Hydrogen Storage Program Area

-Plenary Presentation-

Ned T. Stetson

Fuel Cell Technologies Office

2017 Annual Merit Review and Peer Evaluation Meeting

June 5 - 9, 2017
Goals and Objectives

Objective: Develop H₂ storage technologies with performance to enable fuel cell products to be competitive with conventional technologies.

For Light-Duty Vehicles:
- Comparable driving range
- Similar refueling time (~3 minutes)
- Comparable passenger and cargo space
- Equivalent level of safety

Onboard H₂ storage targets to be reviewed approximately every five years and revised as appropriate.

GOAL: Develop advanced hydrogen storage technologies to enable successful commercialization of hydrogen fuel cell products.
Onboard Storage Target Revisions

Vehicle performance has improved since the 2008/09 target review

- Fuel economy range increase from 48-53 to 49-67 miles per kg H₂
- Autonomie (ANL) available for full vehicle performance analysis

Onboard storage targets are periodically reviewed in terms of current vehicle performance data and revised as appropriate
Revised Onboard H₂ Storage Targets

Revised System Targets for Onboard Hydrogen Storage for Light-Duty Fuel Cell Vehicles

<table>
<thead>
<tr>
<th>Storage Parameter</th>
<th>Units</th>
<th>2020 (previous)</th>
<th>2020 (new)</th>
<th>2025 (new)</th>
<th>Ultimate (previous)</th>
<th>Ultimate (new)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>System Gravimetric Capacity:</strong></td>
<td>kWh/kg (kg H₂/kg system)</td>
<td>1.8 (0.055)</td>
<td>1.5 (0.045)</td>
<td>1.8 (0.055)</td>
<td>2.5 (0.075)</td>
<td>2.2 (0.065)</td>
</tr>
<tr>
<td><strong>System Volumetric Capacity:</strong></td>
<td>kWh/L (kg H₂/L system)</td>
<td>1.3 (0.040)</td>
<td>1.0 (0.030)</td>
<td>1.3 (0.040)</td>
<td>2.3 (0.070)</td>
<td>1.7 (0.050)</td>
</tr>
<tr>
<td><strong>Storage System Cost:</strong></td>
<td>$/kWh net ($/kg H₂)</td>
<td>10 (333)</td>
<td>10 (333)</td>
<td>9 (300)</td>
<td>8 (266)</td>
<td>8 (266)</td>
</tr>
<tr>
<td><strong>Charging / Discharging Rates:</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>System fill time</td>
<td>min</td>
<td>3.3</td>
<td>3-5</td>
<td>3-5</td>
<td>2.5</td>
<td>3-5</td>
</tr>
</tbody>
</table>

System target revisions considered vehicle performance
### New Onboard H₂ Storage Targets

**New System Targets for Onboard H₂ Storage for Light-Duty Fuel Cell Vehicles**

<table>
<thead>
<tr>
<th>Storage Parameter</th>
<th>Units</th>
<th>2020 (new)</th>
<th>2025 (new)</th>
<th>Ultimate (new)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Charging / Discharging Rates:</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average flow rate</td>
<td>(g/s)/kW</td>
<td>0.004</td>
<td>0.004</td>
<td>0.004</td>
<td>New target to differentiate between Average flow rate &amp; Minimum full flow rate</td>
</tr>
<tr>
<td><strong>Dormancy:</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dormancy time target (minimum until first release from initial 95% usable capacity)</td>
<td>Days</td>
<td>7</td>
<td>10</td>
<td>14</td>
<td>New targets to address Dormancy (a challenge for systems that operate below ambient temperate)</td>
</tr>
<tr>
<td>Boil-off loss target (max reduction from initial 95% usable capacity after 30 days)</td>
<td>%</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td></td>
</tr>
</tbody>
</table>

*The full set of onboard H₂ storage targets available online at: [https://energy.gov/node/1315186](https://energy.gov/node/1315186)*
## Current Status vs Targets

