Hydrogen Storage

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2007 DOE Hydrogen Program
Merit Review and Peer Evaluation Meeting

May 15, 2007

¹ Sandia- Retired, on assignment to DOE, Washington DC
Outline

• Goals and Objectives
  Targets & challenges

• Budget

• Progress
  Results in the last year
  R&D examples

• Future Plans
Hydrogen Storage: The “Grand Challenge”

Goal: On-board hydrogen storage for > 300 mile driving range and meet all performance (wt, vol, kinetics, etc.) , safety and cost requirements.

<table>
<thead>
<tr>
<th>Examples of Targets</th>
<th>2010</th>
<th>2015</th>
</tr>
</thead>
<tbody>
<tr>
<td>System Gravimetric Capacity (net)</td>
<td>6 wt.% (2.0 kWh/kg)</td>
<td>9 wt.% (3.0 kWh/kg)</td>
</tr>
<tr>
<td>System Volumetric Capacity (net)</td>
<td>1.5 kWh/L (45 g/L)</td>
<td>2.7 kWh/L (81 g/L)</td>
</tr>
<tr>
<td>Storage System Cost</td>
<td>$4/kWh (~$133/kg H₂)</td>
<td>$2/kWh ($67/kg H₂)</td>
</tr>
<tr>
<td>Min. Full Flow Rate</td>
<td>0.02 g/s/kW</td>
<td>0.02 g/s/kW</td>
</tr>
<tr>
<td>Refueling Time (for 5 kg)</td>
<td>3 min</td>
<td>2.5 min</td>
</tr>
<tr>
<td>Cycle Life (Durability)</td>
<td>1000 cycles</td>
<td>1500 cycles</td>
</tr>
</tbody>
</table>

More targets and explanations at [www.eere.energy.gov/hydrogenandfuelcells/](http://www.eere.energy.gov/hydrogenandfuelcells/)
Current Status vs. Targets

No technology meets targets - results include data from vehicle validation

- Gravimetric Capacity (wt%)
- Volumetric Capacity (g/L)
- USABC 2010 target
- USABC 2015 target
- Complex hydride
- Chemical hydride
- Liquid hydrogen
- Cryocompressed
- "Learning Demo"
- 700 bar
- 350 bar
- ~103-190 miles
  Independent validation (DOE Tech Val Program)

Estimates from developers & analysis results; periodically updated by DOE. "Learning Demo" data is for 63 vehicles.
“...DOE should continue to elicit new concepts and ideas, because success in overcoming the major stumbling block of on-board storage is critical for the future of transportation use of fuel cells.”


Balanced portfolio
• ~ 40 universities, 15 companies, 10 federal labs

Aims to address NAS & other peer review recommendations

Annual solicitation for increased flexibility

Close coordination with basic science

Coordination with industry, other agencies & globally

2. Basic science for hydrogen storage conducted through DOE Office of Science, Basic Energy Sciences
3. Coordinated with Delivery Program Element
Applied R&D Hydrogen Storage Budget

FY2008 Budget Request = $43.9M  
FY2007 Appropriation = $34.6M  
(FY2006 Appropriation = $26.0M)

- **Emphasis:** Ramp up materials R&D through CoE & independent projects
- Tailor materials to focus on T, P, kinetics (as well as capacity)
- New Center of Excellence planned—Engineering Sciences*

Close coordination with Basic Science
$36.4M (FY07)  
$59.5M (FY08)  
Includes basic science for hydrogen storage, production and use (e.g., catalysis, membranes, etc.)

*subject to appropriations
Selected Examples of Progress: High capacity materials also focused on improving thermodynamics, kinetics, regeneration

<table>
<thead>
<tr>
<th>Metal Hydrides</th>
<th>Chemical H₂ Storage</th>
<th>Adsorbents/Carbon</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alane</td>
<td>~8-10 wt%, ~150 g/L (&lt;150 C)</td>
<td>4,7 Phenanthroline (organic liquids)</td>
</tr>
<tr>
<td>Borohydrides</td>
<td>&gt;9 wt%, ~100 g/L (~250 - 350 C)</td>
<td>~7 wt%, ~65 g/L (&lt;225 C)</td>
</tr>
<tr>
<td>Destabilized Binary hydrides</td>
<td>~5-7 wt%, ~60-90 g/L (~250 C)</td>
<td>Seeded Ammonia Borane ~9 wt%, ~90 g/L (&gt;120 C)</td>
</tr>
<tr>
<td>Li Mg Amides</td>
<td>~5.5 wt%, ~80 g/L (&gt;200 C)</td>
<td>Ammonia Borane/Li amide ~7 wt%, ~54 g/L (~85 C)</td>
</tr>
</tbody>
</table>

