

IV.D.1b SRNL Technical Work Scope for the Hydrogen Storage Engineering Center of Excellence: Design and Testing of Metal Hydride and Adsorbent Systems

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Objectives

The Savannah River National Laboratory (SRNL) and the University of Quebec at Trois-Rivieres (UQTR) will:

- Compile property data for:
 - NaAlH₄, a metal hydride.
 - AX-21, an adsorbent.
- Collect property data for other select metal hydrides and adsorbents.
 - Compile list of available analytical techniques to support materials property data requirements.
- Develop acceptability envelope for storage media and vessels.
- Develop numerical models to adequately predict storage system behavior for NaAlH₄ and AX-21-based storage systems.
 - Use the models to design optimized storage systems based on NaAlH₄ and AX-21.
- Develop practical and efficient enabling technologies in the areas of hydrogen purification and demonstrate material compatibility for various systems and components for adsorbent and metal hydride storage materials.

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Storage section (3.3) of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- (A) System Weight and Volume
- (C) Efficiency
- (E) Charging/Discharging Rate

Technical Targets

The goal of the entire Hydrogen Storage Engineering Center of Excellence (HSECoE) is to provide a system model for each material sub-class (metal hydrides, adsorption, chemical storage) which meets the “Technical System Targets: On-Board Hydrogen Storage for Light-Duty Vehicles”, Table 3.3.2 in the DOE Multi-Year Research and Development Plan – April 2009. The end-of-Phase I, Go/No-Go milestone for the entire HSECoE project is that:

1. Four of the DOE 2010 numerical system storage targets are fully met and that
2. The status of the remaining numerical targets must be at least 40% of the target or higher.

For SRNL's specific technical portion of HSECoE, SRNL will:

- Compile thermochemical data.
- Bound media operating characteristics for metal and adsorption hydride material.
- Develop and apply numerical models that couple mass, momentum and energy balances with chemical kinetics and/or isotherms to simulate hydrogen uptake and discharge.
- Identify technology gaps.
- Identify preliminary system designs to achieve DOE 2015 hydrogen storage goals.

Accomplishments

- Compiled and distributed a list of data requirements for storage media.
- Developed general model for NaAlH₄ metal hydride-based storage systems.
 - Validated model against heat transfer and kinetics data.

- Conducted preliminary optimization studies with model.
- Obtained thermodynamic models for AX-21 adsorbent from University of Quebec.
 - Began incorporation of thermodynamic models into a multidimensional model.
- Determined analytical capabilities available within SRNL to support testing for materials properties that are not available in the literature.
- Initiated materials property database for candidate metal hydride storage materials.
- Initiated work on hydrogen purity/quality.



Introduction

SRNL and UQTR will be involved in several critical aspects of the HSECoE. SRNL will focus primarily on metal hydride storage media, and at a secondary level will work to ensure the integration and synergy of adsorbent and chemical hydride systems developed by other team members. SRNL will apply its expertise in modeling dynamic transport phenomena and chemical processes, materials testing, and system modeling to accomplish its objectives in the proposed effort – developing and applying models to identify viable subscale prototype designs, performing design calculations sufficiently accurate for engineering application, and defining the scope and required measurements for experiments with the selected prototypes. Additionally, SRNL will develop technologies to control the purity of hydrogen delivered from the storage systems and will perform analyses and testing to ensure materials compatibility.

UQTR will develop an efficient adsorption model that can be easily implemented but still accurately predict the adsorption characteristics over a range of temperatures. UQTR will extend its current model for hydrogen adsorption on activated carbons to any adsorbent used in the storage system design.

Approach

The project objectives will be carried out in four areas: Transport Phenomena, Hydrogen Purification, Materials Compatibility and Materials Property Database Development. The Transport Phenomena technology area will focus on the processes and components internal to the storage vessel. Models developed within this technology area will be used to downselect and design scaled prototype storage systems for testing. Other efforts by this technology area will include design of test stations and development of test matrices for required data. Data from the prototype

tests will be used to tune and validate the models. The validated models will then be used to predict the performance of full-scale storage systems. Enhancement of storage media performance by optimizing its geometric form and properties will be investigated. The Hydrogen Purification area will address the separation of particulate and gaseous impurities from hydrogen gas discharged from the storage system. The Materials Compatibility area will be responsible for the investigation of the compatibility of the storage media with the materials comprising the vessel wall and components. In addition, to chemical interaction, the technology team will investigate stresses induced by the expansion of storage media during hydrogen uptake. Property data for select metal hydrides will be assimilated by the Materials Property Database area.