<table>
<thead>
<tr>
<th>Storage Targets</th>
<th>Gravimetric kWh/kg (kg H₂/kg system)</th>
<th>Volumetric kWh/L (kg H₂/L system)</th>
<th>Costs 1 $/kWh ($/kg H₂)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2020</td>
<td>1.5 (0.045)</td>
<td>1.0 (0.030)</td>
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<td>2025</td>
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<td>Ultimate</td>
<td>2.2 (0.065)</td>
<td>1.7 (0.050)</td>
<td>$8 ($266)</td>
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</table>

<table>
<thead>
<tr>
<th>Current Status 2</th>
<th></th>
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</thead>
<tbody>
<tr>
<td>700 bar compressed (5.6 kg H₂, Type IV, Single Tank)</td>
<td>1.4 (0.042)</td>
</tr>
</tbody>
</table>


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1 Projected at 500,000 units/year
Objective: Achieve a driving range competitive with conventional vehicles for full span of light-duty vehicles, while meeting packaging, cost, safety, & performance requirements
**FY 2017 Appropriation = $15.6M**

**FY2017 Funding Allocations by Focus Area**

- **Materials R&D**: $9 million
- **Engineering R&D**: $1 million
- **Advanced Tank R&D**: $3 million
- **Analysis**: $2 million

Emphasis is on early phase R&D for H₂ storage materials and lower cost physical storage.

**Number of Projects in Portfolio by Focus Area**
(Includes subs directly funded by DOE)

- Materials: 12 projects
- Engineering: 4 projects
- Advanced Tanks: 6 projects
- Analysis: 24 projects
Physical Storage Activities
Current Status – 700 Bar System Cost Breakout

- **Cost breakdown at 500k systems/yr.**
- System cost is **dominated**, 72%, by **composite materials and processing**
- Carbon Fiber composite cost:
  - ~ 50% Carbon fiber precursor
  - ~ 50% Precursor fiber conversion
- BOP costs are a major cost contributor, especially at low annual production volumes


*Carbon fiber cost reduction is needed to drive down cost of 700 bar storage systems*
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• Precursor development for low-cost, high-strength carbon fiber (CF) for use in composite overwrapped pressure vessel applications
  – Resulting CF to have properties similar to Toray T700S
  – Target cost of $12.60/kg of CF

• Areas of interest:
  – PAN-based fibers formulated with co-monomers and additives that permit lower cost processing to produce the PAN fiber than conventional solution spinning processes, and or that reduce the conversion cost of the PAN-fiber to CF;
  – Polyolefin-based fibers capable of being cost effectively converted into high-strength CF;
  – Novel material precursor fibers that can lead to low-cost, high-strength CF production.
Accomplishments - Project Highlights

Alternative Resin and Manufacturing
[Materia/MSU/Spencer Composites/Hypercomp Engineering]

- Reducing composite volume/mass through use of alternative resin and manufacturing processes
- Improved process cut resin infusion time in half for prototype tanks

Conformable 700 bar H₂ Storage Systems
[CTE/HECR/UT/Stan Sanders]

- Developing conformable 700 bar pressure vessels without use of carbon fiber composites
- Demonstrated vessel with a 34,000 psi burst (2345 bar), exceeding the 2.25 safety margin for 700 bar systems

Addressing cost through reduced carbon fiber use
Accomplishments - Project Highlights

Alternative Materials for BOP [SNL/Hy-Performance Materials]

- Identifying alternative alloys to lower BOP cost and weight through testing and computational material screening
- **Identified alloys with potential to reduce cost and weight by >50% compared to 316L SS baseline**

Insulation for Cryogenic Storage Tanks [Vencore/Aspen Aerogels/Energy Florida/Hexagon Lincoln/IBT/NASA-KSC/SRNL]

- Developing integrated advanced insulation system capable of meeting dormancy requirements for vehicle applications
- **Down-selection of concept technologies in-progress**

Addressing Balance-of-Plant
Institute for Advanced Composites Manufacturing Innovation

