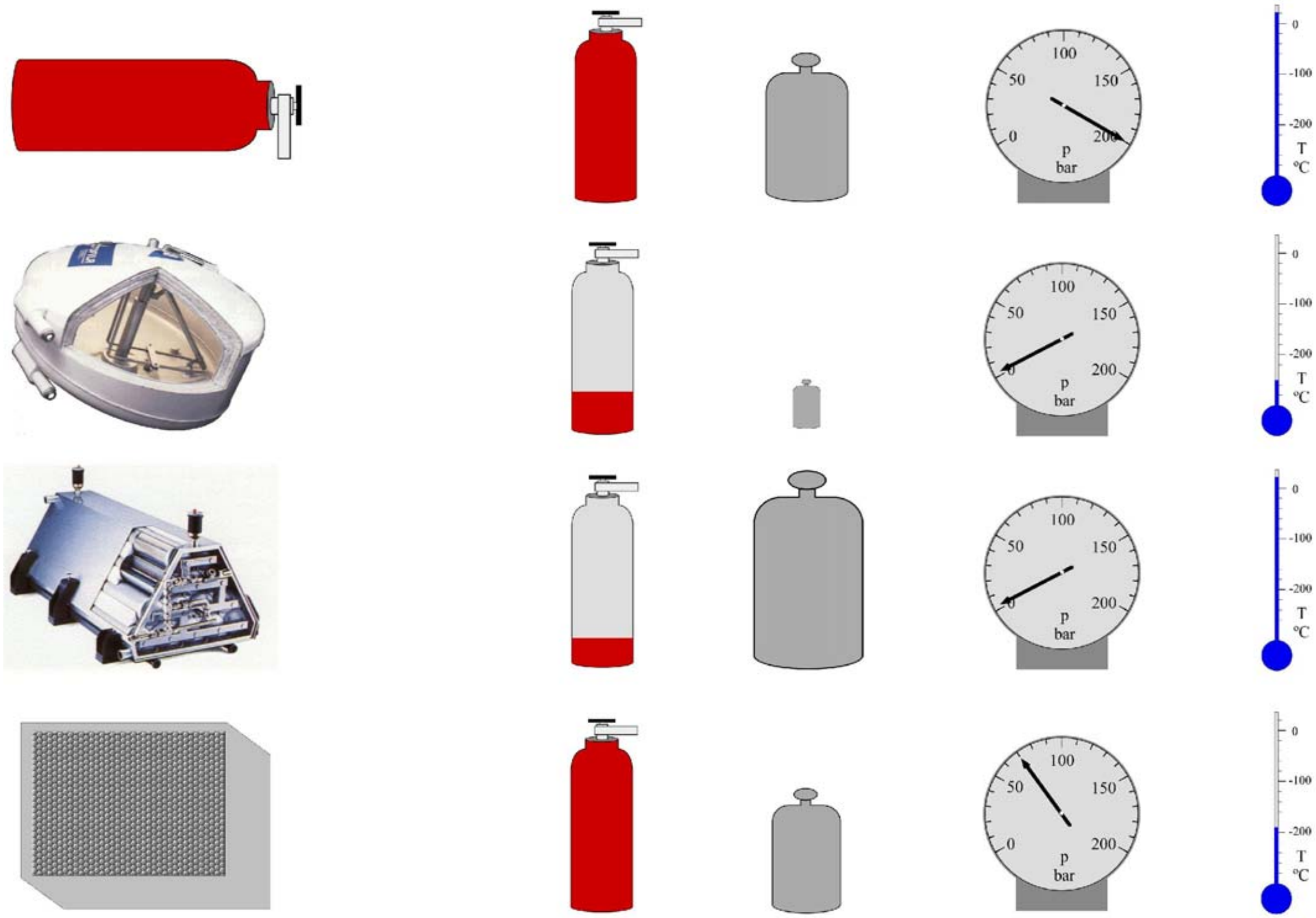

simplest metal

Figure 5. Hydrogen passes through several transformations as the pressure is increased. In the fluid phase *(a)* $H_2$ molecules are dispersed randomly and at low density. The solid phase formed at 54 kilobars *(b)* has a hexagonal close-packed structure; further increases in pressure up to well beyond a megabar drive the molecules closer together but do not alter the fundamental structure *(c)*. In this phase the molecules remain randomly oriented: Viewed classically, the axis defined by the bond between hydrogen atoms can point in any direction, and indeed the molecules are continually tumbling. With further pressure increases hydrogen may enter a phase with orientational order; four candidate structures are shown *(d-g)*, although none of them has been confirmed experimentally at very high pressures. The ultimate fate of hydrogen, as the pressure continues to rise, must be to form an atomic metal *(h)*, where the $H_2$ molecules have ceased to exist.

Ho-kwang Mao and Russell J. Hemley 234 American Scientist, Volume 80 1992 May–June 239

# Hydrogen storage

Storage Media   Volume   Mass   Pressure   Temp.




**Hydrogen gas**
**(298 K, 25°C)**
0.01 mol $H_2 \cdot cm^{-3}$
at 200 bar

**Liquid hydrogen**
**(21 K, -252°C)**
0.0708 $g \cdot cm^{-3}$
0.0354 mol $H_2 \cdot cm^{-3}$
at 1 bar

**Absorbed hydrogen**
**(298 K, 25°C)**
e.g. $LaNi_5H_6$
0.05 mol $H_2 \cdot cm^{-3}$
at 2 bar

**Adsorbed hydrogen**
**(65 K, -208°C)**
0.01 mol $H_2 \cdot cm^{-3}$
at 70 bar

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# VOLUME OF HYDROGEN STORAGE MEDIA

**4 kg hydrogen = 560 $MJ_{therm.}$**

**$Mg_2FeH_6$ $LaNi_5H_6$ $H_2$ (liquid) $H_2$ (200 bar)**

3 l gasoline / 100 km = 9 $kWh_{mech.}$ / 100 km = 32 $MJ_{mech...}$ / 100 km

Ref.: L. Schlapbach & A. Züttel, NATURE, **414,** 2001, 353-358

# LIQUID HYDROGEN AS FUEL FOR CARS


BMW 745i refilled with liquid hydrogen

Liquid hydrogen tank

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# High pressure vessels

Assumptions

gas pressure p = 1000 bar = 100 MPa

tensile strength $\sigma$ = 3000 MPa (modern composites)

cylinder diameter D = 0.1 m

–> wall thickness $d = \frac{P}{20} \cdot D = \frac{100}{6000} 0.1\ m = 1.4\ mm$

composite density $\rho = 3\ g/cm^3$

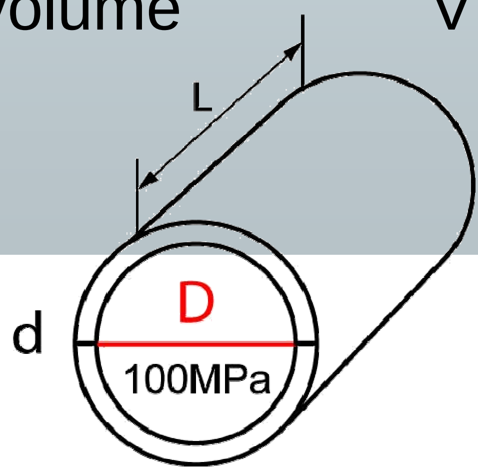
length L = 1m

gas volume V = 8 l

1,5 kg container

0,5 kg $H_2$

–> 25 mass% $H_2$



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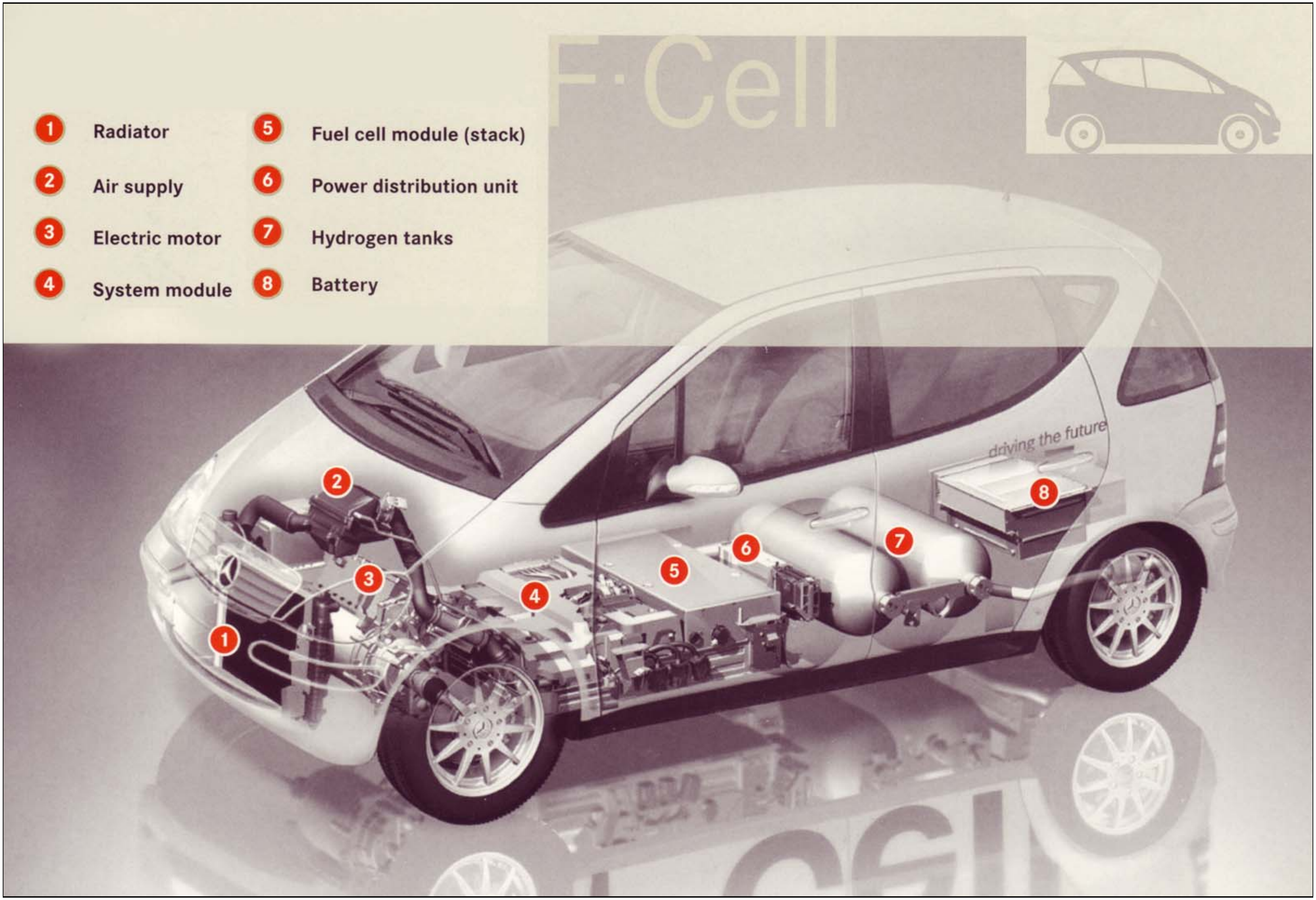
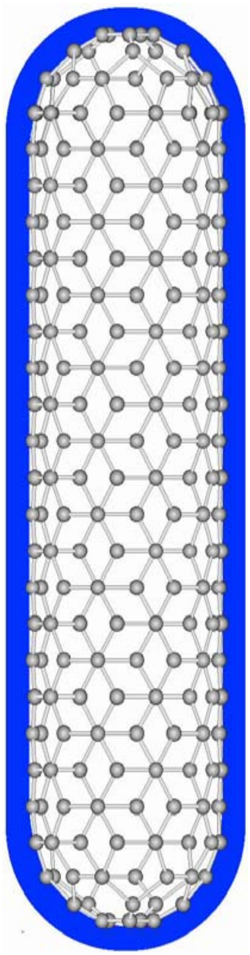
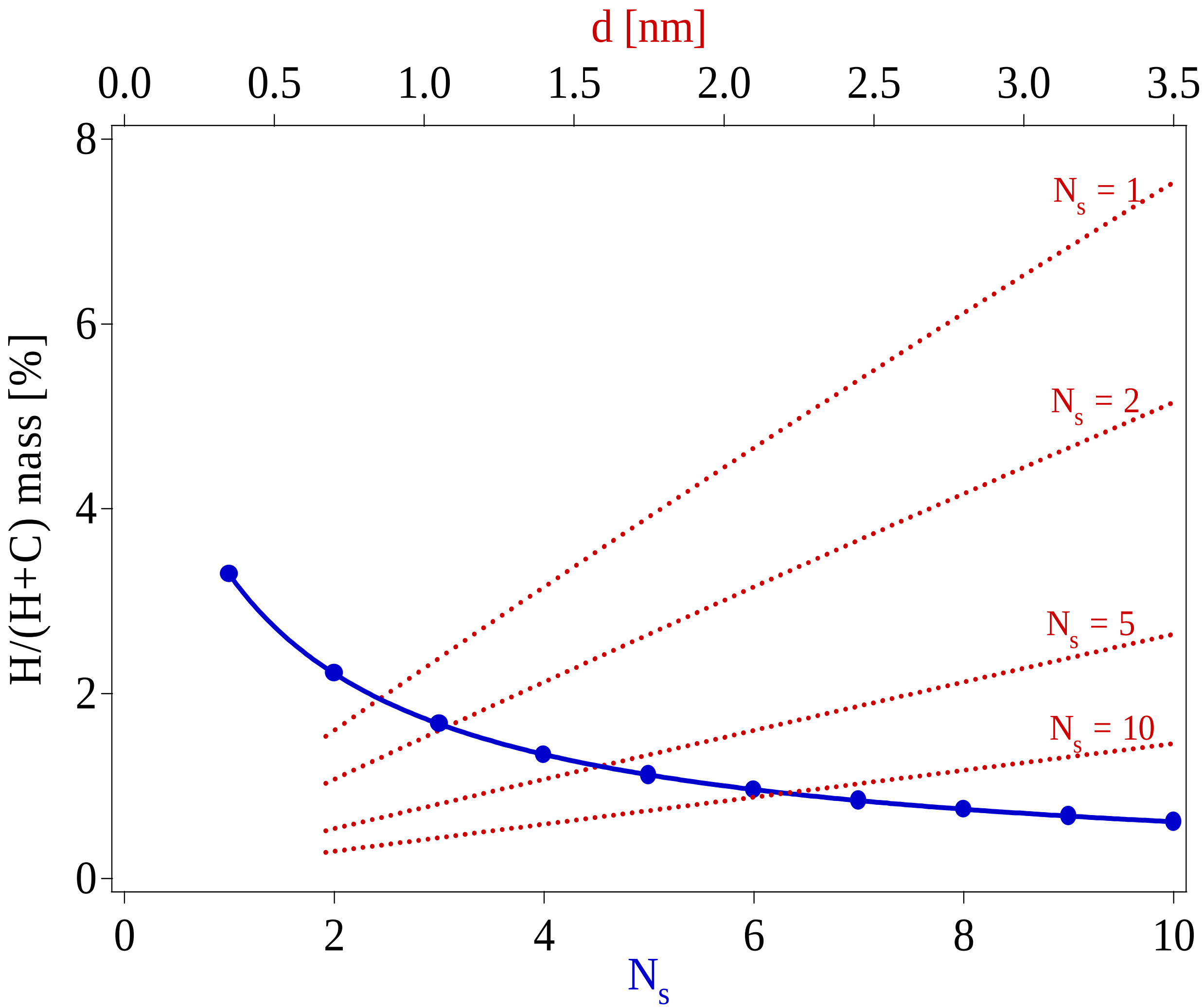
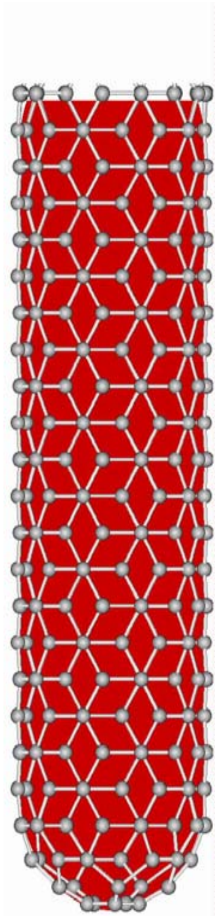

