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Hydrogen Storage: The Key Challenge Facing a Hydrogen Economy

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Abstract:
The development of a viable hydrogen storage system is one of the key challenges that must be met prior to the establishment of a true hydrogen economy. Current hydrogen storage options, such as compressed gas and liquid hydrogen, fall short of meeting vehicle manufacturers' goals for safe and efficient energy storage [1]. The most viable long-term alternative to these options is solid-state storage, which has been proven both safe and efficient. The Savannah River Technology Center (SRTC), with over 50 years of hydrogen storage expertise and over 25 years of expertise in solid-state storage, has assembled a world-class team to meet this key challenge.

The SRTC team is comprised of distinguished scientists and engineers from national laboratories, leading universities, and major corporate research centers that are actively performing research in hydrogen storage on complex hydrides. Their collective expertise in materials development combined with fundamental science and systems engineering, will provide the synergy to meet the hydrogen storage goals. The team's goal is to develop a hydrogen storage system that meets the 2010 U.S. DOE FreedomCAR targets, which includes a system with greater than 6 wt% hydrogen.

This paper will describe the hydrogen storage challenge and also review the approach that the SRTC-led team will follow to help solve this challenge.

Keywords: hydrogen storage, solid-state storage, complex metal hydrides, FreedomCAR, hydrogen fuel initiative

1. Introduction

1.1 Background

On January 28, 2003 in his State of the Union Address, President Bush proposed $1.2 billion for hydrogen infrastructure research and development [2]. This program, together with his previously announced FreedomCAR program, is aimed at enabling industry to make a decision to commercialize a hydrogen fuel cell vehicle by 2015. One of the technologies on the critical path to making hydrogen vehicles a reality is onboard hydrogen storage.

Hydrogen, which has one of the highest gravimetric energy densities of any fuel, unfortunately is also the lightest of all elements. This means that typically large volumes or high pressures are required to store the appreciable amount of hydrogen needed to permit a fuel cell vehicle to exhibit an acceptable operating range. Previous efforts to develop fuel cell vehicles attempted to overcome the hydrogen storage problem by focusing on storing hydrogen chemically bound to hydrocarbon materials, including gasoline, methane and methanol. While some of these materials do provide a highly concentrated form of hydrogen, they require an onboard fuel processor to release and separate the hydrogen. Some of the impurities remaining in the separated hydrogen stream from this onboard processing have been found to adversely affect the performance of fuel cells. Other problems with onboard hydrocarbon reforming are reductions in overall well-to-wheel efficiencies and issues involving carbon dioxide emissions. This has led the U.S. Department of Energy (DOE) and the automotive industry to re-address the onboard hydrogen storage issue.

1.2 Hydrogen Storage Targets

The U.S. DOE, in conjunction with industry, has developed a series of hydrogen storage targets for calendar years 2005, 2010 and 2015 [3]. These targets address gravimetric and volumetric densities, cost, cycle life, refueling rate and loss of usable hydrogen. Some of the 2005 targets are 4.5 wt%, 1.2 kWh/L, and $6/kWh. Future year targets are considerably more challenging. A complete summary of DOE's hydrogen storage targets is shown in Table 1.

Today, only compressed and liquid hydrogen storage systems come close to meeting these targets. Liquid hydrogen storage can meet or exceed many of the gravimetric and volumetric density targets, but costs and energy use, especially with respect to liquifaction, are high. Liquifying hydrogen typically requires 30% of its energy value. Furthermore, liquid hydrogen storage requires dealing with the additional hazard of handling cryogenic hydrogen at 20 degrees above absolute zero. This also leads to hydrogen venting during prolonged periods of storage. Compressed hydrogen storage has become the current standard for fuel cell demonstration vehicles. Newer carbon fiber composite tanks operating at 350 bar have exceeded 6 wt% hydrogen storage density, but the tank volume is still in excess of the goals by a factor of two or more. Increasing compressed hydrogen pressures from 350 bar to 700 bar can reduce the tank volume by about 33%, but at higher safety risk and increased compression energy requirements. Finally, current composite high-pressure storage tanks cost in excess of $100 per kWh ($3300 per kilogram hydrogen storage capacity). Dramatic cost reductions would be needed to meet the DOE goals.