- Institute of Manufacturing USA
- Managed by the EERE Advanced Manufacturing Office
- Technology Focus Areas:
  - Vehicles
  - Wind Turbine Blades
  - Compressed Gas Storage Vessels
  - Design, Modeling & Simulation
  - Composite Materials & Processes

Leveraged project: Thermoplastic Composite Compressed Gas Storage Tanks

- Project lead: DuPont
- Partners:
  - Composite Prototyping Center (CPC)
  - Steelhead Composites
  - University of Dayton Research Institute (UDRI)
- Kick-off: FY2017, Q1

Leveraging efforts of the Institute for Advanced Composites Manufacturing Innovation
Materials-Based Storage Activities
HyMARC: Hydrogen Materials – Advanced Research Consortium
Enabling twice the energy density for onboard H₂ storage

- Applied materials development
  - Novel material concepts
  - High-risk, high-reward
- Concept feasibility demonstration
- Advanced development of viable concepts

- Foundational research
- Material development tools
  - Foundational R&D
  - Computational modeling development
  - Synthetic/characterization protocol development
- Guidance to individual projects
- Database development

- Characterization resources
  - “User-facility” for HyMARC projects
- Characterization method development
- Validation activities
  - Validation of Performance
  - Validation of “Theories”
Effective thermal energy for H₂ release:

\[ \Delta E(T) = \Delta H^\circ (T) + E_a \]

**HyMARC** – Understanding the phenomena of hydrogen interactions with materials.

**Task 1**: Thermodynamics
**Task 2**: Transport
**Task 3**: Gas-surface interactions
**Task 4**: Solid-solid interfaces
**Task 5**: Additives and dopants
**Task 6**: Materials informatics

Focusing on overcoming thermodynamic and kinetic barriers simultaneously.
HyMARC – Understanding the phenomena of hydrogen interactions with materials

**Thermodynamics:**
- Mg-B-H, Li-N-H

- LEIS on model metals (Mg, W)
- Oxidation (NaAlH₄, Mg(BH₄)₂)
- H₂ dissociation (Mg, MgB₂)

- Isotropic interstitial diffusion (PdHₓ)
- Anisotropic vacancy diffusion (MgH₂)
- Complex hydride diffusion (Mg(BH₄)₂)

- Borohydride decomposition pathways in MgBₓHᵧ

- Interstitial topotactic interface (Pd/PdHₓ)
- Simple structural transformation (Mg/MgH₂)
- Complex reactive interface (MgBₓHᵧ/MgBnHm)

- TiF₃/TiCl₃
- Ti in NaAlH₄, MgB₂/Mg(BH₄)₂

- MOF-74, CuBTC
- Graphene (doped/functionalized)

**Surfaces/interfacial diffusion**

**Surface reactions**

**Sorption**

**Additives**

**Phase nucleation & evolution**

**Chemical bond activation**

Studying model systems to isolate physical factors and mechanisms

ST127, ST128, ST129, ST130
HyMARC accomplishments – theory capabilities

Improved sorbent isotherms

Recipes for integrating different levels of theory in sorbent isotherm models

Seedling: Chung/PSU

Accurate hydride thermodynamics

Finite-\(T\) free energy, environment- and morphology-dependent thermodynamics

Seedlings: Liu/ANL, Severa/U. Hawaii

Solid mechanics & interfaces in hydrides

Internal and confinement stress effects; reactive diffuse interfaces

Seedlings: Liu/ANL, Severa/U. Hawaii

Kinetic modeling

Semiempirical kinetic modeling and rate analysis; phase evolution kinetics

Seedlings: Liu/ANL, Severa/U. Hawaii

Additional accomplishments in compiling databases and reference libraries (“Task 6”):

- Simulated & measured spectroscopy database (NMR, FTIR, XAS/XES) for identifying \(\text{MgB}_x\text{H}_y\) (preparing manuscript w/LBNL/SNL/HySCORE)
- Library of analytical free energies for Li-N-H (published) and Mg-B-H (preparing manuscript), with validation at a range of pressures via NMR (w/SNL/HySCORE)
- Database of classical potentials for simulating borohydride mixtures and interfaces (w/SNL)