F·Cell
1 Radiator
2 Air supply
3 Electric motor
4 System module
5 Fuel cell module (stack)
6 Power distribution unit
7 Hydrogen tanks
8 Battery
driving the future

# Hydrogen condensation in and on nanotubes





# Solution and metal hydride formation

pressure composition isotherms

thermodynamics

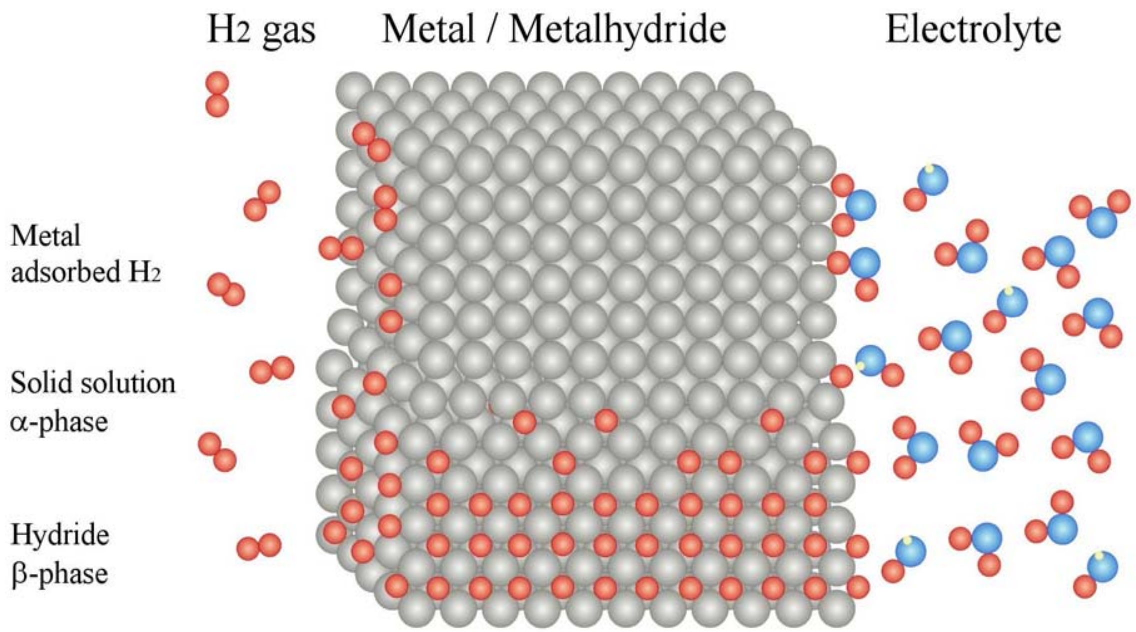
kinetics, diffusion

electronic structure

# Hydrogen absorbtion mechanism

$H_2$ gas phase

alkaline electrolyte

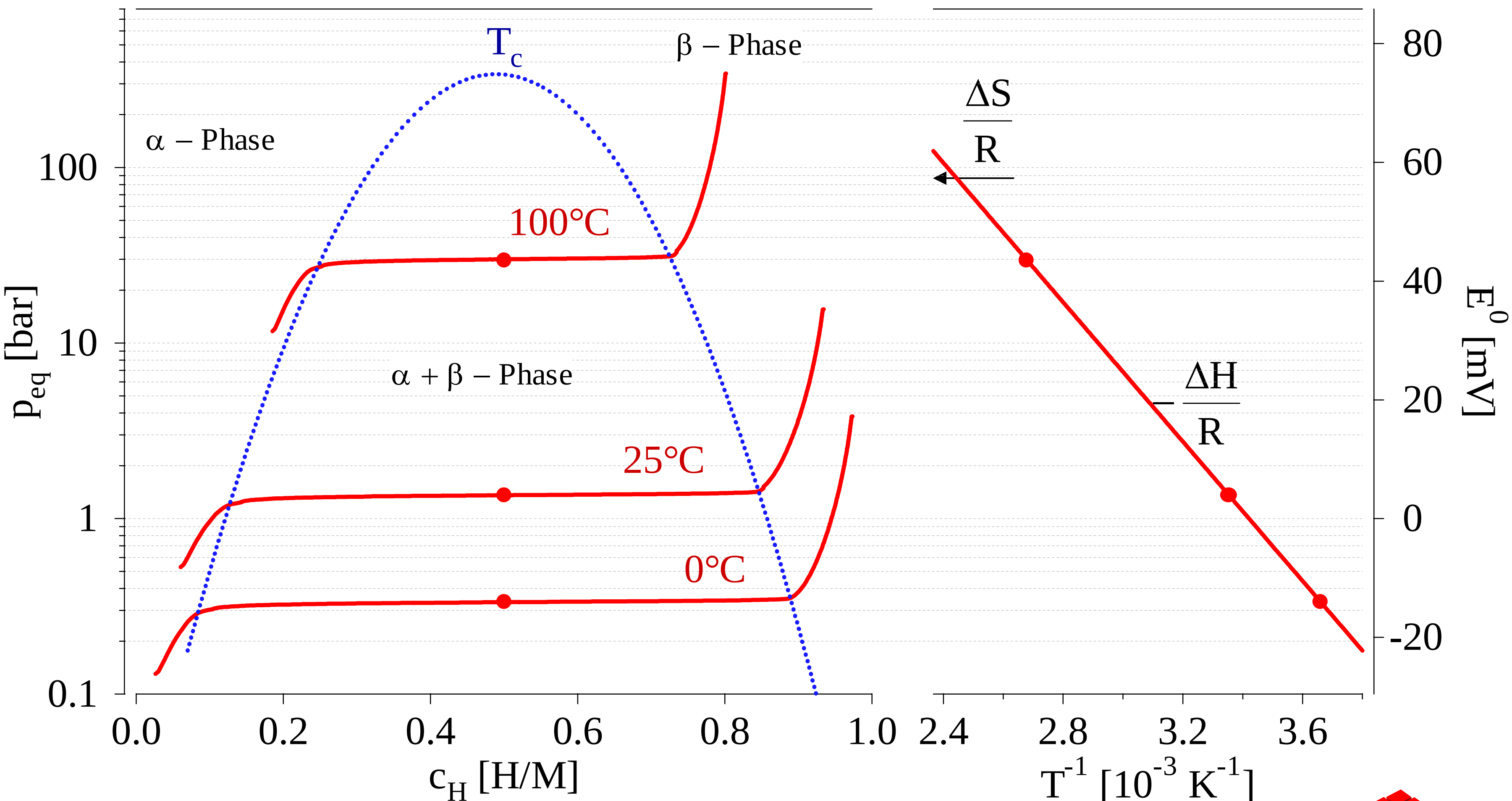


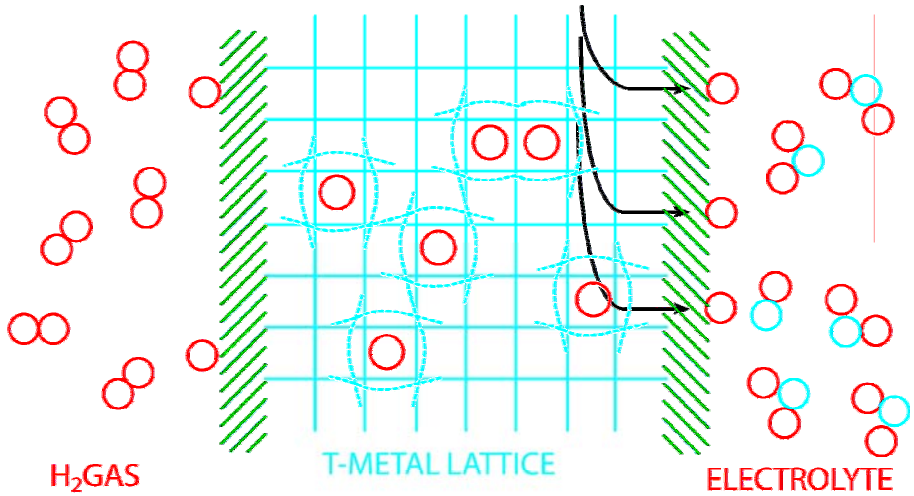

1) Physisorption of $H_2$ molecules
2) Dissociation (activation barrier)
3) Chemisorption of H-atoms
4) Diffusion of H-atoms
5) Intercalation

1) Physisorption of $H_2O$ molecules
2) Electron transfer (desorption of $OH^-$ )
3) Chemisorption of H-atoms
4) Diffusion of H-atoms
5) Intercalation