Table 1. DOE Hydrogen Storage Goals

Storage Parameter	Units	2005	2010	2015
Specific Energy	kWh/kg	1.5	2.0	3.0
	kg H2/kg System	4.5	6.0	9.0
Energy Density	kWh/l	1.2	1.5	2.7
	gm H2/l System*	36	45	81
Storage System Cost	$/kWh	6	4	2
	$/kg H2 capacity	200	133	67
Refueling Rate	kg H2/min	0.5	1.5	2.0
Loss of usable H2	(g/hr)/kg stored	1	0.1	0.05
Cycle Life	Cycles (1/4 to full)	500	1000	1500

*For reference, liquid H2 density is 70 gm/l.

1.3 Solid-state Hydrogen Storage

Solid-state hydrogen storage materials (e.g metal hydrides) have been investigated for over 30 years as on-board hydrogen storage systems due to their excellent volumetric hydrogen storage densities and their inherent low pressure and hydrogen safety aspects. Since hydrides store hydrogen in atomic, rather than molecular form, the volumetric density can exceed that of either liquid hydrogen or the highest pressure compressed gas. For stationary or heavy-duty vehicle applications, traditional interstitial metal hydrides have been successfully utilized [4]. However, their relatively low gravimetric hydrogen density (typically less than 2 wt%) results in excessive storage system weight, limiting their use in fuel cell automobiles. More recently, newer solid-state materials like carbon nanotubes and other forms of carbon have been reported with very high hydrogen capacities. However, as of today, no reliable quantities of these materials have been adequately demonstrated as hydrogen storage materials. Research on carbon systems is expected to continue, and some day future materials may lead to major advances in hydrogen storage technology. Due to the relatively early stage of research, most experts do not expect carbon to be a significant hydrogen storage material for many years.

Perhaps the most promising new solid-state hydrogen storage material is a class of metal hydrides commonly referred to as complex hydrides. They differ from conventional intermetallic hydrides in that complex hydrides are mixed ionic-covalent compounds. They are particularly promising as hydrogen storage materials since they involve lightweight metals, such as aluminum, boron, sodium and magnesium. Absorption and desorption of hydrogen from these materials usually involves solid-phase reactions, and until recently they were believed to be irreversible except under severe conditions. However, in 1997 researchers at the Max Plank Institute in Germany [5] reported that some of these materials could be made to reabsorb hydrogen at reasonable conditions when they are catalyzed by the addition of small amounts of transition metal dopants, such as titanium.

One of these complex hydrides, sodium aluminum hydride, with small quantities of dopants, has been shown to have a reversible hydrogen storage capacity of nearly 5 wt% [6]. Work to improve this material's hydrogen absorption kinetics in order to allow operating conditions compatible with the requirements of fuel cell vehicles is currently under way at the Savannah River Technology Center and other research institutions. Reference [6] above contains more detailed information on this research and other work being performed on additional complex hydride compounds that could lead to hydrogen storage systems with even higher densities that meet or exceed the DOE program goals.

2. Hydrogen Storage at SRTC

The Savannah River Technology Center (SRTC) is a DOE research and development center located at the Savannah River Site (SRS) in Aiken, South Carolina. SRTC has over 50 years of experience in developing and applying hydrogen technology, both through its national defense activities as well as through its recent activities with the DOE Hydrogen Program. The hydrogen technical staff at SRTC comprises over 90 scientists, engineers and technologists, and it is believed to be the largest such staff in the U.S. SRTC has ongoing R&D initiatives in a variety of hydrogen storage areas, including metal hydrides, complex hydrides, chemical hydrides and carbon nanotubes. SRTC has over 25 years of experience in metal hydrides and solid-state hydrogen storage technology research, development and demonstration. As part of its tritium defense mission at SRS, SRTC developed, designed, demonstrated and provides ongoing technical support for the largest hydrogen processing facility in the world based on the integrated use of metal hydrides for hydrogen storage, separation and compression [7].