Seedling projects help focus theory method development prioritization
HyMARC accomplishment – understanding role of additives on sorption kinetics

- Investigated model system Ti-doped NaAlH₄ via AP-XPS, LEIS and Auger spectroscopy
  - Detected no Ti species on sample surface before or during desorption, reappears during absorption
  - Disproved models invoking surface Ti during dehydrogenation reaction

Four Al species detected by AP-XPS during dehydrogenation

Data supports proposed zipper mechanism

Proposed mechanisms are evaluated based on experimental data

HyMARC accomplishment – providing support to seedling projects

NREL and NCNR carries out neutron vibrational spectroscopy measurements on LiBH₄ infiltrated mesoporous carbon samples from UMSL

Can nanoconfinement in functionalized porous materials facilitate reversible hydrogen storage reactions?
• NVS show LiBH₄ infiltrated
• Shifting and broadening show there is an effect of confinement
• Degree of N-doping enhances BH₄⁻ orientational mobilities

Accelerating rate of progress in the development of H₂ storage materials
HyMARC accomplishment – Validating hydrogen sorption measurements and reporting

Led an international inter-laboratory volumetric capacity 
H$_2$ adsorption measurement round-robin study

- Promoted valid comparisons of hydrogen-storage materials 
  - necessary to evaluate implementations of protocols
- Decreased irreproducibility due to systematic and “black box” errors 
  - NREL gives direct feedback on data
- Determining a “natural” spread of data from instrument and operator variables

Promoting standard protocols for performing and reporting sorption measurements
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Errors Corrected from > 400% to < 5% spread

Promoting standard protocols for performing and reporting sorption measurements
Accomplishments: Lab Team Publications

32: Publications published or submitted for publication
4: Patents applications submitted
7: Manuscripts in preparation as of April 2017
2: Selected as cover features


The lab teams are producing high-value R&D and disseminating it to the R&D community.
Accomplishments - HyMARC Project Highlights

Surface functionalized mesoporous carbons [HyMARC seedling—UMSL]

- Demonstrating ability of functionalized mesoporous carbons to facilitate reversible H₂ sorption reactions of hydride materials
- Prepared N-doped carbons and demonstrated infiltration of Al and B-based materials

Electrolyte Assisted Storage Reactions [HyMARC seedling—Liox Power]

- Improving reaction kinetics through use of electrolytes to facilitate atomic rearrangement and diffusion
- Have carried out initial screening studies of possible electrolytes

Accelerating development of improved hydrogen storage materials
Accomplishments - HyMARC Project Highlights

“Graphene-wrapped” hydrides [HyMARC seedling—ANL]
- Encapsulating nanoparticles of complex hydrides with graphene to enhance reversibility and kinetics
- Demonstrated 9 wt% uptake in NaBH₄ systems with 80% regenerable release over 6 cycles

SEM of NaBH₄ nanoparticles wrapped in graphene

Magnesium boride etherates [HyMARC seedling—U. Hawaii]
- Improve reversibility of Mg(BH₄)₂ through formation of MgB₂-etherates
- Demonstrated the formation of significant amounts of β-Mg(BH₄)₂ at 300 °C

TGA of hydrogenated ball milled MgB₂-THF

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Accelerating development of improved hydrogen storage materials
Accomplishments - HyMARC Project Highlights

**Novel boron-containing polymers [HyMARC seedling—Penn State]**
- Developing novel boron containing porous polymers with higher H₂ binding energy
- Designed and synthesized two new classes of microporous polymers that contain boron.