# Phase Diagram of Metal Hydrides (LaNi5)

$$R \cdot T \cdot \ln\left(\frac{p}{p_0}\right) = \Delta H - T \cdot \Delta S$$






$H_2 \rightleftharpoons 2H_{ads}$

$H_2O + e^- \rightleftharpoons H_{ads} + OH^-$

CRYSTAL STRUCTURE

LATTICE EXPANSION

ELECTRONIC STRUCTURE

→ smaller band width

PROTON: ATTR.POT. for $e^-$

→ lowering of states

→ H-induced bonding band

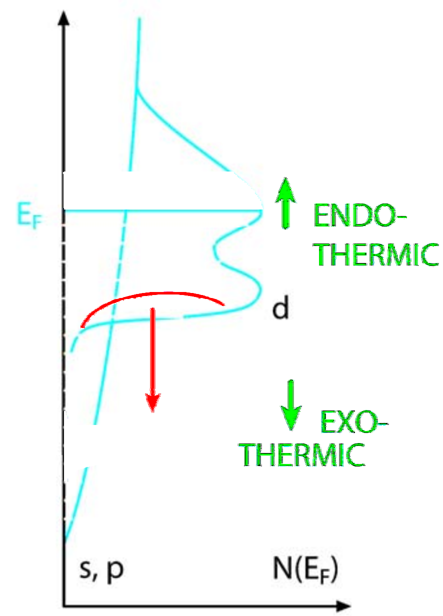
H-ELECTRON

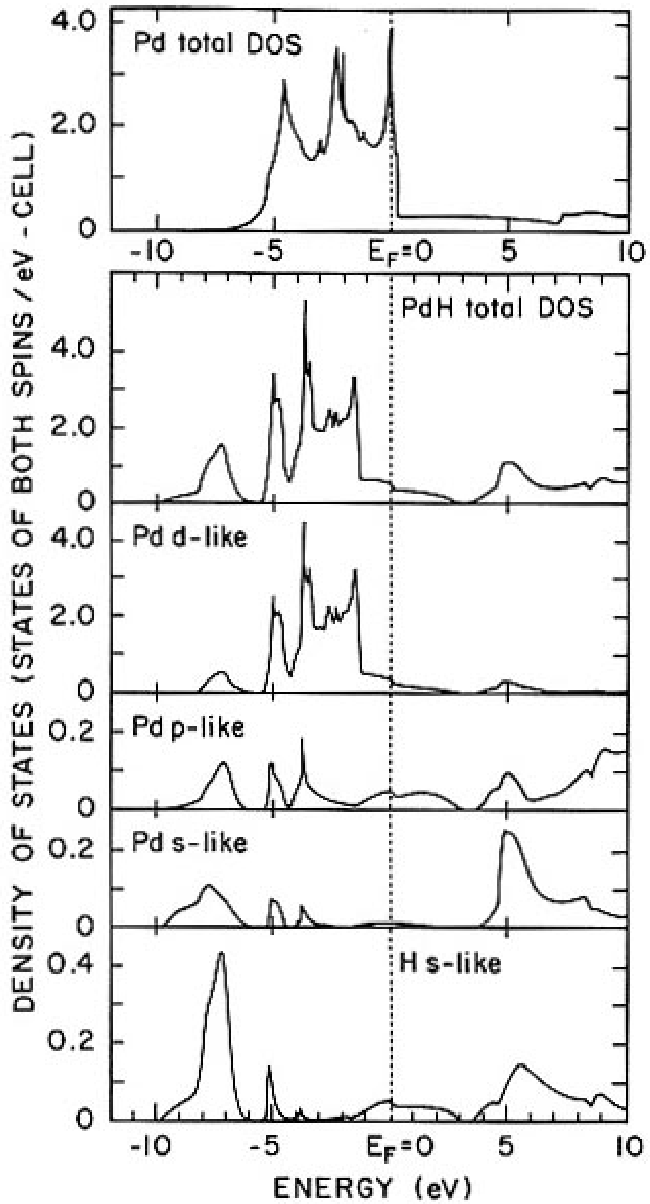
→ shift of $E_F$

→ charge transfert

H-H INTERACTION

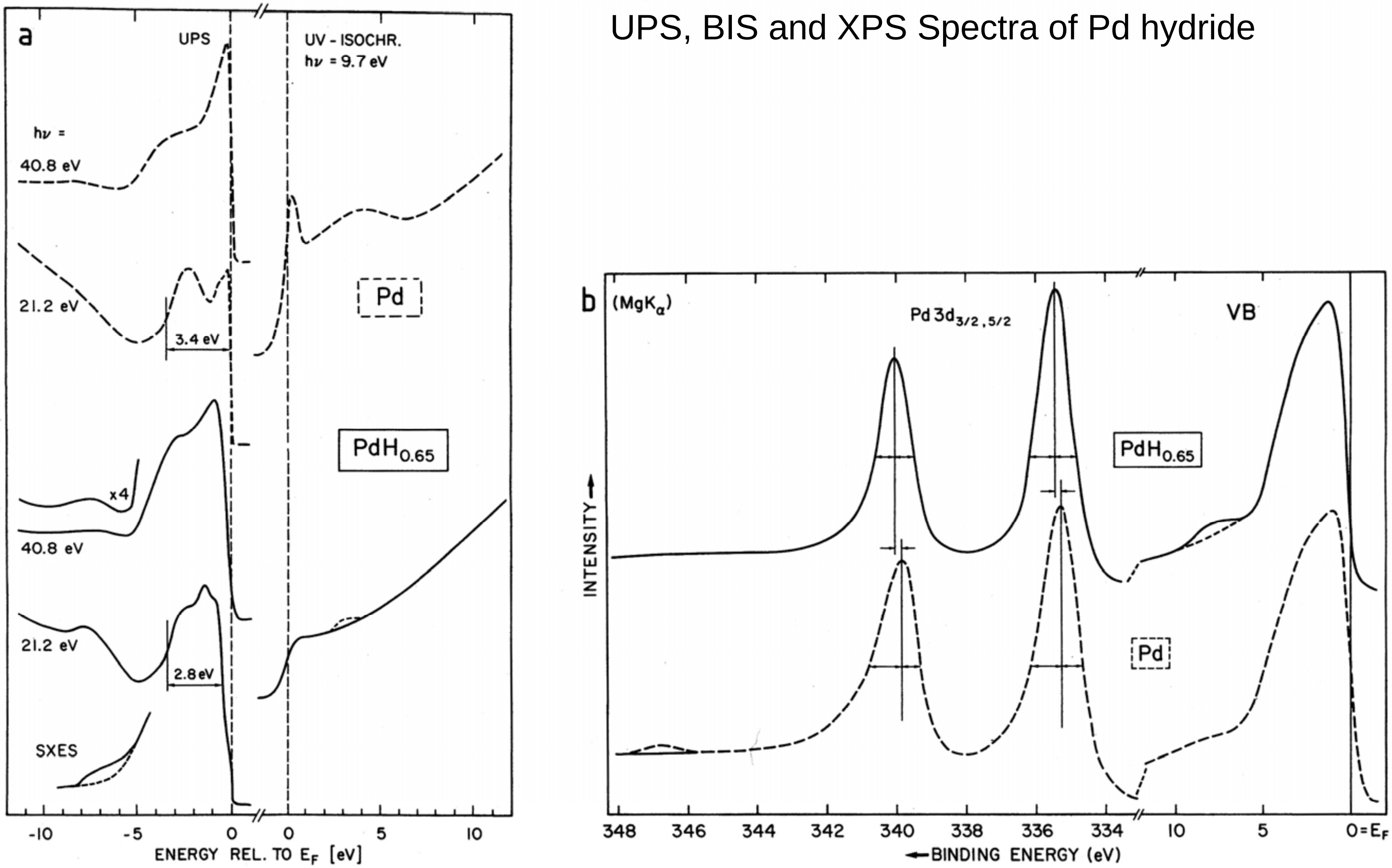
PHONONS → harder






Slater-Koster density of states for Pd
And total and partial wave
Analysis of the DOS of PdH
(D. Papaconstantopoulos)

UPS, BIS and XPS Spectra of Pd hydride

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# Intermetallic compounds for reversible hydride formation

| | |
|---|---|
| hexagonal | $AB_5$, $LaNi_5$, |
| cubic | $AB$, FeTi |
| | $A_2B$, $Mg_2Ni$ |
| | $AB_2$, $Zr_2Ni$, $Mn_2Ni$, |

$AB_5$, $AB$, $AB_2$ type hydrides:
Reversible cycling 10 000 times
Safe room temperature operation
Compact, but very heavy (1.5 -3 w% H) and expensive

# Other materials to adsorbe or intercalate hydrogen

layered structures

| | |
|---|---|
| hexagonal | $Al\ B_2$ |
| trigonal | $Ca\ Si_2$ |
| orthogonal | $Ru\ B_2$ |
| hexagonal | $Re\ B_2$ |
| hexagonal | $W\ B_2$ |

high surface area nanostructures

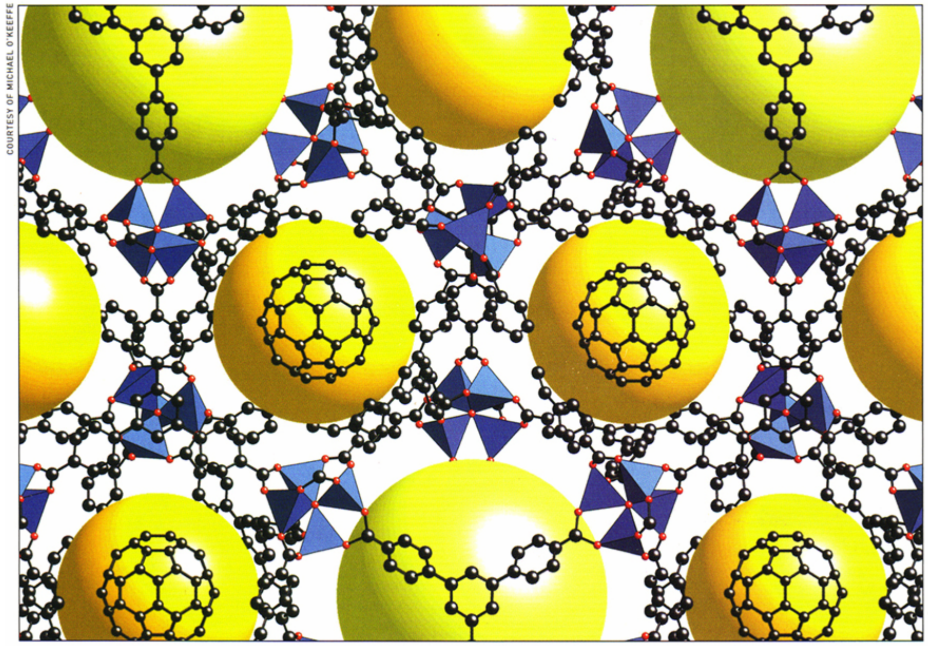
$Li\ Al\ O_2$

high (open) porosity nanostructures

zeolites

# Some metal hydrides discovered and/or characterized at the Uni Geneva (Klaus Yvon)

| compound | space group | structure | H density (wt %) | H density (gH/ler) | Tdes (1 bar) |
|---|---|---|---|---|---|
| **Complex transition metal hydrides**)** | | | | | |
| $BaReH_9$ | $P6_3/mmc$ | $[ReH_9]^{2-}$ttp | 2.7 | 134 | <100°C |
| $Mg_2FeH_6$ | Fm-3m | $[FeH_6]^{4-}$oct | 5.5 | 150 | 320°C |
| $Ca_4Mg_4Fe_3H_{22}$ | P-43m | $[FeH_6]^{4-}$oct | 5.0 | 122 | 395°C |
| **Intermetallic hydrides**)** | | | | | |
| $Zr_2CoH_5$ | P4/ncc | new type | 2.0 | 123 | >400°C |
| $Zr_6FeAl_2H_{10}$ | P-62c | filled $Fe_2P$ | 1.5 | 80 | >400°C |
| **Saline hydrides**)** | | | | | |
| $Ca_4Mg_3H_{14}$ | P-62m | new type | 5.7 | 98 | >400°C |
| $Ba_6Mg_7H_{26}$ | C2/m | $Ba_6Zn_7H_{26}$ | 2.6 | 82 | >400°C |




**MOLECULAR CONTAINERS** Developing suitable storage media for hydrogen is critical to capitalizing on the gas's potential benefits as an energy carrier. Among other candidates, this metal-organic framework compound—MOF-177, composed of zinc clusters (blue) and 1,3,5-benzenetribenzoate units—is being studied for gas uptake because of its large pore volume (yellow spheres).