2006

<table>
<thead>
<tr>
<th>Alane (AlH₃) regeneration Chemical, electrochemical, supercritical fluids</th>
<th>1,6-Naphthrydine ~7 wt%, ~70 g/L (275 C) Surface supported catalyst</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiBH₄/C aerogels</td>
<td>Amine boranes Ionic liquids</td>
</tr>
<tr>
<td>6-8 wt.%, ~55-75 g/L (~300 C)</td>
<td>~7 wt.% , 39 g/L (85 C)</td>
</tr>
<tr>
<td>Reversible Ca(BH₄)₂</td>
<td>AB/LiNH₂, AB/LiH</td>
</tr>
<tr>
<td>~9.6 wt.%, ~105 g/L (~350 C)</td>
<td>~9 wt.% , ~70 g/L (85 C)</td>
</tr>
<tr>
<td>Mn(BH₄)₂</td>
<td>Solid AB</td>
</tr>
<tr>
<td>9-13 wt.% (&gt;100 C)</td>
<td>&gt;16 wt.% , &gt;199 g/L (155 C)</td>
</tr>
<tr>
<td>Mg(BH₄)₂</td>
<td>(&gt;3g/s/kgAB)</td>
</tr>
<tr>
<td>9-12 wt.%, ~110 g/L (~350 C)</td>
<td>Liquid AB/catalyst</td>
</tr>
<tr>
<td>Destabilized hydrides</td>
<td>~ 6 wt.% (~ 80 C)</td>
</tr>
<tr>
<td>DFT identified new reactions</td>
<td>Regeneration</td>
</tr>
<tr>
<td>LiBH₄/MgH₂, CaH₂/LiBH₄, LiNH₂/LiH/Si</td>
<td>2 step process, est.&gt;50% eff.</td>
</tr>
</tbody>
</table>

2007

<table>
<thead>
<tr>
<th>Bridged cat./IRMOF-8 &gt;3 wt.%, 100 bar (25 C) ~20 kJ/mol</th>
<th>Bridged cat./AX-21 &gt;1 wt.%, 100 bar (25 C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C aerogels</td>
<td>C aerogels</td>
</tr>
<tr>
<td>~5 wt.%, ~30 g/L (77 K)</td>
<td>~2 wt.% (77 K)</td>
</tr>
<tr>
<td>Metal-doped C aerogels</td>
<td>~7-7.5 kJ/mol</td>
</tr>
<tr>
<td>~7-7.5 kJ/mol</td>
<td>PANI</td>
</tr>
<tr>
<td>2.8 wt.%, 25 bar (25 C)</td>
<td>2.8 wt.%, 25 bar (25 C)</td>
</tr>
<tr>
<td>Release at ~100-220 C</td>
<td>Release at ~100-220 C</td>
</tr>
</tbody>
</table>
Results: Theory Guided Materials Discovery

Theory for rapid screening

- >160 compounds
- >300 reactions
- Energetically favored systems identified

Example: Experimental progress

Theory predicted promising properties

$$3\text{Ca(BH}_4\text{)}_2 \rightarrow \text{CaB}_6 + 2\text{CaH}_2 + 10\text{H}_2$$

$$\Delta H = 53 \text{ kJ/mole}$$

9.6 wt. %

Developed synthesis route (75-80% yield)

Reversibility demonstrated

$$\text{CaB}_6 + 2\text{CaH}_2 \xrightarrow{100 \text{ bar H}_2, 390 ^\circ \text{C}} 1\% \text{ rehydrided}$$

$$\text{CaB}_6 + 2\text{CaH}_2 \xrightarrow{700 \text{ bar H}_2, 400 ^\circ \text{C}} \text{fully rehydrided}$$

Theory guides experiment

Experiment refines theory

Alapati, Johnson and Sholl,

Majzoub, D. Johnson, Bowman et al., MHCoE

Majzoub, D. Johnson, Bowman et al., MHCoE

E. Ronnebro, E. Majzoub et al. Sandia

*Phys Rev B* 74, 155122 (2006)
Results: Sorbent Materials

Independent verification of MOF-177 (O. Yaghi et al. - highest capacity to date worldwide; > 7 wt.%, 77 K)

Independent verification of > 2x increase in capacity due to spillover (R. Yang et al.)