**High-capacity Hydrogen Storage Materials via Mechanochemistry [Ames Laboratory]**
- Prepare high hydrogen capacity silicon-based borohydrides through mechanochemical methods
- Demonstrated several new materials with reversibility for part of their total capacity

“Li₂SiS₂(BH₄)₂” desorption measurements

Accelerating development of improved hydrogen storage materials
FY2017 FOA Topic

- **Hydrogen Storage Materials Discovery (HyMARC)**
  - innovative, high-risk, high-payoff concepts for hydrogen storage materials
  - project teams will be integrated into HyMARC as individual projects
  - phase I Go/No-Go milestone must provide confidence that the proposed concept has reasonable potential to result in a hydrogen storage material capable of meeting automotive performance requirements

- **Areas of interest:**
  - novel, advanced **onboard-rechargeable** hydrogen storage materials
  - physi- and chemisorption materials acceptable

- Only Phase I effort will be supported until Go/No-Go criteria is met, additional support will be contingent on meeting criteria
Accomplishments - Project Highlights

Computational Screening of MOFs with High Volumetric Density [U. Michigan]
- Identifying high-performing MOF’s through screening of large structure databases
- Synthesized and tested several MOFs for their H₂ adsorption properties; IRMOF-20 and DUT-23(Co) both projected to surpass MOF-5 in system performance

- [Graphene-based carbon sorbents [Caltech]]
  - Design and synthesize porous graphene materials as high-capacity H₂ sorbents
  - Demonstrated progress in preparing high-surface area carbons and inserting metal atoms to achieve higher heats of adsorption

- [SEM of high surface area graphene prepared from graphene oxide]

Developing improved adsorbent storage materials
Low-cost methods for $\alpha$-alane production [SRNL, Greenway; Ardica, SRI]

- Developing and demonstrating low-cost processes for scale-up of alane ($\text{AlH}_3$) preparation
- Demonstrated improved crystallization and passivation process to produce high-purity, stable $\alpha$–alane from chemical synthesis in batches of up to 200 grams (SRNL, Greenway)
- Demonstrated ability to yield $\alpha$-alane from electrochemical synthesis, however further improvements are needed (Ardica, SRI)

XRD of crystalized $\alpha$-alane from chemical (left) and electrochemical (right) syntheses

Crude Product for $\text{AlH}_3$-$\text{N}^+\text{PrMe}_2$ conversion to $\text{AlH}_3$ at 77 °C

Developing low-cost $\alpha$-alane ($\text{AlH}_3$) production processes
Engineering
Accomplishments - Project Highlights

Maintenance and Enhancements for HSECoE Models [NREL/PNNL/SRNL]

- Collaborative effort to maintain, update and enhance system models developed under HSECoE to provide a resource to hydrogen storage materials developers
- Posted models include metal hydride, chemical, and sorbent H₂ storage systems
- Improved framework utility for materials researchers through new isotherm fitting and estimator tools.

Online system models maintained and accessible to the research community
Accomplishments - Project Highlights

Materials-based H₂ Storage for UUV Applications [SRNL/US Navy/Ardica]

- Developing a materials-based H₂ storage system to extend UUV mission duration
- Preliminary analysis indicate ≥2 times longer mission capability over battery operation

Metal Hydride H₂ Storage for Forklift Applications [Hawaii H₂ Carriers/SRNL]

- **Small Business Voucher** project to demonstrate MHHS performance on a forklift under realistic conditions and its fast fill capabilities; perform preliminary DFMA analysis
- System originally designed and built under a SBIR program

Leveraging HSECoE models and capabilities for high-value applications
Analysis
Accomplishments – Project Highlights

Hydrogen Storage System Performance [ANL] and Cost Analyses [SA/PNNL/ANL]

- Analyses are carried out to estimate system performance and cost of various technologies to help identify focus areas for the Program and to gauge technology development progress
- Cryo-compressed H₂ storage systems were evaluated for heavy duty fleet (bus) applications
- 500 bar, 40 kg H₂ capacity systems projected to be able to achieve 7.3 wt.% and 43 g/L storage densities with a cost of $15/kWh

Analysis of a 40 kg H₂ capacity, 500 bar cryo-compressed system for bus applications