# FILLING UP WITH HYDROGEN

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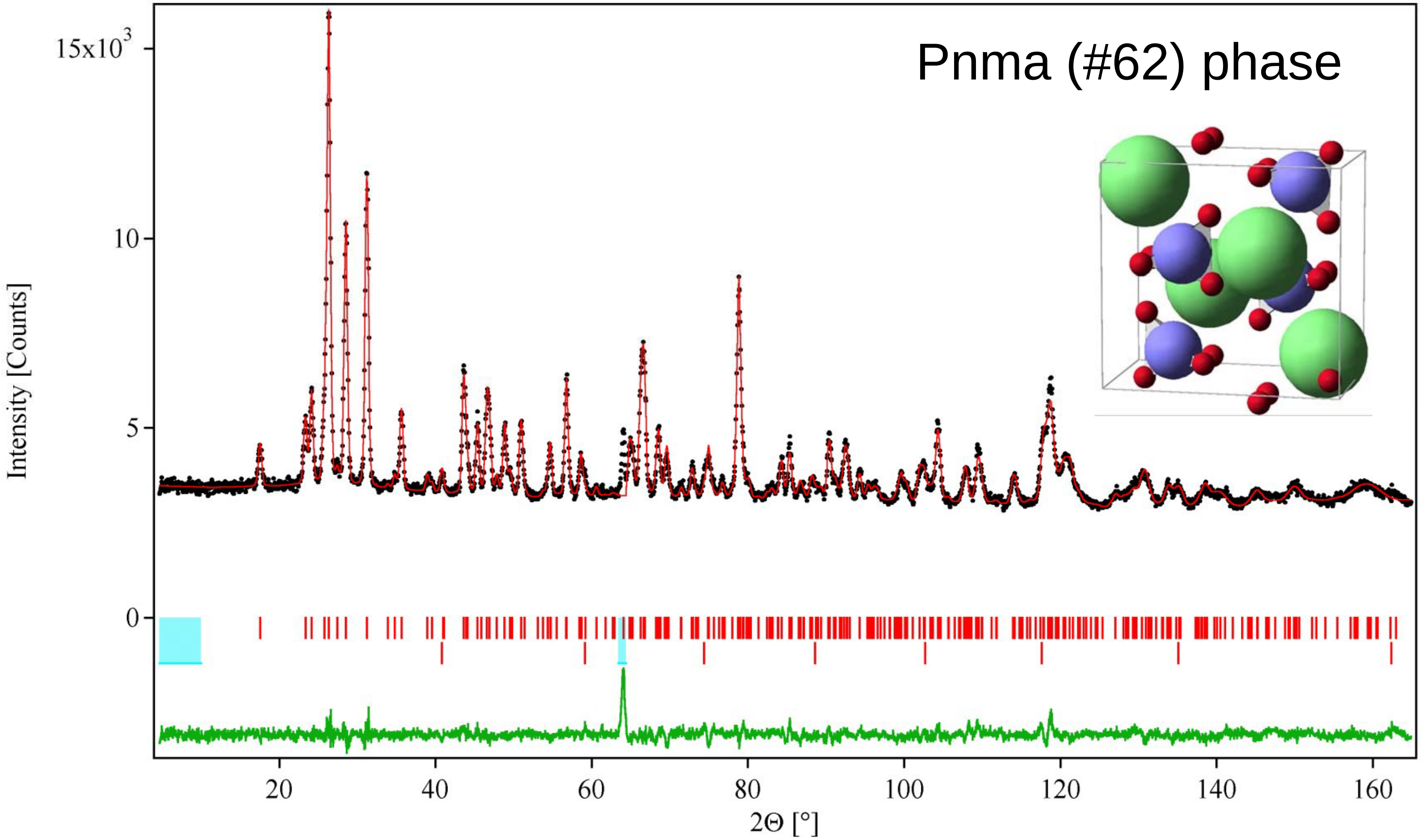
**Complex hydrides**
**Hydrogen generation by the hydrolysis of alkaline borohydrides**

| MH complex | Mol. mass | H-content (mass%) | H-generated (mass%) (Hydrolysis) |
|---|---|---|---|
| $LiAlH_4$ | 37.93 | 10.53 | 10.82 |
| $LiBH_4$ | 17.85 | 22.41 | 14.86 |
| $KAlH_4$ | 70.08 | 5.71 | 7.54 |
| $KBH_4$ | 53.91 | 7.42 | 8.90 |
| $NaAlH_4$ | 53.97 | 7.41 | 8.89 |
| $NaBH_4$ | 37.70 | 10.61 | 10.85 |

# NEUTRON DIFFRACTION OF $LiBD_4$




A. Züttel et al., J. of Alloys and Compounds 356–357 (2003), 515–520

# STRUCTURE OF $LiBH_4$ AT 408K (135°C)

hexagonal symmetry

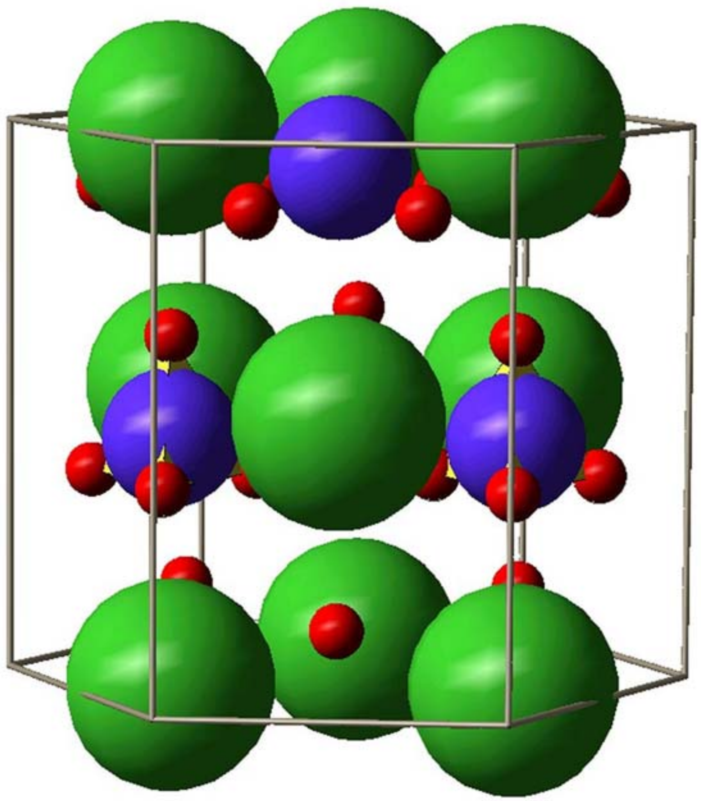
space group: **$P6_3mc$ (#186)**

a = 4.27631(5) Å

c = 6.94844(8) Å

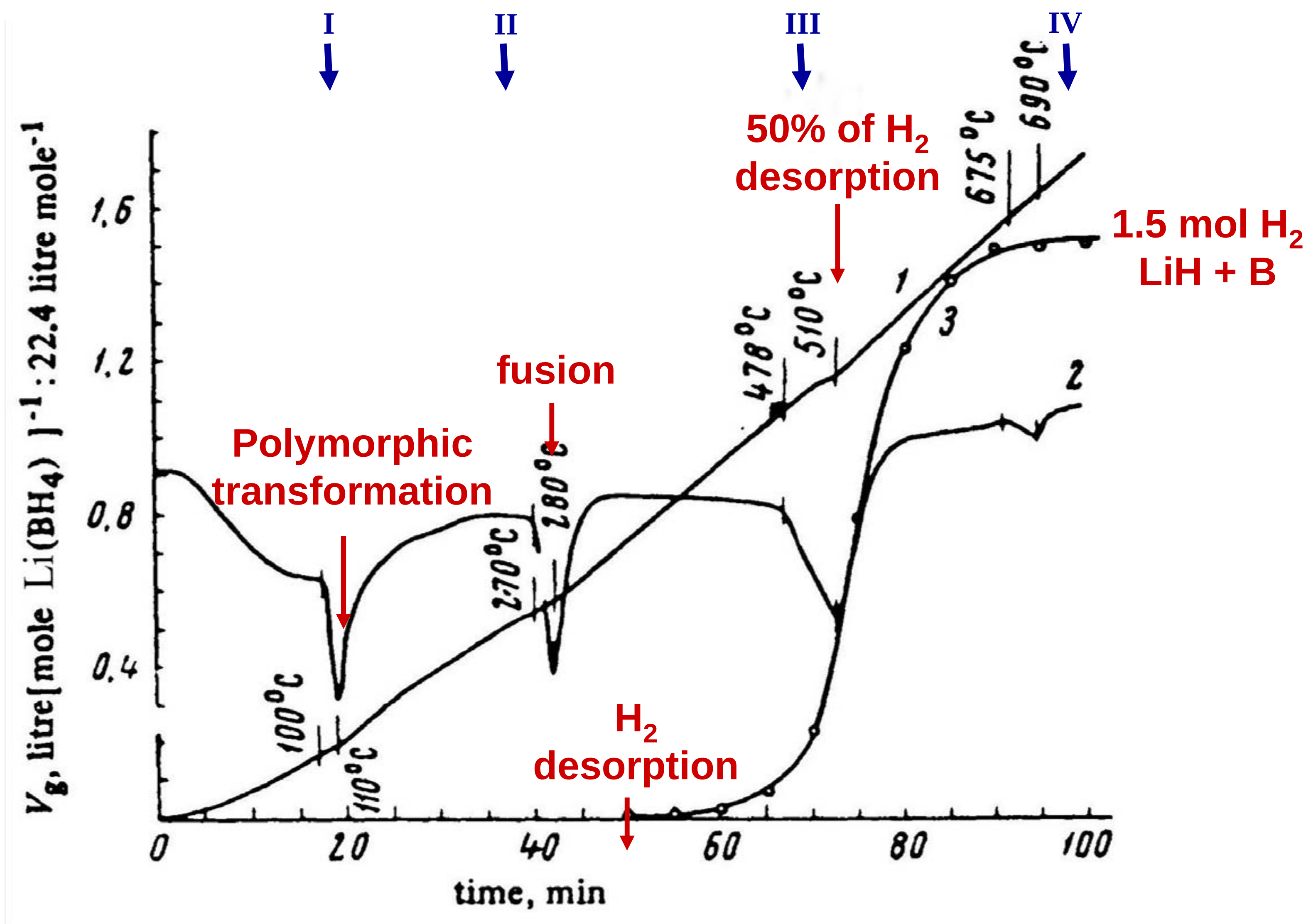
Vol:  110.041 Å$^3$, Z = 2

| Atom | x | y | z |
|---|---|---|---|
| Li | 0.3333 | 0.6666 | 0.0000 |
| B | 0.3333 | 0.6666 | 0.553 |
| $H_1$ | 0.3333 | 0.6666 | 0.370 |
| $H_2$ | 0.172 | 0.344 | 0.624 |



