Hydrogen for Automobiles

The key issues to an efficient H energy infrastructure
Why Hydrogen Storage?

What are the problems?

• Mediocre volumetric energy density
• Do we have a hydrogen mine?
• Gaseous under most circumstances
Typical H Storage Means

- High pressure
- Cryogenic
- Chemical Hydrides
- Metal Hydrides
- Physical Sorption
Basic H Storage Requirements

- H mass percentage (~6% wt at least)
- Volumetric density (~0.15 kg/liter at least)
- Low cost
- Ease of recharge or regeneration
- Fast release, fast recharge
- Environmentally sound
High Pressure H Storage

- 3000, 5000, 7000 psi, maybe up to 10000
- Gravimetric density up to 3%-wt H
- Volumetric density ~ 0.06 kg/liter
- Cost high for bottles > 7000 psi
- Environmentally sound
- But how about safety? it’s like a bomb!
- Relative ease of refueling though taking time
- Composite construction with metal liner
64.9 kg composite usage in the 1st hybrid vessel vs. 76.0 kg in the baseline tank (FW alone)

- The end-user H₂ storage system weight efficiency = 1.67 kWh/kg vs. 1.50 kWh/kg in the system with the baseline tank
- The end-user H₂ storage system cost efficiency:
  - $11/lb CF  Baseline $23.45  Fully Integrated $21.91  Fully Separate $21.75
  - $6/lb CF  Baseline $18.74  Fully Integrated $17.79  Fully Separate $17.63
Approach: Advanced Fiber Placement- Boeing

- **Advanced Fiber Placement**: A CNC process that adds multiple strips of composite material on demand.
  - Maximum weight efficiency - places material where needed
  - Fiber steering allows greater design flexibility
  - Process is scalable to hydrogen storage tanks
  - Optimize plies on the dome sections with minimal limitation on fiber angle
  - Reinforce dome without adding weight to cylinder
Strength

- Tank preparation and validation test

Representative smallest polar opening that the AFP process can currently make

The localized reinforcement protected the dome regions very well

- Static Burst Result: 23420 PSI > 22804 PSI, EN standard (New European Standard superseding EIHP)
- 64.9 kg composite usage in the 1st hybrid vessel vs. 76 kg in the baseline tank (FW alone)

11.1 kg (14.6%) Savings!
Cryogenic H Storage

-252.87°C!

- Very energy consuming to cool
- Energy consuming to maintain
- Gravimetric density up to 8~9%
- Volumetric density ~ 0.08 kg/liter
- Cost high
- Environmentally sound and safe
- Relative ease of refueling
- Vacuum Dewar
Relevance: High density cryogenic hydrogen enables compact, lightweight, and cost effective storage

- **Cost effective:** Cryogenic vessels use 2-4x less carbon fiber, reducing costs sharply at higher capacity

- **Compact:** 235 L system holds 151 L fuel (10.3-10.7 kg H₂)
Relevance: Cryogenic pressure vessels can exceed 2015 H₂ storage targets and approach ultimate
Approach: reduce/eliminate H₂ venting losses by researching vacuum stability, insulation, and para-ortho conversion

- Determine para-ortho effect on pressurization and venting losses
- Directly measure para-ortho populations
- Determine vessel heat transfer mechanism (radiation vs. conduction)
- Evaluate vacuum stability by measuring pressure vessel outgassing
- Test ultra thin insulation for improved vessel volume performance
- Improve vessel design based on experimental results
Hydrogen has two nuclear spin states: para-H\(_2\) (stable at 20 K) and ortho-H\(_2\)

- para-H\(_2\) stable at 20 K
- para-H\(_2\) converts to ortho-H\(_2\) when heated
- Normal H\(_2\) (25% para, 75% ortho) is stable at 300 K
Para-ortho conversion absorbs energy & increases dormancy (equivalent to a second evaporation)

\[ \Delta U = 700 \text{ kJ/kg} \]

\[ \Delta U = 452 \text{ kJ/kg} \]
Chemical Hydrides

- Examples: NH3, N2H4, B2H6, NaBH4...
- Gravimetric density up to 20%-wt (LiBH4)
- Volumetric density up to 0.2 kg/liter
- Many are safe and sound, but not always
- Cost high except NH3 and hydrocarbons
- Regeneration has been problematic
- Utilization is less straightforward than H2.
Chemical Hydrides: Examples