Results

SRNL activities for the HSECoE began on February 1, 2009. The main focus was the development of models for use in optimizing storage system designs and performing parametric sensitivity studies for design optimization. Implementation of the models required data for the storage media. SRNL's charter was modeling and design of storage systems based on metal hydride and adsorbent storage media. Based on the availability of data and apparent performance the media selected for use in the first models were TiCl_3 catalyzed NaAlH_4 as the metal hydride media and AX-21 as the adsorbent media. Data compilation for other media will continue.

A prioritized list of data required for modeling all media classes – metal hydrides, chemical hydrides and adsorbents – was issued. Analytical capabilities within SRNL were identified for the measurement of media properties not available in the literature. A preliminary compilation of metal hydride properties for NaAlH_4 and other media was compiled from current databases. A literature review was completed for binders used to pelletize hydrogen storage media.

A general multidimensional model for NaAlH_4 systems, based on the integrated model in [1], was developed and issued to United Technologies Research Center (UTRC), UQTR and Oregon State University (OSU) who will assist with validation and optimization studies. The model was validated against heat transfer and kinetics data and some characterization of thermal contact resistance for heat transfer surfaces was performed. Thermal contact resistance is particularly important because the principal challenge for metal hydride based storage systems is the removal of heat released during hydrogen uptake by the media. The very low thermal conductivity of the media necessitates the use of heat transfer elements in the vessel. Thus far fins have been considered as the primary enhancements to heat removal. However, SRNL and the Microproducts

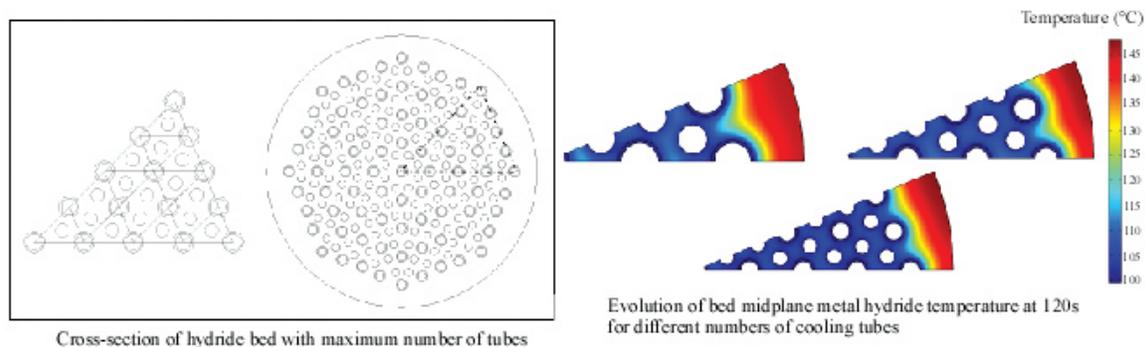


FIGURE 1. Sensitivity of Temperature during H_2 Uptake to the Number of Coolant and H_2 Injection Tubes

Breakthrough Institute at OSU are investigating novel micro heat exchange systems for this purpose. Optimization studies have been performed that examine the placement, number and dimensions of coolant tubes and fins within the storage vessel, see Figure 1 [2]. For heat removal with fins, preliminary results indicate that fin orientation relative to the axis of coolant tubes within the storage vessel affects the efficiency, in terms of heat removal per mass of the fins.

A thermodynamic model for hydrogen uptake and discharge by AX-21 was provided to SRNL by UQTR [3,4]. Currently both organizations are working to incorporate the thermodynamic model into a multidimensional model for an adsorbent-based storage system.

As part of the Materials Compatibility area, several papers have been identified detailing studies of the stress in containers of expanding hydride materials. Future work will build a computational model of container wall stress due to hydride expansion, using these studies.

Within the Hydrogen Purification area, a literature review of contaminant gettering and membrane separations has been initiated. Draft lists of candidate storage media have been obtained and technical challenges, including delivery of hydrogen at approximately 3 bar to fuel cells and hydrogen purification at the tank/fueling station and the tank/fuel cell interfaces, are being evaluated.

Conclusions and Future Directions

- Continue optimization studies using model for $NaAlH_4$ -based storage vessels.
- Complete development of multidimensional model for AX-21 adsorbent-based storage systems:
 - Validation of models.
 - Parametric studies.

– Optimization of storage system designs.

- Develop acceptability envelope for media and storage vessel as an interactive model that identifies required system and media properties and dimensions required to meet storage system performance requirements.
- Data compilation for other adsorbent and metal hydride media than AX-21 and $NaAlH_4$.

References

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2. M. Bhourri. "Sensitivity Study of Alanate Hydride Storage System." Draft report from the Hydrogen Research Institute, University of Quebec at Trois Rivieres, June 2009.
3. M.-A. Richard, P. Benard, R. Chahine. "Gas Adsorption Process in Activated Carbon Over a Wide Temperature Range Above the Critical Point. Part 1: Modified Dubinin-Astakhov Model." *Adsorption*, 15, 43-51, 2009.
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