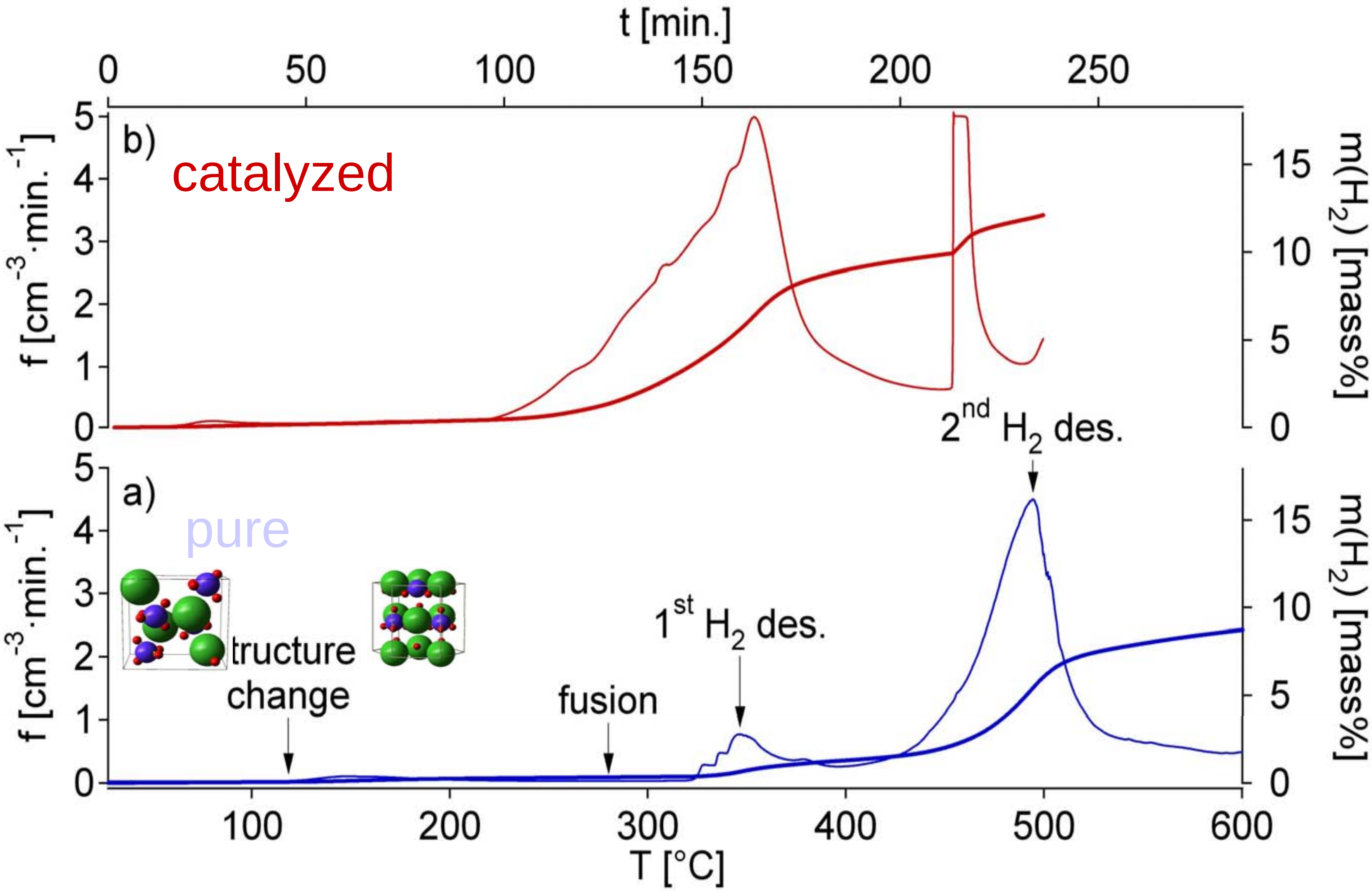
J-Ph. Soulié, G. Renaudin, R. Cerny, K. Yvon, J. Alloys and Comp. 346 (2002), 200

# THERMAL DESORPTION OF $H_2$ FROM OF $LiBH_4$

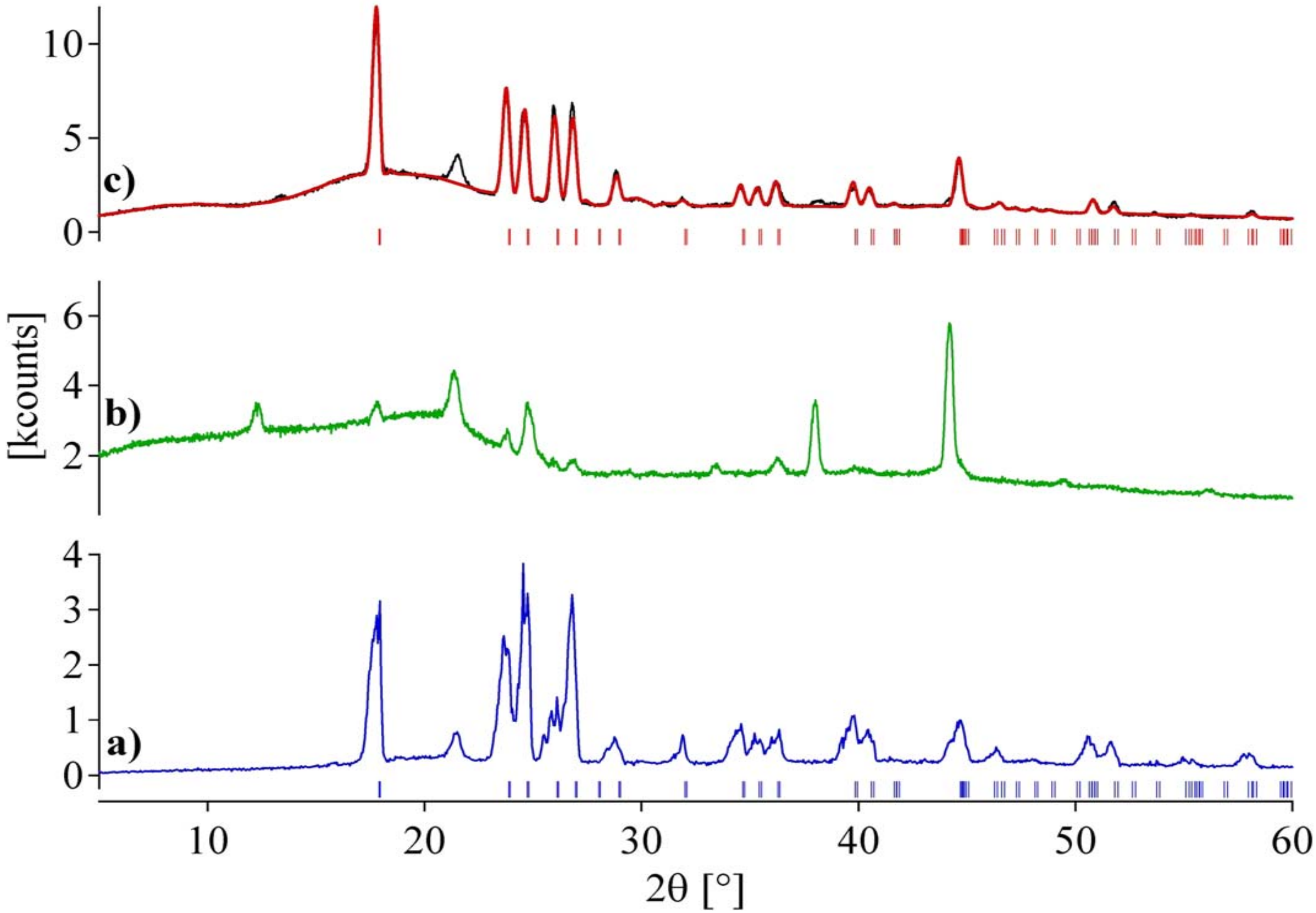


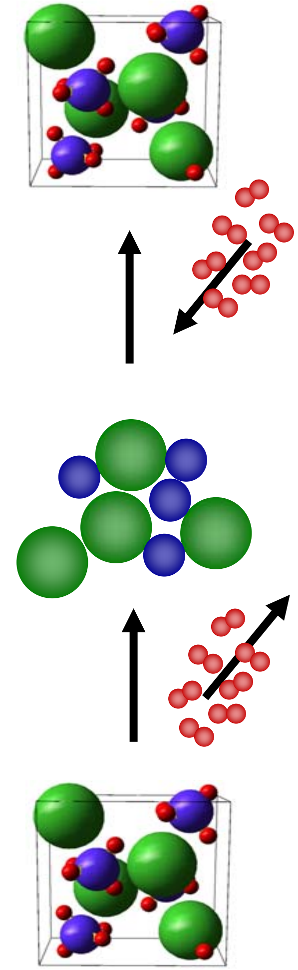

D. S. Stasinevich and G. A. Egorenko, Russian J. Inorg. Chemistry 13 (1968), 341

# THERMAL $H_2$ DESORPTION FROM $LiBH_4$ into vacuum

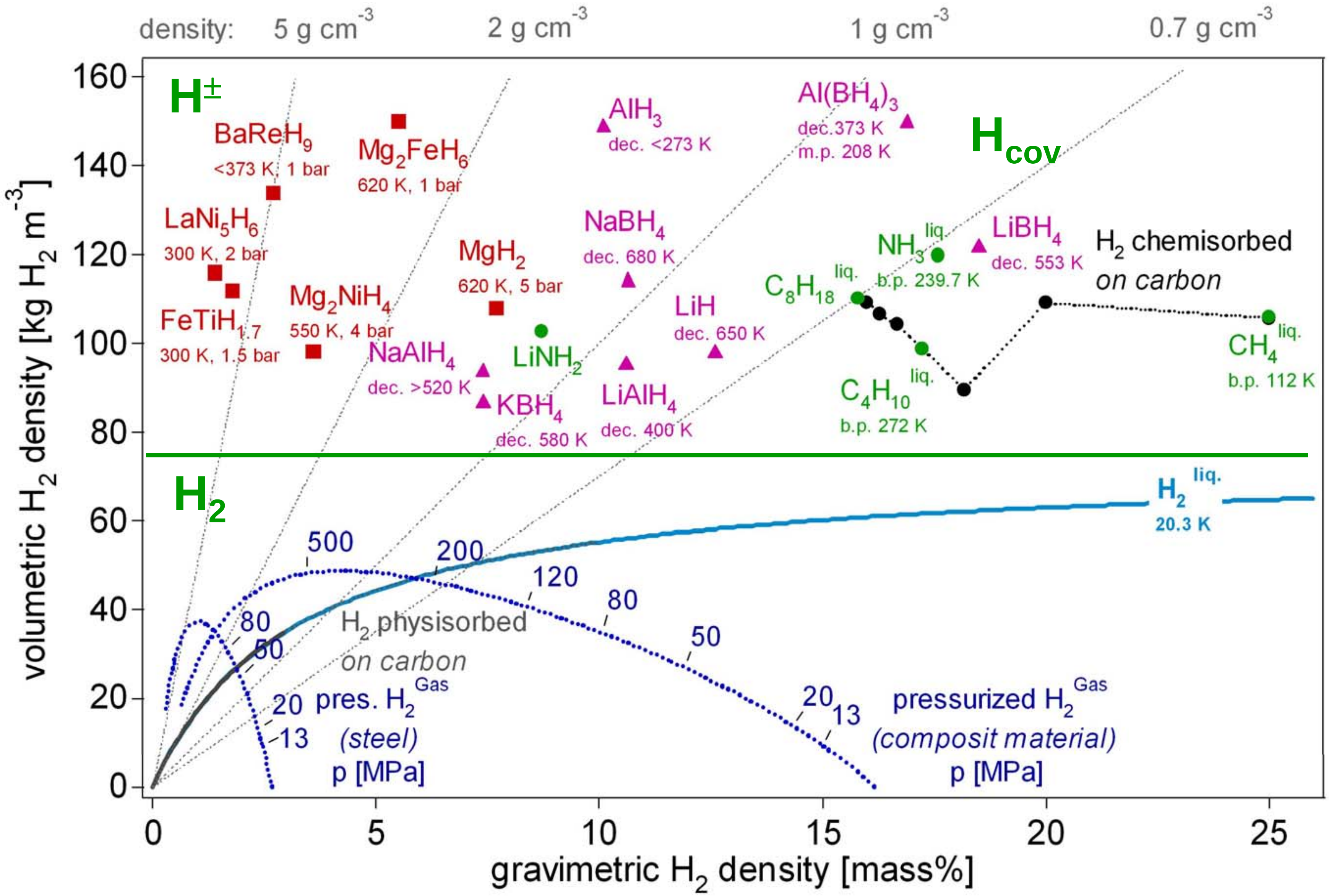


# REVERSIBILITY OF $LiBH_4$



# HYDROGEN DENSITY




Ref: A. Züttel, “Materials for hydrogen storage”, materialstoday, Septemper (2003), pp. 18-27

# Future Developments

- H storage by light weight metal alloys
- Kinetics: nanosized particles (short diffusion path) however: high thermal conductivity
- Thermodynamics: include side reactions which balance $\Delta$ H
- Catalysts to lower reaction temperature (3d, 4d metals with high DOS@$E_F$
- Alkaline and alkaline earth metals and their compounds (borides, alanates,...)
- Adsorption on other nanoporous structures
- *Accept and use more efficient energy technologies*


CAR OF THE YEAR
2005
HYBRID SYNERGY DRIVE
The power to move forward
PRIUS
Mehr Intelligenz, weniger Verbrauch.