- Hydrocarbons: CH4, C2H6… (complicated reforming → H2, dirty byproducts)
- NH3 (Ammonia) N2H4 (hydrazine) (toxic and … it stinks)
- B2H6 (diborane) (highly toxic)
- Borohydrides (LiBH4, NaBH4…) (relatively safe)
- Alanates (NaAlH4…) (highly reactive)
Chemical Hydrides: Borohydrides

- LiBH4, very high H content, but not soluble
- NaBH4, 12%-wt H dry
- NaBH4, can be made to 30% H2O solution
- NaBH4, 6%-wt H in 35% H2O stabilized with ammonium hydroxide
- Safe, low toxicity
- Still a challenge in regeneration
2009 Progress & Accomplishments

Status at 2009 AMR Review

Material capacity must exceed system targets

Observed H₂ Capacity, weight %

metal hydrides

Mg(BH₄)₂(NH₃)₂
Ca(BH₄)₂
Mg(BH₄)₂
LiBH₄/Ca
MgH₂
LiH₂/MgH₂
Mg-Li-B-N-H
PCN-12
C aerogel
carbide-derived C
B/C
MOF-74
bridged cat./MRI MOF-8
MD C-foam
bridged cat./AX21
Ti-MOF-16
M-doped CA
PANI
NaBH₄
M-B-N-H
LiBH₄/MgH₂
LiBH₄/CA
Li₃AlH₆/Mg(NH₂)₂
Ca(BH₄)₂
MgH₂
LiH₂/MgH₂
M-B-N-H

sorbents

IRMOF-177
PCN-12
C aerogel
carbide-derived C
B/C
MOF-74
bridged cat./IRMOF-8
MD C-foam
bridged cat./AX21
Ti-MOF-16
M-doped CA
PANI
NaBH₄
M-B-N-H
LiBH₄/MgH₂
LiBH₄/CA
Li₃AlH₆/Mg(NH₂)₂
Ca(BH₄)₂
MgH₂
LiH₂/MgH₂
M-B-N-H

2015

H₂ sorption temperature (°C)

Temperature for observed H₂ release (°C)

NPHE 470 H Sys. & Fuel Cells
Observed H₂ Capacity, weight %

- Mg(BH₄)(AlH₄)
- solid AB (NH₃BH₃)
- MD C-foam
- CsC₂₄
- Ti-MOF-16
- Na₂Zr(BH₄)₆
- PANI
- AB ionic liq.
- Bridged cat/AX21
- Mg(BH₄)₂(NH₃)₂
- BC8
- sorbents
- LiBH₄/CA
- MPK/PI-6
- PCN-6
- IRMOF-177
- AC (AX-21)
- PCN-12
- C aerogel
- carbide-derived C
- B/C
- MOF-74
- AB/IL (20% bminCl)
- Li-AB
- AB/Cat.
- AB/LiNH₂
- AB/AT/PS soln
- KAB
- LiAB
- AB/AT/PS soln
- DADB
- solid AB (NH₃BH₃)

Metal hydrides:
- Mg(BH₄)₂(NH₃)₂
- Mg(BH₄)₂
- MgH₂
- Mg(Li-B-N-H)
- Mg-Li-B-H
- LiBH₄/MgH₂
- LiBH₄/CA
- LiBH₄/Mg₂NiH₆
- Ca(BH₄)₂
- LiBH₄/Mg(NH₃)₂
- Mg₂NiH₆

Chemical hydrides:
- AB/LiNH₂
- LiBH₄/MgH₂
- M-B-N-H
- LiMgN
- Li₃AlH₆/Mg(NH₂)₂
- 1,6 naphthyridine
- LiBH₄/Mg₂NiH₆
- Ca(BH₄)₂
- MgH₂
- Mg(Li-B-N-H)

Ultimate
- LiMn(BH₄)₃
- NaMn(BH₄)₄
- NaAlH₄
- NaBH₄
- PANI

Material capacity must exceed system targets

H₂ sorption temperature (°C) vs. Temperature for observed H₂ release (°C)