Spillover by Yang, U Michigan

Room Temp!
NH₃BH₃ → BNHₓ + 3H₂  19.4 wt.%, 160 g/L (theoretical material capacity)

New ionic liquids enhance H release; eliminate induction time

Chemical promoters enhance H release in ammonia borane (AB)

Sneddon, et al, U. Penn and CH Center
Results: Liquid Carriers and Systems Analysis

Organic liquid carriers & catalysts

1,6-Naphthyridine
Liquid at room temp.
7.2 wt. % H₂, ~70 g H₂/l
release temp. ~275°C

A. Cooper, et al., Air Products

Regeneration of Ammonia Borane

Example of Reactor Modeling & Sensitivity Analysis

ΔH (kJ/mol) = 35 40 45 51

R. Ahluwalia, et al. ANL
### Results: Regeneration Option Assessments

**Example of systematic approach to down-selects**

<table>
<thead>
<tr>
<th>Option Criterion</th>
<th>Weighting</th>
<th>Schlesinger</th>
<th>Metal Reduction</th>
<th>Echem</th>
<th>Borane</th>
<th>Metathesis</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Chemistry demonstrated</strong></td>
<td>Pref</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Cost/per unit H2 (NaBH4)</td>
<td>25</td>
<td>1</td>
<td>5</td>
<td>7</td>
<td>7</td>
<td>3</td>
</tr>
<tr>
<td>Energy consump (theor efficiency)</td>
<td>25</td>
<td>3</td>
<td>8</td>
<td>7</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>Raw material consump - high conv /yields</td>
<td>5</td>
<td>7</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Low operating severity</td>
<td>5</td>
<td>5</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Few chemical reactions</td>
<td>5</td>
<td>5</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Few separation/processing steps</td>
<td>5</td>
<td>5</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Capital cost, $ per unit H2 (NaBH4)</td>
<td>10</td>
<td>6</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Low complexity</td>
<td>10</td>
<td>6</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Low technical risk</td>
<td>5</td>
<td>10</td>
<td>7</td>
<td>7</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>EHS (environmental / health / safety)</td>
<td>10</td>
<td>10</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Emissions, wastes, CO2</td>
<td>5</td>
<td>8</td>
<td>7</td>
<td>6</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>toxicity, safety, flammability, H2O-reactive</td>
<td>5</td>
<td>8</td>
<td>7</td>
<td>6</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>other ecological components?</td>
<td>5</td>
<td>8</td>
<td>7</td>
<td>6</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>Logistics (supply / distribution)</td>
<td>5</td>
<td>10</td>
<td>7</td>
<td>10</td>
<td>7</td>
<td>8</td>
</tr>
<tr>
<td>Abundant raw materials</td>
<td>5</td>
<td>10</td>
<td>7</td>
<td>10</td>
<td>7</td>
<td>8</td>
</tr>
</tbody>
</table>

- **Assess efficiency potential & other factors for viable chemical carriers**
- **Develop & demonstrate chemistry & processes to improve regeneration efficiency**

Different weighting factors can be analyzed

Linehan, Lipiecki, Chin, Rohm & Haas and CH Center
Results: Systems, Safety, Testing & Analyses

2nd Gen Prototype Built (Ti-NaAlH4)

Estimated 2.0 wt% & 21 g/L
(Projected 2.3 wt.% and 24 g/L)

Examples of Storage System Cost Analyses

Sensitivity analysis shows key cost drivers

System Cost, $/kWh

Cryo-Compressed $2
LH2 $4
Carbon $6
NaBH4 $8
NaAlH4 $10
5,000 psi $12
10,000 psi $14
2010 Target ($4/kWh)

SwRI- Independent testing underway

New: Storage Materials & Systems Safety (U.S., Japan, Germany, Canada)

Cryo-Compressed Tank Concept
Demonstrated w/ DOE Tech Val.

4.7 wt. %
30 g/L
(ANL estimate)

Lasher, et al, TIAx LLC

Mosher, et al, UTRC

Aceves, Berry, et al, LLNL
Examples of Hydrogen Storage Collaboration

IEA – HIA TASK 22

A total of 43 projects have been proposed for Task 22. This includes participation by 15 countries, 43 organizations, and 46 official experts.