# HY-LIGHT ®


- **A purpose designed vehicle**
- Curb Weight: 850 kg
- 4 seats + trunk
- Acceleration 0-100 km/h: < 12 Sek.
- Range (@ 80 km/h const.): 400 km
- Consumption: < 25 kWh/100km compressed $H_2$
- Electrical damping and steering
- Advanced wheel motors 30 kW per wheel
- Fuel storage integrated in the vehicle structure
- Fuel cell stack: 30 kW, based on $H_2$ and $O_2$
- Supercaps: 32-45 kW @ 17 s
- The vehicle participated at the ***Challange Bibendum 2004*** in Shanghai

A collaboration between Michelin and PSI:
PSI contributed the fuel cell system and the supercap-module

# Mobile Future Hydrogen

# F600 HY **Genius**

(Mercedes-Benz Research Vehicle)
Zero emission compact class car
Fuel cell–Li battery–electric motor hybrid car

**Hydrogen Reservoir:**
**4 kg 700 bar**

**Fuel Cell:**
**with air turbocharger**

**Li-ion Battery:**
**30/55 kW**

**Electric Motor:**
**60/85 kW const/peak**
**Torque 250-350 Nm**

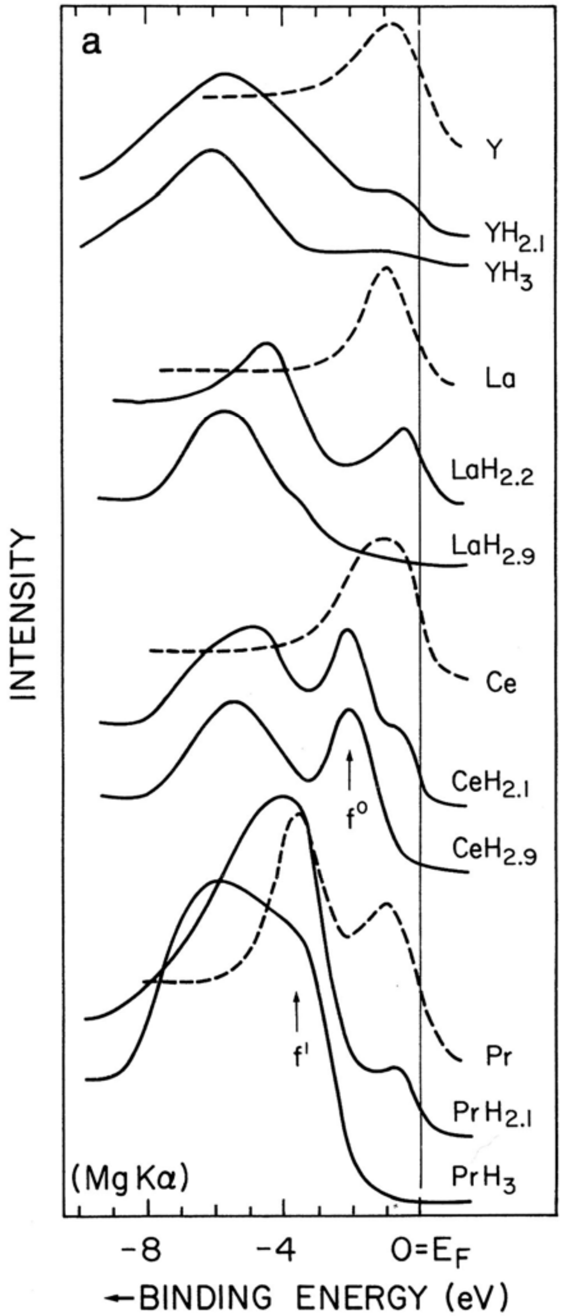
**Range, Consumption:**
**400 km, eq. 2.9 l diesel/100 km**

# Hydrogen interactions with materials

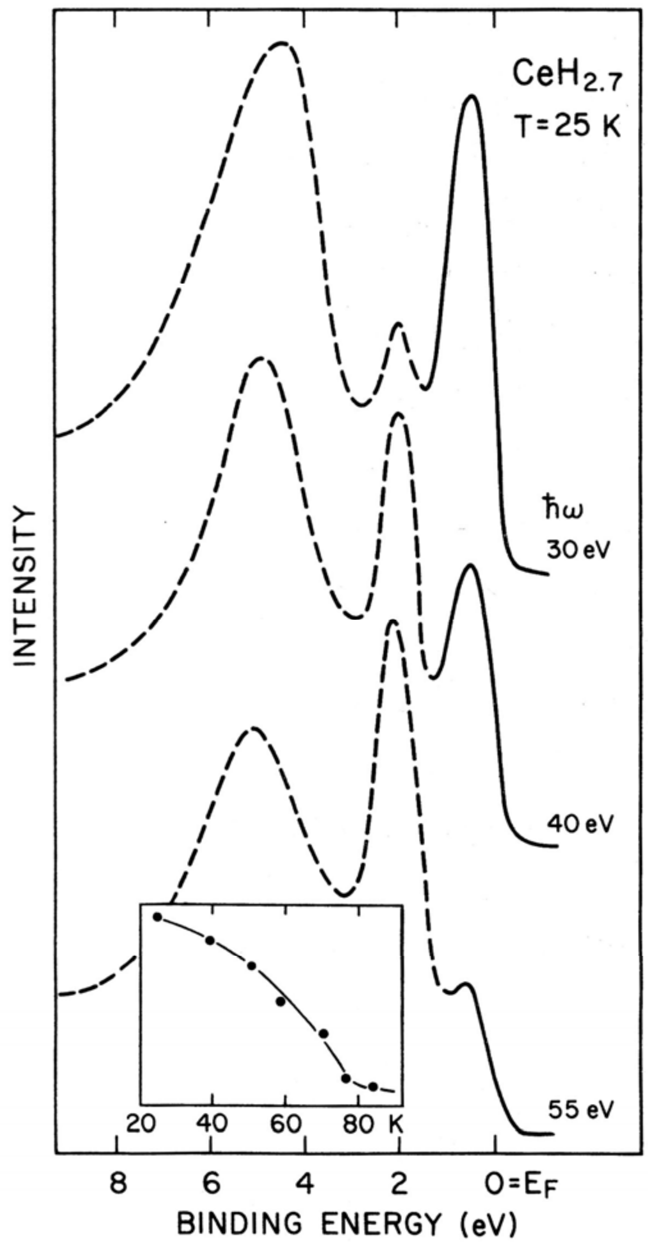
Metal semiconductor transition of Rare Earth hydrides

Valence fluctuation in Yb hydride

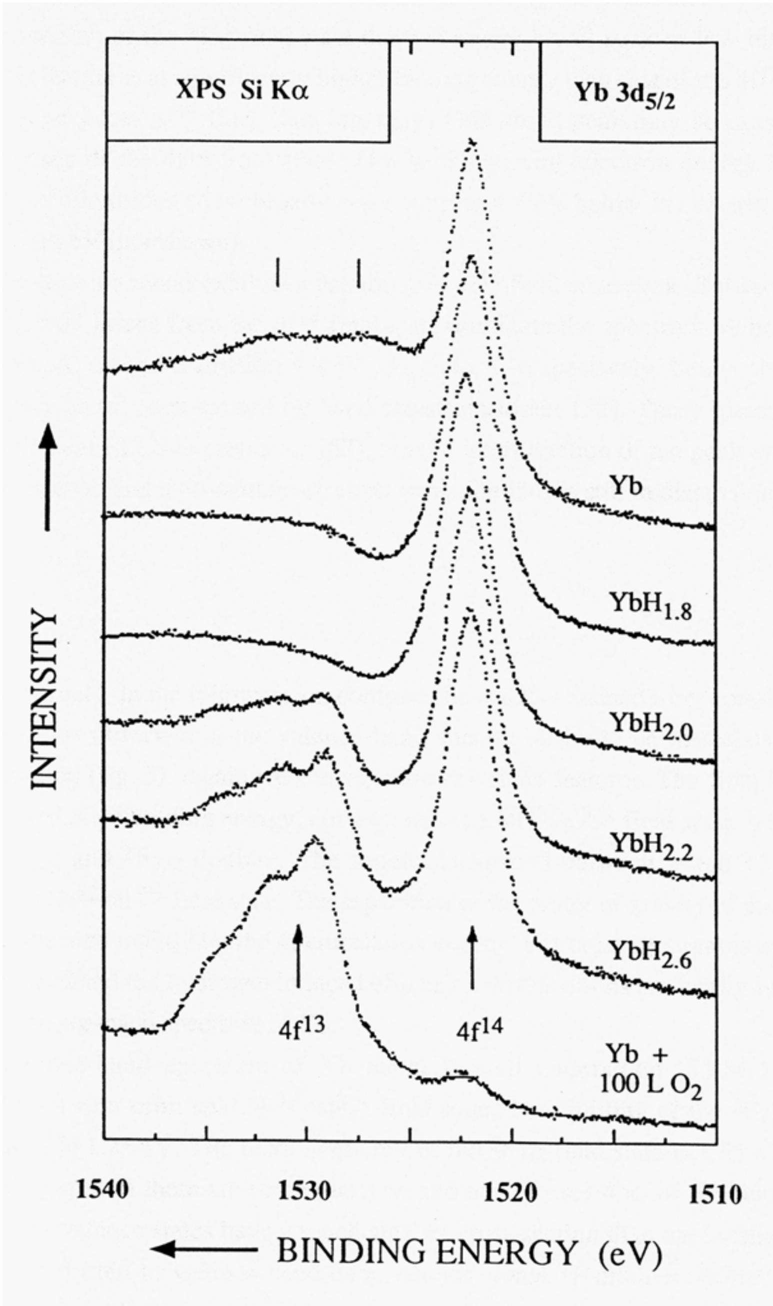
Hydrogen induced defects on graphitic carbon




VB spectra of rare earth hydrides, metal-semiconductor transition




VB spectra of Ce hydride at low temperature

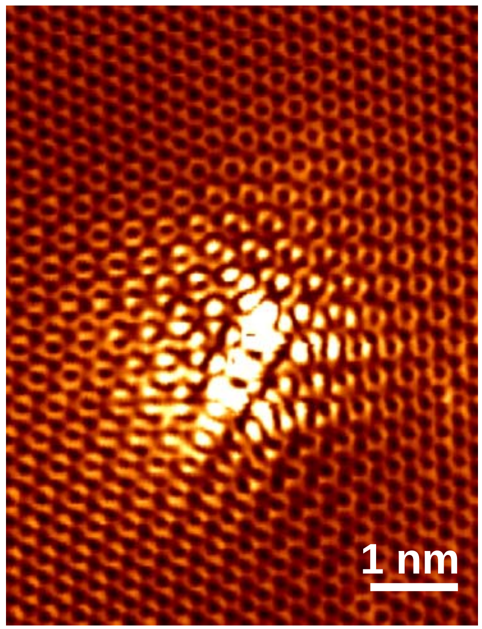



S. Büchler, Diss. ETH No 8785

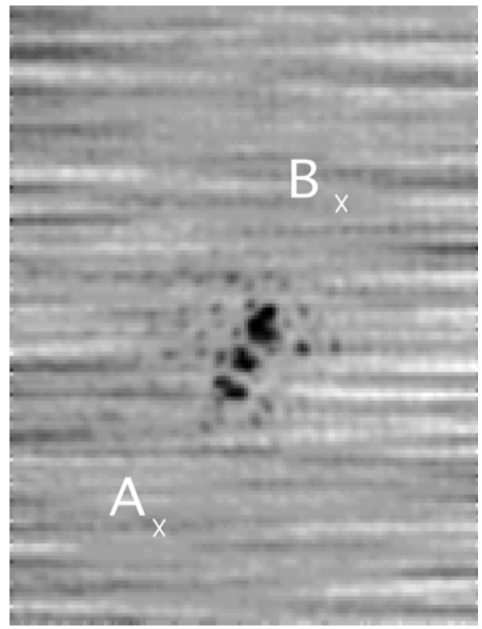
# Individual hydrogen defects on graphite

**Localized structural changes induced by the interaction with hydrogen species lead to long-ranged (~5 nm) electronic effects**