2010 Progress & Accomplishments
Metal Hydrides

Simple metal hydrides

- Examples: NiH, PdH, LaNi$_5$H$_6$, MgH$_2$
- Metallic bond, H share mobile electrons with the metal atom
- Hydrogen mobility is generally high
- Gravimetric density from 1% ~ 8%
- Metal hydrides with lower H-content tend to have better reversibility
Simple Metal Hydrides: Classification

- $\text{AB}_5$  - LaNi$_5$H$_6$
- $\text{AB}_2$  - ZnMn$_2$H$_3$
- $\text{AB}$  - TiFeH$_2$
- $\text{A}_2\text{B}$  - Mg$_2$NiH$_4$
- Solid solution type  - V$_{0.8}$Ti$_{0.2}$
- MgH$_2$ class (alkaline earth metal hydride)
Metal Hydrides: Isotherm

The isotherm tells us the working temperature and pressure of the hydride and how much H it can store.
Metal Hydrides: LaNi$_5$H$_6$

- Most widely utilized MH today
- Gravimetric density $\sim 1.3\%$-wt H
- Volumetric density $\sim 0.1$ kg/liter
- Cost high due to nickel, lanthanum (rare earth)
- Relative ease of refueling (near ambient pressure)
- It’s the most representative AB$_5$ alloy
- Can be utilized in electrochemical cells (batteries and fuel cells) directly
The chemical elements

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**Lanthanide series**

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**Alkaline metals**

- Lithium (Li)
- Sodium (Na)
- Potassium (K)
- Rubidium (Rb)
- Caesium (Cs)
- francium (Fr)

**Alkaline Earth**

- Calcium (Ca)
- Strontium (Sr)
- Barium (Ba)
- Lanthanum (La)
- Cerium (Ce)
- Praseodymium (Pr)
- Neodymium (Nd)
- Promethium (Pm)
- Samarium (Sm)
- Europium (Eu)
- Gadolinium (Gd)
- Terbium (Tb)

**Rare Earth**

- Lanthanum (La)
- Cerium (Ce)
- Praseodymium (Pr)
- Neodymium (Nd)
- Promethium (Pm)
- Samarium (Sm)
- Europium (Eu)
- Gadolinium (Gd)
- Terbium (Tb)
- Dysprosium (Dy)
- Holmium (Ho)
- Erbium (Er)
- Thulium (Tm)
- Ytterbium (Yb)
LaNi$_5$H$_6$: Structure
Metal Hydrides: MgH$_2$

- Gravimetric density ~ 8%-wt H
- Volumetric density >> 0.1 kg/liter
- Cost is low, very affordable
- Abundant element
- Clean
- Medium temperature absorption and desorption ~ 300 degrees C
- It's the most representative alkaline earth metal hydride
- Not ideal for mobile H storage but ideal for stationary type applications
MgH$_2$: Structure
MgH$_2$: Isotherm

**Graph Description:**
- The graph shows the dependence of hydrogen pressure ratio $p_H/p_0$ on hydrogen concentration $w_H$.
- Data points represent Mg (2% Ni) with different absolute temperatures:
  - Abs.: 653 K
  - Abs.: 623 K
  - Abs.: 593 K
  - Abs.: 573 K
  - Abs.: 547 K
- The pressure $p_0$ is 0.1 MPa.

**Axes:**
- **Y-axis:** Hydrogen pressure ratio $p_H/p_0$.
- **X-axis:** Hydrogen concentration $w_H$ in %.

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**NPTE 470 H Sys. & Fuel Cells**
MgH$_2$: Kinetics

- Absorption and release is slow.
- ~ a few hours for a typical Ab/De-sorption cycle.
- Fast enough for stationary storage of renewable energy nevertheless.
- Can be expedited with innovative heating.
- For example inductive heating.
MgH$_2$: Fast release with induction Heating

- **Hydride Storage**
- **Fuel Cell**
- **Pressure Control**
- **Induction Coil**
- **Power Source ~ 40 kHz**
- **Electrically and Thermally Insulating Container**
- **Hydrogen In & Out**

NPREG 470 H Sys. & Fuel Cells


MgH₂: Fast release with induction heating

Fuel cell performance with and without induction heating
MgH₂: Fast release with induction heating

