

Hydrides for Processing and Storing Tritium

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Abstract

The Savannah River Site (SRS) has 50 years of experience in handling and processing tritium for defense and other special applications. During the past 20 years, a new technology, metal hydride technology, was introduced to the tritium facilities. This technology dramatically changed the way tritium and the other hydrogen isotopes were handled and processed at SRS. Metal hydrides allowed tritium to be stored much more compactly and at much lower pressures, thereby minimizing accidental release and enhancing operational safety. The use of metal hydrides also simplified many of the processes, resulting in smaller and more efficient operations, which led to significant cost savings.

Multimillion-dollar cost savings have been realized in the existing tritium facilities at SRS by using metal hydride technology. Similar cost savings are expected in several of the new tritium projects. New tritium applications continue to be developed at SRS to ensure the reliability of our nation's tritium reserves and to support our nation's commitment to a strong defense.

In recent years, the Department of Energy and SRS have supported the development of "dual-use" metal hydride technology, which provides benefits not only for defense but also for future energy applications. SRS has collaborated on international energy programs aimed at demonstrating nuclear fusion as a potential, clean, and plentiful source of future energy. SRS has also partnered with government, industrial, and academic institutions to apply its expertise on metal hydrides to clean, non-polluting, hydrogen-powered energy systems. Benefits from these dual-use activities have allowed SRS to maintain its expertise in metal hydrides and have led to substantial cost savings for SRS facilities.

Introduction

What are metal hydrides?

While almost all metals can be made to react with hydrogen under some conditions, only a few metals do so "reversibly" at room temperature and near atmospheric pressures. These materials are generally referred to as "reversible" metal hydrides. Reversible metal hydrides have the ability to absorb and release hydrogen like a solid sponge (Sandrock and Huston 1981). They can do this over and over again. An analogy is that of a household sponge, which can absorb and release water as needed. Reversible metal hydrides can be either pure metal such as palladium, titanium, or zirconium. They can also be intermetallic compounds or alloys made up of two or more metals such as iron-titanium or lanthanum-nickel.

Reversible metal hydrides offer a number of advantages in storing hydrogen versus compressed gas or a cryogenic liquid. Hydrides have an extremely high volumetric density for hydrogen. That means that a lot of hydrogen can be packed in a very small compact space. In fact, most metal hydrides can store hydrogen several times more compactly than high-pressure gas and often more compactly even than liquid hydrogen. This is because the hydrogen atoms in a hydride are bound to metal atoms more closely than they can bind to themselves either as a gas or a liquid. Hydrides can store hydrogen at very low pressure, which affords a higher level of safety. Hydrides often react only with hydrogen, which makes them ideal for use in many separation processes. The disadvantage of hydrides is their relatively high cost and weight.

History of Metal Hydrides

While the ability of some pure metals to absorb hydrogen has been well known for over a 100 years, the discovery of a new class of intermetallic alloys that reversibly absorb and release hydrogen did not come about until the late 1960s. One of the first intermetallic alloys to be “hydrided” was iron-titanium. Brookhaven National Laboratory reported this in 1969 (Sandrock and Huston 1981). Iron-titanium was one of the first practical metal hydrides. It readily absorbs hydrogen at room temperature and is relatively inexpensive. However, iron-titanium also has some disadvantages. It can be easily poisoned by small amounts of oxygen, and substantial heating of the material is required the first time that it is exposed to hydrogen. The initial conditions, required of a metal hydride, to first absorb hydrogen are normally referred to as its “activation” conditions.