**Current image (STM)**

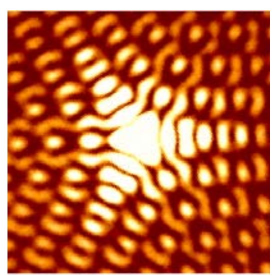



***V = 30 mV***

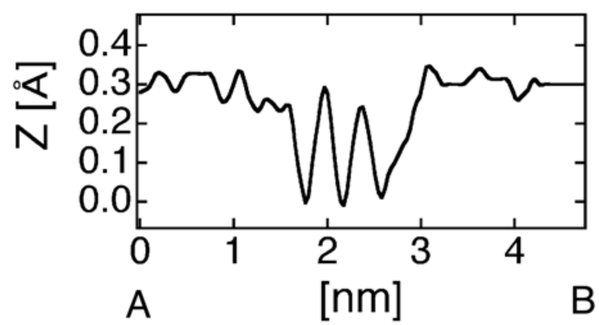



**Topography (AFM)**

**Tight Binding Calculation**



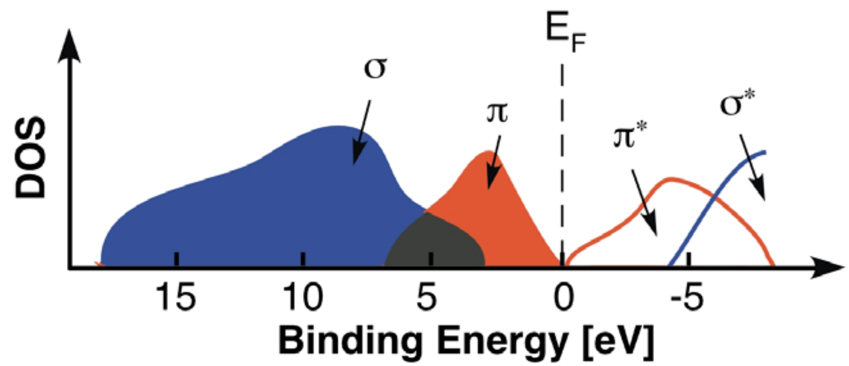
**by M. Franco and F. Zerbatti**
**University of Bologna**




P. Ruffieux et al., Phys. Rev. Lett. 84, 4910 (2000)

# Hydrogen chemisorption on sp$^2$-bonded carbon

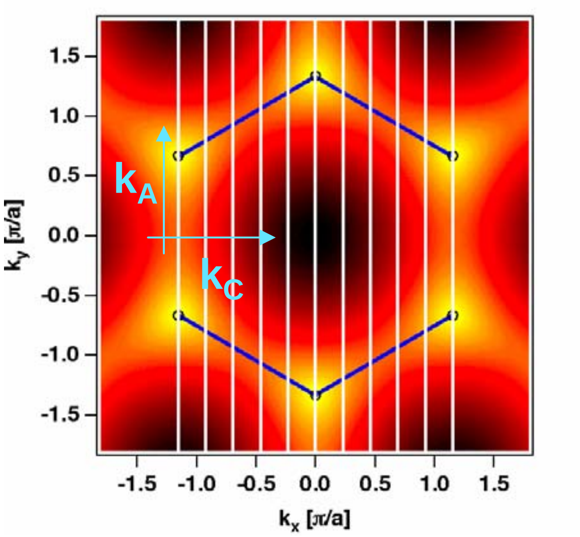
*Local change of the hybridization from sp$^2$ to sp$^3$*

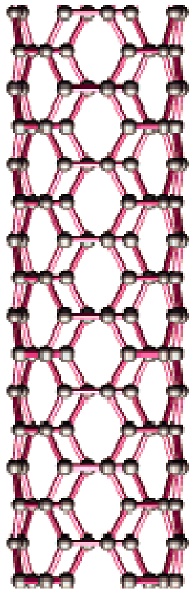



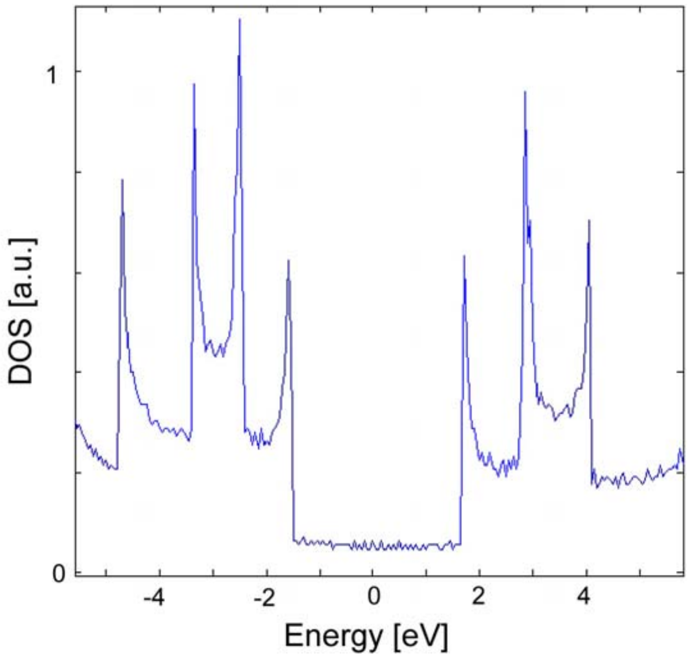
# Electronic structure of SWNT

$$\left|\Delta\vec{k}_C\right| = \frac{2\pi}{\left|\vec{C}_h\right|} >> \left|\Delta\vec{k}_A\right| = \frac{2\pi}{L}$$

*Armchair (5,5) SWNT*






**Armchair (n,n) SWNT are always (in principle) metallic, because the band going through the $\Gamma$-point (origin of the k-space) always goes through a Fermi-point !**

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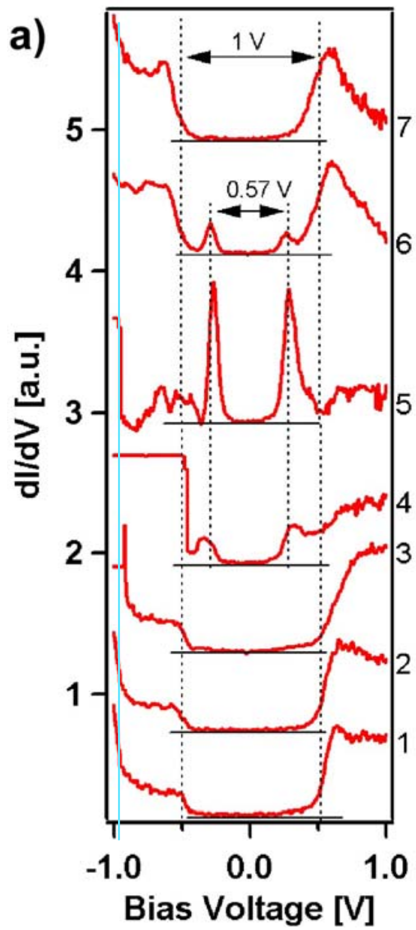
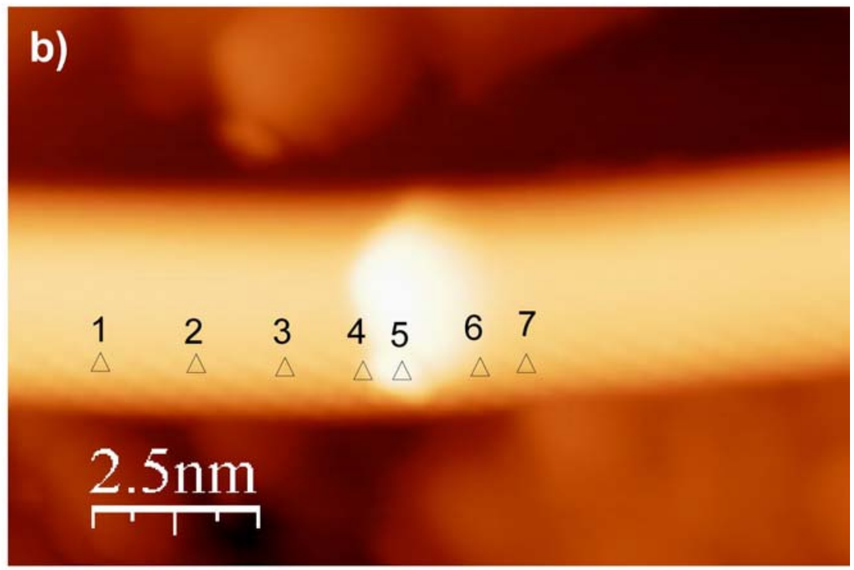
# Local modification of the electronic properties of CNT

## Formation of symmetric states in the gap of semiconducting SWNT




$U_s$=1 V; $I_s$=0.1 nA; $\Delta z$=0.85 nm

LT-STM measurement @ 77 K

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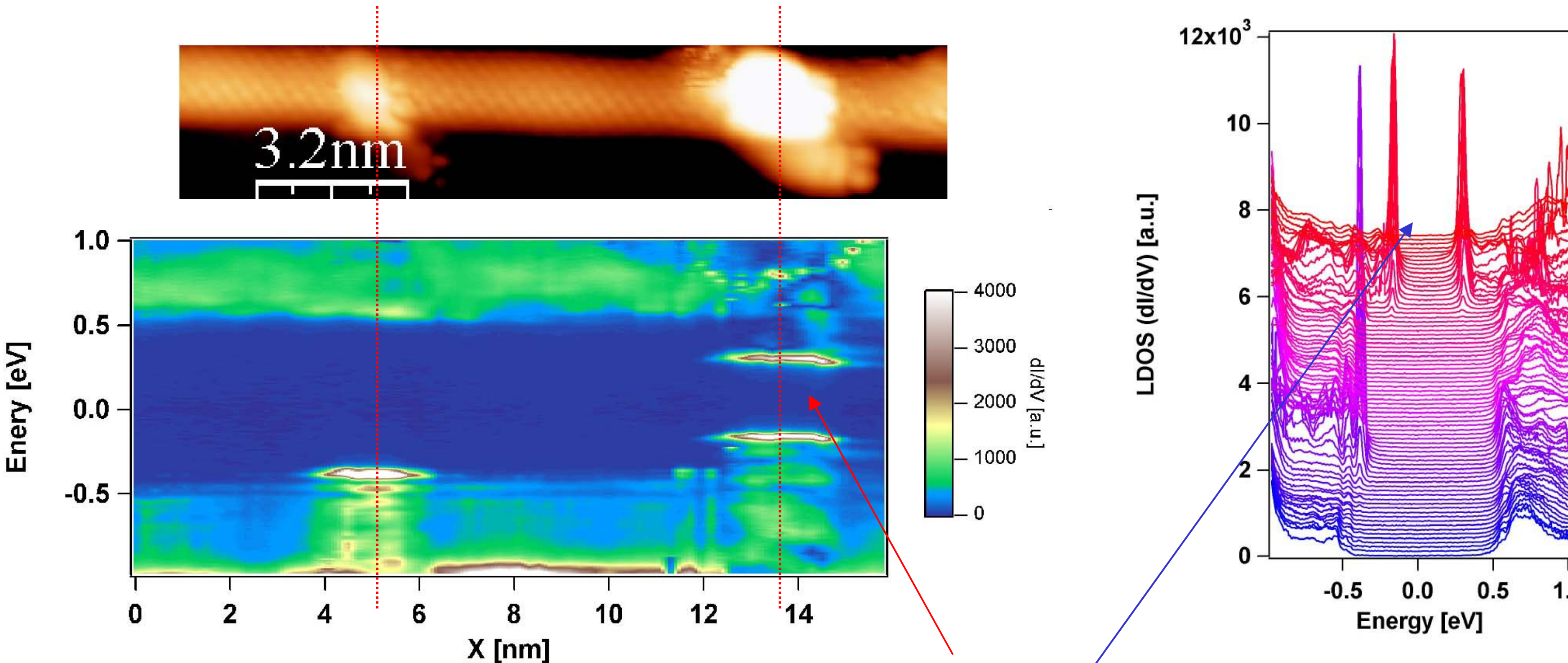
# Local modification of the electronic properties of CNT by hydrogen induced defects: Band Gap Engineering




LT-STM @ 5 K

**Sharp (FWHM < 30 meV) twin state in the bandgap of a semiconducting tube. May be active as radiative recombination centers => Point photon source.**