In 1994, SRTC initiated a project with industrial and academic partners to develop the world's first hybrid-electric, hydrogen-fueled transit bus operating with a metal hydride, storage system [8]. The storage system contained lanthanum-nickel metal hydride and had a capacity of 15 kg of hydrogen. The overall storage system weighed 2000 kg, making it one of the largest vehicle hydrogen storage systems ever built. It operated at 100 psig and used waste heat from an internal combustion engine at 80ºC. The bus operated successfully in transit service in Augusta, Georgia, and was later transferred to Las Vegas, Nevada as part of a DOE hydrogen demonstration program. A photograph of one of the two metal hydride storage containers is shown in Figure 1. SRTC also developed a smaller 2 kg hydrogen storage system for a John Deere "Gator™" industrial fuel cell vehicle [9]. Figure 2 shows a photograph of the metal hydride storage system developed by SRTC.

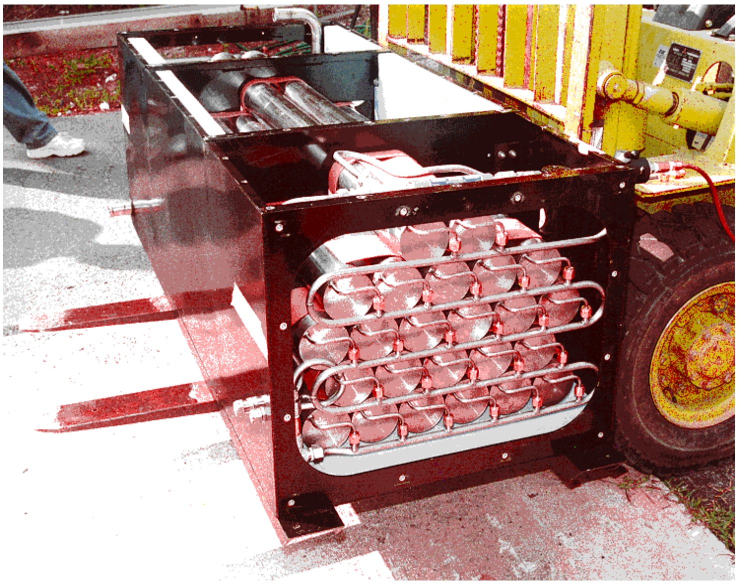

Figure 1. SRTC metal hydride container for H2Fuel hybrid bus.

Figure 2. SRTC metal hydride container for fuel cell "Gator" vehicle.

3. DOE Grand Challenge for Hydrogen Storage

The DOE Office of Hydrogen, Fuel Cells and Infrastructure has recently initiated a "Grand Challenge" to the scientific community for basic and applied research in hydrogen storage [10]. A major, $150 million, 5-year solicitation was issued in the summer of 2003. Funding to support this effort is expected in October of 2004. The DOE's goal is to expedite the research, development and demonstration of on-board hydrogen storage systems capable of meeting the performance goals for hydrogen automobiles. The DOE program includes 4 topical areas which are: virtual centers for hydrogen storage materials research and development; new storage materials concepts; on-board compressed and liquid hydrogen tank technology; and off-board hydrogen storage systems. The DOE hopes to create 3 to 4 Centers of Excellence led by national laboratories and concentrating on each of the primary hydrogen storage technologies: (1) metal and complex hydrides, (2) carbon, and (3) chemical hydrides. Each Center will be responsible for leading and coordinating an integrated hydrogen storage effort that includes both basic and applied research. The Centers are required to include participation by at least 6 universities and one or more industrial participants. Participation by other national and federal laboratories is encouraged. The final deliverable for each Center is to provide a 1 kg hydrogen storage system that meets or exceeds the DOE 2010 targets. Figure 3 shows the main features of the DOE Hydrogen Storage Program.