Project Types:
- Experimental
- Engineering
- Theoretical Modeling (scientific or engineering)
- Safety Aspects of Hydrogen Storage Materials

Classes of Storage Media
- Reversible Metal Hydrides
- Regenerative Hydrogen Storage Materials
- Nanoporous Materials
- Rechargeable Organic Liquids and Solids

DoD: DEFENSE LOGISTICS AGENCY

New Storage Awards (4/07):
- High throughput - Combinatorial Screening: U of Central Florida, UC Berkeley & Symyx, Miami U (Ohio) & NREL
- Reversible System Dev’t & Demonstration: Energy Conversion Devices, U of Missouri (phase 1 design)

Interagency Hydrogen R&D Task Force (OSTP)

- NSF - proposal review in process (5/07)
- NIST - neutron scattering
Strategy & Execution
Example - maps portfolio & requirements to meet targets

Hydrogen Storage Tech Team Outcomes Map

Current R&D:
- System Project: High Capacity H₂ Storage System Design and Demonstration, UTROG
- Distributed Technology Programs, DOE
- Storage Systems and Performance Analysis, RNL
- Analyses of H₂ Storage Material, Cost, and On-Board Systems, TRAX
- H₂ Sorption@Center NREL, NIST, and others
- NH Center, SNL, and others
- Metal Hydride Storage Materials, U of Hawaii
- Complex Hydride Compounds with Enhanced H₂ Storage Capacity, UTRC
- Novel Complex Metal Hydrides for H₂ Storage, JGIP
- Direct Microreactor for Hydrogen, MIT, U of New York, and SNL
- Metal Hydrides for H₂ Storage, Michigan Technological U
- Nanomaterials for H₂ Storage, University of California-Berkeley and LLNL
- Nanogranular New Materials for H₂ Storage, LLNL, U of California-Santa Barbara
- Lithium Nitride Materials for H₂ Storage Sub, U of Connecticut, FNINL
- Vesic-Organic Frameworks for H₂ Storage, UCL
- Carbon-Carbon Carbon and Carbon Nanomaterials for H₂ Storage, U of Florida
- Non-Graphite Graphite-Based Materials for H₂ Storage, Graz Technology Institute
- Biomimetic Approaches to New Adsorbent Materials, Minn U of Ohio
- Nanomaterials for Hydrogen Storage, SNL, and others
- Advanced Storage Concepts, LAI, and others

- Advanced Safety Concepts, UTROG, and others
- Advanced and Conformable Tanks, LLNL, Quantum
- Safety Projects (SNL, SNL, and UTROG)

Legend:
- H₂ storage R&D Focus (Storage Team) (limited by government and industry)
- Other Focus Area (limited primarily by industry or others)

Requirements:
- Lightweight H₂ storage
- Lightweight container
- Efficient thermal management systems
- High packing density
- Compact container
- Compact components (less than 1% system unit)
- Efficient storage system integration (compact system design)
- Breakthrough in high H₂ capacity material (9 wt%)
- High density material (e.g., >0.9 g/cm³)
- H₂ accessibility (e.g., 20%)
- Multifunctional materials design (e.g., micromachined/heat exchanger)
- Corrosion-resistant, degradable (if not necessary)
- Low cost storage material (including catalyst, etc.)
- Low cost container
- Low cost components
- Low maintenance, high durability
- High performance, volume capacity (500,000 units)

Targets:
- System Weight:
  - 2010: 2 kWhr/kg (50 wt%)
  - 2015: 3 kWhr/kg (60 wt%)
- System Volume:
  - 2010: 1.5 kWhr (46 g/L)
  - 2015: 2.7 kWhr (81 g/L)
- System Cost:
  - 2010: $400/kW
  - 2013: $200/kW

FreedomCAR
Fuel Partnership
Keep up the sustained effort and high technical quality work & be flexible! Address volumetric capacity, T, P, kinetics, etc. (not just wt. %!)

*Subject to appropriations and direction*
Hydrogen Storage Team

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www.hydrogen.energy.gov
Acknowledgements

DOE Researchers
Global Hydrogen Storage R&D Community
FreedomCAR & Fuel Partnership Technical Team
Reviewers

In Memoriam
Professor Alan MacDiarmid*
1927-2007

*2000 Nobel Prize in Chemistry (conducting polymers)
Thank you
Additional Information
# Applied R&D Hydrogen Storage “Grand Challenge” Partners:
Diverse Portfolio with University, Industry and National Lab Participation

## Centers of Excellence

<table>
<thead>
<tr>
<th>Metal Hydride Center</th>
<th>Hydrogen Sorption Center</th>
<th>Chemical Hydrogen Storage Center</th>
</tr>
</thead>
<tbody>
<tr>
<td>National Laboratory: Sandia-Livermore</td>
<td>National Laboratory: NREL</td>
<td>National Laboratories: Los Alamos, Pacific Northwest</td>
</tr>
</tbody>
</table>