Around the same time that iron-titanium was being explored as a practical metal hydride, another important hydride material, lanthanum-nickel, was discovered. The hydride properties of this material were discovered entirely by accident (Sandrock and Huston 1981). The researchers at the time were working on developing permanent magnets when they stumbled on the hydrogen properties of lanthanum-nickel. This became a new and exciting reversible hydride material. Lanthanum-nickel has a high hydrogen capacity and readily absorbs hydrogen at ambient pressures. Furthermore it can be easily activated at room temperature without additional heating and is considerably more resistant to oxygen and other hydride poisons. In the early 1970s, researchers all around the world began exploring the properties of this new reversible hydride along with its many variations. It was discovered that substituting other metals for some of the nickel in the formulation could significantly change the hydrogen properties of the material. Scientists and engineers could now customize their own hydride materials and come up with

operating conditions to match the needs of their application. No longer did they have to settle for a hydride whose properties were just in the “neighborhood”. An engineer could now select the right metal hydride material to meet a specific hydrogen storage temperature and pressure.

Commercial Applications of Metal Hydrides

Metal hydrides have been used for many years as chemical additives or reducing agents in chemical processes. But it was not until the advent of modern room temperature in the early 1970s that reversible hydrides spawned a wide variety of useful applications (Lynch 1991). These applications range from hydrogen storage, separation, and refrigeration to metal hydride batteries used in many of our notebook computers and other electronic devices.

NASA developed some of the first applications of metal hydrides. The objective was to demonstrate long-term hydrogen storage for potential use in space propulsion. Automotive hydrogen storage applications soon followed in several locations around the world. Metal hydrides provided a compact and safe method for storing hydrogen both for engines as well as fuel cell applications. In the 1970s, several metal hydride vehicles, including automobiles, vans, forklifts and even mining vehicles, were successfully demonstrated.

Other demonstrations involved using metal hydrides as compressors to deliver hydrogen at high pressures and as heat pumps and refrigeration systems. Hydride refrigerators usually employ pairs of hydrides with different operating pressures. Hydrogen is allowed to flow from one hydride to the other. The hydride, which is losing hydrogen, naturally cools, and this cooling can be used to provide refrigeration. A waste heat source can be used to return the hydrogen to the first hydride allowing the cooling cycle to be repeated.

One of the most important recent applications of metal hydrides, Nickel Metal Hydride (NiMH) batteries occurred in the late 1980s. These batteries are now widely used in small electronic devices and are being developed for larger applications such as electric vehicles. Some of the benefits of NiMH batteries are that they are rechargeable, have a high-energy density, have no memory effect, and have low toxicity compared to traditional Nickel Cadmium (NiCd) batteries (Lynch 1991). The next generation electric automobile, the General Motors EV1™, is expected to operate on NiMH batteries.

Another important use of metal hydrides is in gas separation. Several companies have developed hydrogen purifiers using metal hydrides. The ability of hydrides to absorb only hydrogen in many gas streams makes them useful in many chemical separation processes. One of the areas where this has been used most successfully is in tritium processing applications.

History of Metal Hydrides at SRS

In the early 1980s, the Savannah River Laboratory began a major program to develop and utilize metal hydrides in its tritium production facilities. Metal hydrides turned out to be ideally suited for tritium handling and processing. Tritium is the radioactive form, or isotope, of hydrogen, produced for defense programs by SRS for over 40 years. Tritium behaves chemically very much like normal hydrogen, except that it is radioactive and decays over time to a form of helium. It should be noted that metal hydrides were used to process tritium at other Department of Energy sites prior to the 1980s. These typically involved using mostly uranium for tritium storage. This material has many drawbacks compared to the materials developed by SRS. Uranium hydride materials require high temperatures to remove the tritium, which leads to tritium permeating or escaping through walls of the container. In addition the pyrophoric or flammable nature of uranium led to many safety concerns with regards to its use at SRS (Ortman et al. 1985).