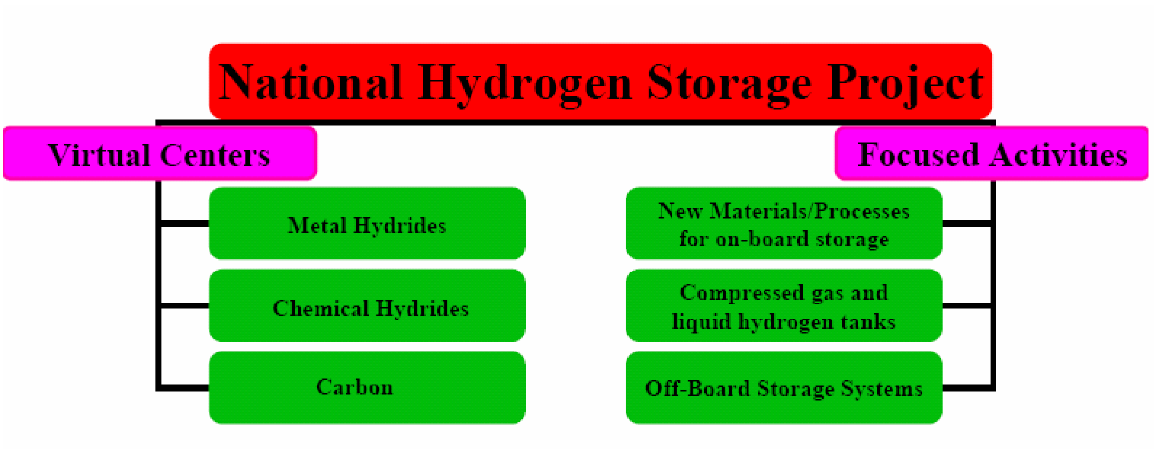

Figure 3. DOE Hydrogen Storage "Grand Challenge" Program

4. SRTC Proposed Center of Excellence

SRTC has responded to the DOE solicitation with a proposal for a Center of Excellence focused on metal and complex hydrides. A world-class team comprised of distinguished scientists and engineers from national laboratories, leading universities, and major corporate research centers has been assembled to meet the DOE challenge. Table 2 lists the major organizations expected to participate in the SRTC Center. All of the key organizations are currently performing research in hydrogen storage on complex hydrides. Their collective expertise in materials development, combined with fundamental science and systems engineering, will provide the synergy to meet the program goals.

Table 2. Planned Partners for SRTC Center of Excellence

Universities	**National and Federal Labs**	**Industry**
University of Hawaii	Argonne National Laboratory	United Technology
Iowa State University	Ames National Laboratory	Intermatix Corp.
Clemson University	Brookhaven National Laboratory	
University of South Carolina	NIST Center for Neutron Research	
Northwestern University	Naval Research Laboratory	
Georgia Institute of Technology		
Virginia Commonwealth University		

To deliver a 1-kg hydrogen capacity storage bed that meets or exceeds the 2010 FreedomCAR targets, the Center will carefully balance the material development and fundamental science efforts with advanced materials and systems engineering. The Center will conduct parallel efforts in system engineering and material development (see Figure 4). Periodic evaluations of small-scale systems will be conducted to better measure the material properties, and to determine their effect on future system performance. These engineering studies will be evaluated by the material developers to determine if a material property can be improved or enhanced. This approach will enable the material engineering and system performance requirements to drive the material development effort, thus allowing the material development effort to remain focused on the delivery of the 1-kg hydrogen storage system—the ultimate goal.