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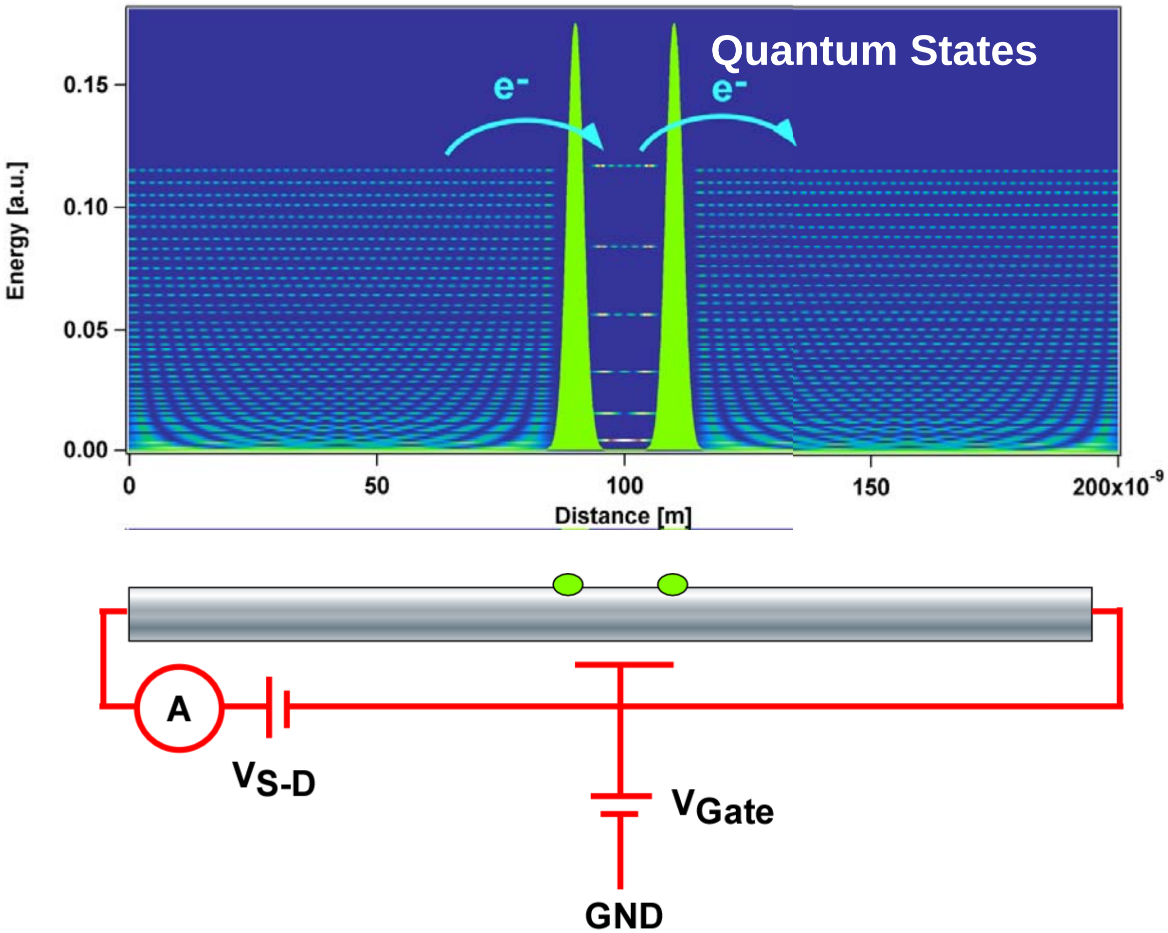
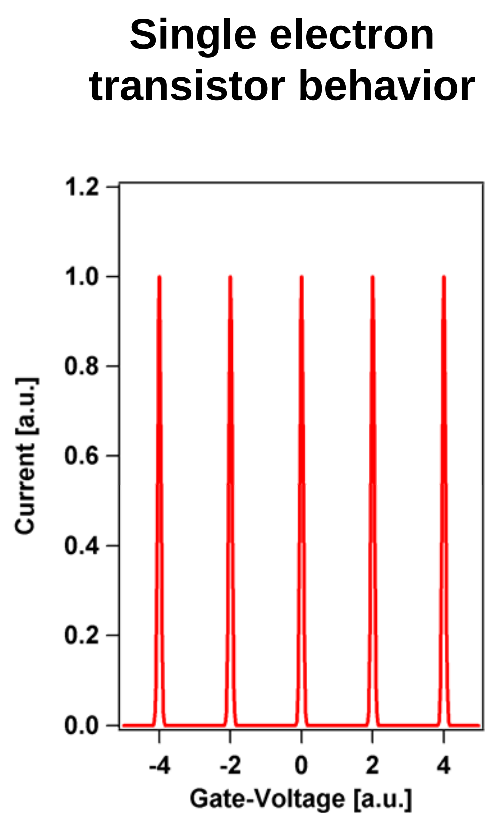
# What is so interesting with defects and SWNT?

***(single walled carbon nanotube SWNT)***

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# Again:
# Is working with hydrogen safe?

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# LAVOISIER

JEAN-PIERRE POIRIER

*Préface de*
**ALAIN PEYREFITTE**
*de l'Académie française*

Pygmalion
Gérard Watelet

## Chronologie des travaux de physique et de chimie de Lavoisier

1765 : Mémoire « Sur l'analyse du gypse ».
1772 : Expériences sur la calcination ; pli cacheté à l'Académie.
1773 : Calcination du plomb et de l'étain dans des cornues.
1774 : *Opuscules physiques et chimiques.*
1777 : Mémoire « Sur la respiration des animaux » ;
mémoire « Sur la combustion en général ».
1780 : Expériences sur les acides.
1781 : Travaux sur la chaleur (en coll. avec Laplace).
1783 : Mémoire « Sur la composition de l'eau » ; « Réflexion sur le phlogistique ».
1785 : Grande expérience de synthèse et analyse de l'eau.
1787 : Mémoire « Sur la nécessité de réformer et de perfectionner la nomenclature chimique ».
1789 : *Traité élémentaire de chimie* ; premier volume des *Annales de chimie.*
1792 : Travaux pour le système métrique.

# ANTOINE LAURENT LAVOISIER,

## FERMIER GÉNÉRAL NÉ A PARIS LE 16 AOUT 1743.

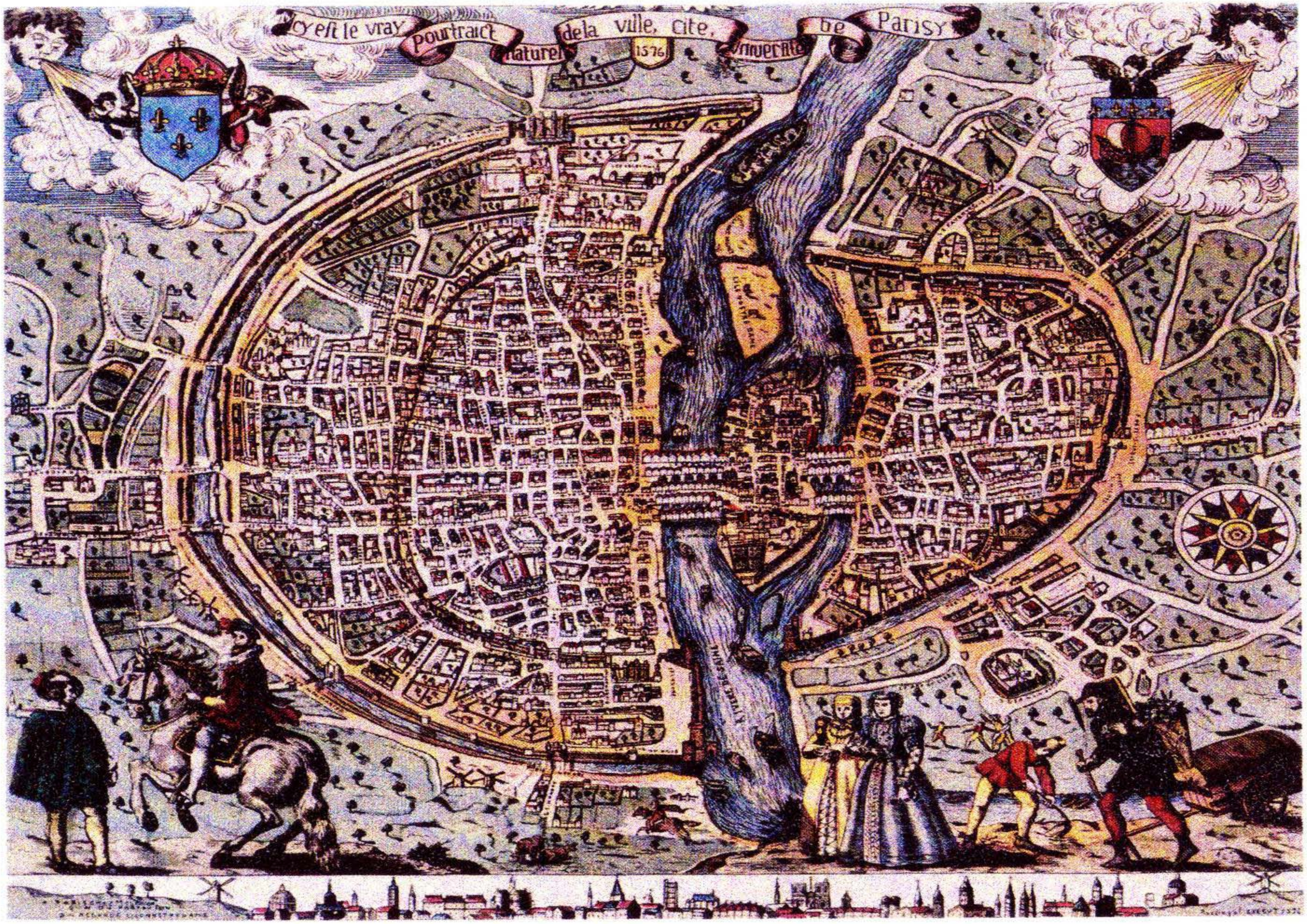
### Jugé le 16 floréal l'an 2.

*Robespierre s'attachoit plus particulièrement à proscrire tous les hommes à qui les talens, les vertus assuroient la vénération, les hommages de leurs contemporains et de la postérité. A ces titres il devoit proscrire Lavoisier. Quel homme fut en effet plus vertueux? quel homme mérita plus des sciences et des arts? toutes les branches des sciences mathématiques et physiques eurent des droits sur ses veilles; et il sembloit destiné à contribuer également au progrès de toutes, lorsqu'une circonstance, telle qu'il ne s'en présente que rarement dans les fastes de l'esprit humain, décida son choix, l'attacha exclusivement à la Chymie, et le conduisit rapidement à l'immortalité. Nous parlons de la découverte si célèbre des Fluides élastiques. Blak, Cavendish, Mackbride et Priestley venoient de faire connoître aux Physiciens un monde nouveau. Lavoisier s'empressa de répéter leurs expériences, de confirmer et d'étendre leurs résultats.*

*Un Chymiste distingué a comparé de la manière suivante Lavoisier et Priestley. « dans les ouvrages de Lavoisier tout est régulier et méthodique, on voit dans ses nombreux mémoires la série d'un immense travail, la même pensée-mère qui plane sur les détails, les lie, et les rapporte à un centre commun: dans Priestley une foule d'expériences, de découvertes, s'offre de toutes parts; vous êtes étonné par le nombre et la diversité des faits nouveaux, mais en même tems frappé de leur incohérence, de leur opposition, de leur contradiction. »*

*Traduit au tribunal révolutionnaire, Lavoisier demanda à ses juges, ou plutôt à ses bourreaux, quinze jours pour terminer des expériences nécessaires à un travail dont il s'occupoit depuis plusieurs années. Le féroce Coffinhal qui présidoit le tribunal, fit cette réponse barbare. « la République n'a pas besoin de Savans, ni de Chymistes. » Ainsi Lavoisier fut confondu dans la fournée des fermiers généraux. Il monta à l'échafaud d'un pas ferme; et mourut, comme il avoit vécu, en philosophe, en sage.*

* Lavoisier arrêté dans son laboratoire, par le Comité Révolutionnaire




Mittelalterlicher Stadtplan von Paris.

# Remaining Challenges e.g. Mobility

Goal: Safe, comfortable and fast transport of 1 or a few persons

2.5 ton vehicle as mid-size car (US DOE)?

Introduce ***mass ratio „brain to car“*** as a quality criteria?
Or „**mass $CO_2$/km**“

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**Incremental increase of storage capacity of 2.5 ton cars or new concepts for efficient cars?**


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Zero emission compact class car
Fuel cell–Li battery–electric motor hybrid car

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**Electric Motor:**
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**Torque 250-350 Nm**

**Range, Consumption:**
**400 km, eq. 2.9 l diesel/100 km**

**Availability of energy**
**is a product of nature,**
**science, technology,**
**culture**
**not of economics, nor**
**politics or war**

**Ethical behaviour**
**respects energy and its**
**efficient use**

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