The 1980s

The first metal hydride small-scale test at SRS began in 1981. A few years later, the first metal hydride applications in the tritium facilities were introduced in a new tritium loading operation. The facility required both near-term storage and compression of tritium. The hydride material chosen for this application was lanthanum-nickel-aluminum (LANA). By substituting a small amount of aluminum for some of the nickel in the lanthanum-nickel hydride, it was found that the operating pressure of the hydride system could be controlled. Therefore, a higher aluminum content alloy with 6% aluminum by weight could be used for low pressure, safe storage of tritium, while a lower aluminum content alloy with only 2% aluminum by weight could be used to pump or compress tritium to higher pressures (Ortman et al. 1990). Another advantage of using the LANA metal hydrides for this application was that pure tritium was always delivered to the loading process (Nobile 1991). In traditional tritium pumping and loading applications, if the tritium stays in the system for a prolonged period of time, some of the tritium is converted to 3-helium. Tritium naturally decays to 3-helium, a non-radioactive form of helium gas at a rate of 5.5% per year. Though this process of radioactive decay is small, over an 8-hour shift, it can produce enough helium to effect the purity of the tritium. The LANA, metal hydride is able to retain all of the decay helium and release virtually pure tritium on demand.

Later in the mid 1980s, another metal hydride material was added to the SRS loading system to create a higher pressure, or second stage, compressor. The entire system was very simple and reliable with no moving parts other than valves.

Another early application of metal hydrides in the tritium facilities was as a pump/separator. In 1987, a metal hydride pump/separator was put in service to provide pumping to the cryogenic distillation columns, as well as to purify inert gasses (i.e., helium, argon, nitrogen)

from the hydrogen isotope stream. Prior to the use of the pump/separator, the feed system to the cryogenic distillation columns, which are used to separate the different isotopes of hydrogen, always operated above ambient pressure. This was done to ensure that no gases such as nitrogen and oxygen could leak into the system. Since the cryogenic distillation columns operate at very low temperatures, any gases leaking into the system, other than hydrogen, could freeze out in the line and plug up the columns. The pump/separator used a very low-pressure hydride (palladium) that was able to efficiently separate the hydrogen isotopes from the other gasses and deliver the pure hydrogen isotopes directly to the cryogenic distillation columns. The pump/separator was also able to do this safely from tanks that were below atmospheric pressure, thereby improving the overall performance of the system (Nobile 1991).

Following the successful adaptation of metal hydrides in the existing facilities, a new tritium facility, based on more extensive use of metal hydrides, was planned. The new facility, originally called the Replacement Tritium Facility (RTF), began construction in 1986. The RTF was designed to take advantage of the latest technology to enhance operational safety, increase safeguards and security, and to minimize tritium releases to the environment. The facility was located underground to help prevent unauthorized entry. Thick, reinforced concrete outer walls combined with redundant safety systems provided protection against natural disasters (i.e. tornadoes and earthquakes) and assured that the facility could be safely shut down with no threat to the environment. Other technological improvements included using nitrogen gloveboxes to provide secondary containment for the tritium processes, and "dry" (oil and mercury free) pumps to eliminate generating a major mixed waste stream. The RTF also introduced laser cutting to replace a mechanical shearing systems for unloading tritium reservoirs and a state-of-the-art computer-based control system to improve product quality and process operations (Motyka 1992).

Of all the improvements in the RTF, the most significant new technology introduced was metal hydride technology. The use of metal hydrides to store isotopes in place of tanks substantially reduced the size of the overall facility, thereby significantly lowering the cost of the project. The size reduction also facilitated confining the process equipment into gloveboxes, thereby minimizing atmospheric releases. Other hydride applications for pumping, separating, purifying, and compressing hydrogen isotopes not only simplified process operations, but also improved the reliability of many of the plant operations.

The 1990s

The RTF became fully operational in 1994. To date all of the metal hydride applications introduced into the facility have met or exceeded all expectations. Metal hydrides were integrated throughout the entire gas handling process. In the RTF, metal hydrides are used to separate, store, compress, and purify hydrogen isotopes. A variety of pure metal and metal alloys were selected to meet the facility operating requirements. Each of these materials and applications was tested and evaluated by the Savannah River Technology Center (SRTC) prior to introduction in the RTF. Samples of metal hydride material were put into a long-term test program to ensure that the materials performed as expected over time. Both small- and large-scale performance tests on each of the metal hydride applications in the RTF were evaluated as part of the development program. Also a large pilot-scale test program, which integrated many of the individual hydride applications to determine how they would work together, was undertaken. The facility used for these integrated tests was also used to help instruct and train operators and engineers prior to RTF startup. The development and testing of the metal hydride systems by SRTC personnel played a major role in the successful startup and operation of the RTF.