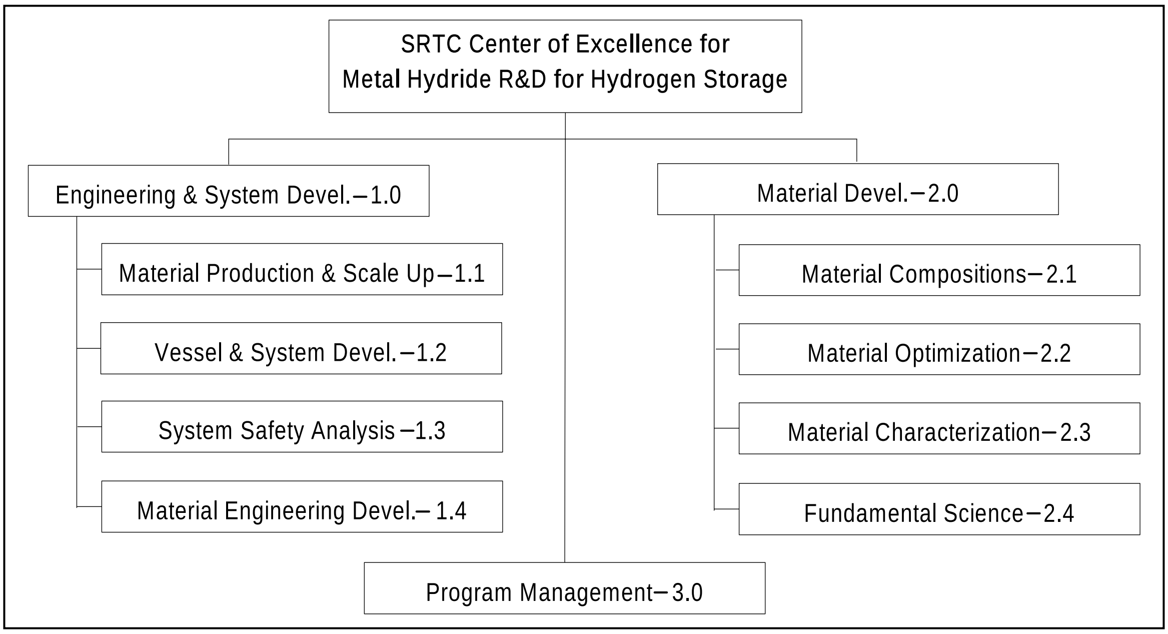

Figure 4. SRTC Proposed Center of Excellence Structure

Many years of development experience by SRTC and its other team leads have shown that careful consideration of engineering requirements in a development program prevents serious consequences. An integrated approach will ensure the balance necessary to be successful. The material development approach takes advantage of ongoing, state-of-the-art activities in complex hydride development, as well as a number of new material composition paths, which should lead to breakthrough results. New compositions will be based on modified alkali and alkaline earth metal complex hydrides and a variety of intermetallic and complex hydride substitutions. New material pathways will include nanomaterial processing techniques as well as a new “molten-state” process developed by SRTC (patent pending). The Center’s material development efforts will be guided by theoretical and experimental analyses, which will give us a better understanding of hydrogen and material interactions. Modeling will be conducted at multiple levels from *ab initio* to thermodynamic to integrated analysis of system performance and cost. Combinatorial techniques will be employed that, when joined with other analysis tools, will substantially minimize experimental efforts and maximize results.

The results from safety, component, and systems engineering tasks will provide feed back into the material development efforts to improve and optimize material properties and characteristics, while meeting the system requirements. A systems engineering approach, which considers optimization of, and tradeoffs among, all components and system requirements, will enable the team to achieve the scientific breakthroughs needed to meet the demanding FreedomCAR goals. This comprehensive development approach, integrating activities from basic science through practical application, is a hallmark of SRTC, and will be integral to the operation of the Center of Excellence in Metal Hydrides.

5. Summary and Conclusion

The future of a world energy system based on hydrogen as a major energy carrier will depend on finding solutions to several key economic, technical and political problems. One of these key problems is the need for low-cost, efficient and safe on-board hydrogen. The hydrogen storage in a fuel cell vehicle must be able to provide comparable performance to that of today’s gasoline fueled vehicles. Passenger room, vehicle range, refueling times and overall vehicle performance, along with actual and perceived safety, must all be addressed. The U.S. DOE has developed a major national program to tackle this challenge. A major portion of the program includes the creation of National Centers of Excellence in several new promising areas of hydrogen storage technology. One of these areas is metal and complex metal hydrides. SRTC, a DOE federal laboratory with an international reputation in metal hydride technology, has put together a world-class team to help meet the hydrogen storage challenge. SRTC will combine both basic and applied research to deliver a 1 kg hydrogen storage system that meets the DOE and FreedomCAR 2010 targets. Further work will be required to meet the more challenging targets needed for vehicle introduction in 2015.

6. References

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