Fast fuel cell ramping with induction heating
Complex metal hydrides

The hydrogen bonding is more covalent or localized

• Examples: Ca(BH$_4$)$_2$, Mg(BH$_4$)$_2$, LiNH$_2$, LiAlH$_4$

• New development

• Many issues exist, like regeneration, volatiles, safeties
Final Year Downselection Path

Materials examined in final year of the MHCoE

11 More Downselects (Removing from Study)

- CaB_{12}H_{12}/CaH_2 (not reversible)
- Li_2B_{12}H_{12}/6MgH_2 (too high T_{des})
- Ti(BH_4)_3 (not reversible)
- Li_3AlH_6/2LiBH_4 (too high T_{des})
- Li(NH_3)_xB_12H_{12} (NH_3 release)
- NaBP_2H_8 (not reversible)

- 4LiBH_4/Mg_2NiH_4 (low wt. %)
- Mg(B_3H_8)_2 (too unstable)
- Li_2B_{12}H_{12}/2CaH_2 (too high T_{des})
- Mg(NH_3)_xB_10H_{10} (NH_3 release)
- Mg(NH_3)_6B_12H_{12} (NH_3 release)
Physical/Chemical Sorption

• Basically utilize the relatively weak forces: Van Der Waals force, hydrogen bonding…
• Sometimes the sorption could also have a chemical nature.
• Examples: activated carbon, zeolite, MOF (metal organic framework), COF (covalent organic framework), nanotubes…
MOF

- **One of best known MOF 177:**
  \[ \text{Zn}_4\text{O(BTB)}_2, \text{ where } \text{BTB}^3- = 1,3,5\text{-benzenetribenzoate} \]
  
  Theoretical gravimetric density
  - 7.1 wt% at 77 K, 40 bar
  - 11.4 wt% at 77 K, 78 bar
  
(not including dewar and pressure vessel)
MOF 177

Bridged MOF-177

Pt/AC - MOF-177

MOF-177

P2 AUGERDEG (W T%) vs Pressure (MPa)

NPRE 470 H Sys. & Fuel Cells
Physical/Chemical Sorption

Some remarks

• MOF still not matching the AB$_5$ metal hydride in gravimetric density
• Generally poor volumetric density (puffy material)
• Cycling and cycle life?
• Good with cryogenic means
New energy cars

- Electric (hybrid) cars (80Wh/kg)
- Natural gas cars (>800Wh/kg)
- Fuel cell cars with H (stored in various forms)
  (compressed H > 500Wh/kg)
- Others...

The problem?
1. Energy density
2. Cost
Battery cars

Nissan Leaf has a 24-kWh EPA range of 73 miles

(CNN) -- President Barack Obama's goal of putting 1 million electric cars on U.S. roads by 2015 could run into a huge roadblock -- the American consumer.
Battery cars

Are they really clean or green?

A bit of inconvenient truth?

Coal fired Power station $\rightarrow$ Electric grids $\rightarrow$ charger $\rightarrow$ battery $\rightarrow$ Wheel

Battery cars have an overall efficiency only 30.2%!!

Compared to the gasoline engine cars of $\sim$30%

And what is cleaner? Coal vs Oil

NPRE 470 H Sys. & Fuel Cells
Battery cars

Some remarks

• Unless there is a major breakthrough in batteries, say doubling the current energy density, battery cars will be a niche.

• Put in perspective, battery chemistry improves from the 1859 Plante lead acid cell (40Whr/kg) to today's lithium ion (80Whr/kg). It doubled in 150 years!
Natural gas cars

The rationale

- NG is 1/3 the price of gasoline equivalent
- It at least triples the range of a battery for less than ½ of the added weight compared to a Li-ion battery car
- For 2 thousand dollars you can modify your car to burn NG, with a range of 70+ miles, bettering that of Chevy Volt!
Natural gas cars

- Most cars can be converted to burn NG + gasoline
- The NG is good enough to daily commute
Natural gas cars

• Then you can charge it overnight at your home
• $2000 conversion vs plug-in hybrids (PHEV) of $12000 battery
• 1/3 of gasoline operation cost, on par with PHEV or cheaper
• More NG reserve than oil. And NG is going up with shale NG and methane hydrates
• NG twice clean as gasoline, four times as coal
Natural gas cars

- The technology is mature
- Only problem existing is political