Following the RTF, new tritium programs resulted in the additional demand for metal

hydride technology. In the mid 1990s, the tritium facilities needed a new method for safely and compactly storing tritium for prolonged periods of time, 7-10 years. A new Hydride Storage Vessel (HSV) was developed to address this problem. The HSV used a form of titanium as the hydride material. Using this material allowed the excess tritium to be stored at very low pressure, which eliminated the need for an external cooling system to remove any of the tritium's heat of decay. The use of metal hydrides in this application provided a safe and compact solution to the long-term tritium storage problem and avoided the use of more dangerous and more expensive high-pressure gaseous storage.

Also in the mid 1990s, the tritium facilities became involved in several new projects. One of these was the Tritium Facility Modernization and Consolidation Project. The goal of this project, which is referred to as Tritium Consolidation, was to reduce the overall physical size of the tritium facilities and to upgrade and modernize its capabilities. Again, to achieve the project goals, new metal hydride applications were required. A new storage bed design was developed that eliminated the need for a forced heating and cooling system, thereby minimizing the size of the facility. Another process improvement was the introduction of another class of metal hydrides, often referred to as "getters". These materials have been developed to remove small amounts of impurities from gas streams. One of the major commercial uses is to provide extremely clean gas to the semiconductor industry. These getters are typically comprised of zirconium-based metal alloys. In Tritium Consolidation, these getters will be used to remove small amounts of water and hydrogen from nitrogen and other process flush gases. Finally, improvements to the metal hydride, isotope separation system, which separates tritium from the other hydrogen species, were also made. These improvements not only reduced the overall space required but also were estimated to save the project \$20

million.

A new Tritium Extraction Facility (TEF) is being constructed at SRS to process the tritium-bearing rods produced in a commercial-type nuclear reactor. This newly produced tritium will be stored in metal hydride beds similar to those developed for the Tritium Consolidation Project. The TEF will also use a new hydrogen separator, which will greatly reduce the amount of gas that will need to be processed in the facility. The metal-hydride-based separator will separate the tritium and other hydrogen isotopes from the byproduct gases, which is mostly helium. The purified hydrogen isotopes will be measured in accountability tanks and then sent on to isotope separation operations. The waste gas will be further purified to remove any residual tritium contamination and then released. The hydride used in this process is palladium, which is also used in many of the RTF separation processes. The design of the separator, however, has been substantially improved by eliminating a large auxiliary heating and cooling system, thus making the unit much more compact and efficient.

Benefits of Metal Hydrides for Tritium Applications

Many of the benefits of using metal hydrides for commercial hydrogen as well as tritium applications have been described above. These benefits include safe and compact storage as well as efficient hydrogen separation and purification. While many of these benefits have commercial importance, they are perhaps more important in tritium applications. The use of metal hydrides in the tritium facilities has led to a major reduction in the size of the process equipment, allowing the equipment to be easily contained and isolated in gloveboxes. Virtually all potential tritium leaks can be captured, and the release of tritium to the environment can be avoided. Another advantage of metal hydrides is enhanced safety. Hydrogen and tritium can be stored on metal hydrides at low pressure.

This provides an added measure of safety when dealing with tritium, which is not only extremely flammable, but is also radioactive. Simplicity is another advantage of hydride technology. Hydrogen isotopes can be transferred from one location to another, or even compressed, simply by heating and cooling the metal hydride materials. This permits simple and reliable designs for tritium equipment, often with no moving parts other than valves. Since most valves can be operated remotely, many of the hydride systems can be easily automated and controlled. This is a very important advantage in a nuclear material handling facility.