## Independent Projects

<table>
<thead>
<tr>
<th>Advanced Metal Hydrides</th>
<th>Sorbent/Carbon-based Materials</th>
</tr>
</thead>
<tbody>
<tr>
<td>UTRC, UOP</td>
<td>UCLA</td>
</tr>
<tr>
<td>Savannah River Nat’l Lab</td>
<td>State University of New York</td>
</tr>
<tr>
<td>Univ. of Connecticut</td>
<td>Gas Technology Institute</td>
</tr>
<tr>
<td>Sorbent/Carbon-based Materials</td>
<td>UPenn &amp; Drexel Univ.</td>
</tr>
<tr>
<td>UCLA</td>
<td>Miami Univ. of Ohio</td>
</tr>
<tr>
<td>Sorbent/Carbon-based Materials</td>
<td>Chemical Hydrogen Storage</td>
</tr>
<tr>
<td>Air Products &amp; Chemicals</td>
<td>Air Products &amp; Chemicals</td>
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<tr>
<td>RTI</td>
<td>RTI</td>
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<tr>
<td>Millennium Cell</td>
<td>Millennium Cell</td>
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<tr>
<td>Safe Hydrogen LLC</td>
<td>Safe Hydrogen LLC LLC</td>
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<tr>
<td>Univ. of Hawaii</td>
<td>Univ. of Hawaii</td>
</tr>
<tr>
<td>Chemical Hydrogen Storage</td>
<td>Other New Materials &amp; Concepts</td>
</tr>
<tr>
<td>Air Products &amp; Chemicals</td>
<td>Alfred University</td>
</tr>
<tr>
<td>RTI</td>
<td>Michigan Technological University</td>
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<td>Millennium Cell</td>
<td>UC-Berkeley/LBL</td>
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<td>Safe Hydrogen LLC LLC</td>
<td>UC-Santa Barbara</td>
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<tr>
<td>Univ. of Hawaii</td>
<td>Argonne Nat’l Lab</td>
</tr>
<tr>
<td>Other New Materials &amp; Concepts</td>
<td>Tanks, Safety, Analysis &amp; Testing</td>
</tr>
<tr>
<td>Alfred University</td>
<td>Lawrence Livermore Nat’l Lab</td>
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<tr>
<td>Michigan Technological University</td>
<td>Quantum</td>
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<tr>
<td>UC-Berkeley/LBL</td>
<td>Argonne Nat’l Lab, TIA LLC</td>
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<tr>
<td>UC-Santa Barbara</td>
<td>SwRI, UTRC, Sandia Nat’l Lab</td>
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<tr>
<td>Argonne Nat’l Lab</td>
<td>Savannah River Nat’l Lab</td>
</tr>
</tbody>
</table>

## Coordination with: Basic Science (Office of Science, BES)

MIT, U.WA, U. Penn., CO School of Mines, Georgia Tech, Louisiana Tech, Georgia, Missouri-Rolla, Tulane, Southern Illinois; Labs: Ames, BNL, LBNL, ORNL, PNNL, SRNL.
Assessment of CoE model:

Multi-institutional critical mass applied R&D is proving to be effective

Programmatic Results: Focus on strategy & execution

- Annual Solicitation (6 new projects complement portfolio)
- No-go (FY06) on pure SWNTs (doped C/basic science still go)
- Assessment of Cryo-compressed tanks complete
- Outcomes maps done & targets revisited through FreedomCAR
It’s not just about capacity– much research is focused on tailoring kinetics & thermodynamics…

- **Hydride heat of formation**
  - pressure limits (~20-35 kJ/molH₂)
  - refueling (<20 kJ/molH₂)

- **Surface heat of adsorption**
  - operating temperature
  - release temperature

- **Activation barrier for regeneration**
  - energy efficiency
  - near thermo-neutral
Apply theory & experimentation to design & develop novel, high-performance materials to meet specific performance targets:
• Develop new materials, leverage knowledge from basic research
• Optimize materials and testing to improve performance
• Design, develop and demonstrate materials, components and prototype systems to meet milestones

Develop and use theoretical models & fundamental experimentation to generate knowledge:
• Fundamental property & transport phenomena
• Novel material structures, characterization
• Theory, modeling, understand reaction mechanisms

Test Systems under Real World Conditions
• Demonstrate and validate performance against targets
• Gain knowledge (e.g. fueling time, driving range, durability, cost, etc.) and apply lessons learned to R&D
Summary

• New Materials & Concepts are critical- address volumetric capacity, T, P, kinetics, etc. (not just wt. %!)

• Basic science is valuable to develop fundamental understanding & complements applied research & development

• Engineering issues need to be considered
  – System issues, thermal mgmt, safety, refueling, testing, etc

• Examples of Essential Capabilities:
  – Modeling & Analysis
  – Combinatorial/high throughput methods
  – Material properties measurements
  – Standardized & accurate testing