Weight breakdown

<table>
<thead>
<tr>
<th>Component</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Composite</td>
<td>48%</td>
</tr>
<tr>
<td>Liner</td>
<td>25%</td>
</tr>
<tr>
<td>Shell</td>
<td>10%</td>
</tr>
<tr>
<td>H₂</td>
<td>8%</td>
</tr>
<tr>
<td>BOP</td>
<td>9%</td>
</tr>
<tr>
<td>MLVSI</td>
<td>&lt;1%</td>
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</tbody>
</table>

Cost Breakdown @ 5000/yr

$19,907/System @ $14.93/kWh

- BOP: 42%
- Liner: 6%
- Insulation: 8%
- Other mfg. steps: 10%
- Composites and Fiber Winding: 34%
Collaborations

INTERNATIONAL ACTIVITIES
• IEA HIA Task 32 Hydrogen-based Energy Storage

DOE – EERE - FCTO
Hydrogen Storage Applied R&D
• Physical Storage
• Fiber Composites
• Materials-based Storage
• System Engineering
• Testing and Analysis

INDUSTRY
• U.S. DRIVE
  Tech teams:
  ➢ H₂ Storage
  ➢ H₂ Delivery
  ➢ Codes & Standards
  ➢ Fuel Cells
  ➢ Fuel Pathways
  ➢ Vehicle Systems

TECHNOLOGY VALIDATION

National Collaborations (inter- and intra-agency efforts)

Collaborating and leveraging of national and international activities
Summary

• **Physical Storage**
  - Focus is on developing technologies to lower the cost of 700 bar systems
  - On-going projects on alternative materials and manufacturing processes
  - Conformable tank designs may provide improved packaging onboard vehicles
  - FOA topic on low-cost, alternative precursors for high-strength carbon fiber

• **Materials-based Storage**
  - Focus is to accelerate development of H$_2$ storage materials with targeted properties
  - HyMARC core team performing foundational research to develop computational tools
  - Rechargeable metal hydrides and hydrogen sorbents are primary materials areas
  - First round of seedling projects underway and FOA topic to select second round
  - Engineering activities leverage prior work to meet needs of high-value applications

<table>
<thead>
<tr>
<th>FY 2017</th>
<th>FY 2018</th>
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<tbody>
<tr>
<td>HyMARC team to prepare sorbent strategy prioritization</td>
<td>First round of seedlings have go/no-go decisions</td>
</tr>
<tr>
<td>First round of seedlings working with HyMARC</td>
<td>Second round of seedlings working with HyMARC</td>
</tr>
<tr>
<td>Second round of seedlings to be selected</td>
<td>Low-cost high-strength CF precursor projects up and running</td>
</tr>
<tr>
<td>Low-cost high-strength CF precursor projects to be selected</td>
<td></td>
</tr>
</tbody>
</table>
## Contacts – H₂ Storage Team

<table>
<thead>
<tr>
<th>Name</th>
<th>Title</th>
<th>Phone</th>
<th>Email</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ned Stetson</td>
<td>Program Manager</td>
<td>202-586-9995</td>
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</tr>
<tr>
<td>Grace Ordaz</td>
<td></td>
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<tr>
<td></td>
<td>Now retired and</td>
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<tr>
<td></td>
<td>enjoying life after DOE!</td>
<td></td>
<td></td>
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<tr>
<td>Katie Randolph</td>
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<td><a href="mailto:chris.werth@ee.doe.gov">chris.werth@ee.doe.gov</a></td>
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BACK UP
HyMARC: Accelerating the discovery of breakthrough H$_2$ storage materials

HyMARC provides **capabilities** and **foundational understanding** of phenomena governing thermodynamics and kinetics for the development of solid-state hydrogen storage materials.

HyMARC delivers **community tools and capabilities:**
- **Computational models and databases** for high-throughput materials screening
- **New characterization tools and methods** (surface, bulk, soft X-ray, synchrotron)
- **Tailorable synthetic platforms** for probing nanoscale phenomena

**Website:** [hymarc.org](http://hymarc.org)
Baseline system projections based on single tank design

Lowest cost, but most difficult to package onboard a vehicle
All current commercial FCEVs have dual tank designs.

Higher cost, but most easier to package onboard a vehicle.