Future of Metal Hydrides at SRS

As long as there is need for a strong nuclear deterrent, SRS will continue to play a major role in providing the tritium needed to support the nuclear stockpile. To support this mission, SRS will continually need to modernize and update its tritium handling facilities. A major part of the future modernization will involve metal hydride technology. In recent years, however, many of the metal hydride scientists at SRS have begun to look elsewhere for metal hydride development opportunities. The down sizing of the nuclear stockpile, coupled with the completion of several major tritium initiatives, has allowed SRS scientists the opportunity to see if the metal hydride experience gained from defense work could be applied to many of today's environmental and energy problems.

Fusion

One area that can benefit substantially from the transfer of metal hydride technology is the major worldwide effort to develop nuclear fusion as a future energy source. The United States, Europe, Japan, and Russia have collaborated on fusion energy research and development programs. One major program, the International Thermonuclear Experimental Reactor (ITER), has a goal of providing a large-scale demonstration of a fusion power plant. The preferred fuel for a modern fusion power plant

is a combination of the hydrogen isotopes, deuterium and tritium (D-T). The product of a D-T fusion reaction is a tremendous amount of energy. Basically, the reaction is similar to what takes place naturally to power the stars and our own sun. The major byproduct is nonradioactive helium. The fusion reaction is not self-sustaining, like a fission reaction, and no long-lived fission products are produced.

For a fusion reactor to proceed, a substantial amount of tritium and deuterium will be required. Initially this tritium will be stored and eventually separated and recycled back into the reactor. Many of the processes being considered for these tritium operations are based on metal hydride technology. Metal hydrides offer the same safety and reliability benefits to the fusion program as they do for defense applications. Fusion scientists from around the world have developed several new metal hydride materials and applications. Japanese scientists have developed a new low-pressure, metal hydride material (zirconium cobalt) that has similar storage and operating properties to that of uranium metal. One of the major benefits of this new material over uranium is that it is less pyrophoric and much safer to use. Other metal hydride applications have also been proposed and evaluated. SRS and the other DOE organizations have actively supported fusion energy initiatives over the past 30 years. The fusion community can benefit immensely from long-term SRS experience with metal hydride and other tritium handling technologies. Most of the SRS value to the fusion program comes from its many years of safely handling tritium, which in a large part is due to its extensive use and reliance on metal hydrides. SRS can also directly benefit from support of fusion programs. Future fusion facilities will be using the latest state-of-the-art tritium handling equipment. Extensive research and development on new and better ways to handle and process tritium will be required for fusion facilities to operate safely and efficiently. Therefore SRS is in good position not only to contribute its expertise but also to learn from these future tritium facilities.

Hydrogen Energy

While the world waits for the development of future energy sources like fusion, a more near-term energy solution will be required. The U.S. dependence on foreign oil has increased substantially since the oil shortages of the early 1970s. The U.S. now imports more than half of its annual demand for oil. The energy problem is compounded even further on the world scene, where increasing development in third world countries will place an even greater demand on limited world energy supplies. How long will the world energy reserves last when the billions in Asia and other parts of the world demand the same automobiles, air conditioners, and home appliances found in the industrialized nations? Further complicating this issue is the fact that the majority of the world's oil reserves are located in politically unstable regions.

The debate on how long our energy supplies might last will continue but another perhaps even more important issue remains—*pollution*. Most of the major cities in the U.S. have serious and even life threatening air pollution and smog problems. When combined with the right atmospheric conditions, smog can seriously effect the health of the elderly and the very young in our cities. The situation around the world is much worse. Many major metropolitan areas in other countries are now restricting traffic and industrial operations during peak times of the day in an attempt to reduce the level of air pollutants. Another issue that also needs to be considered is global warming. While scientists continue to debate the extent of this problem, it cannot be disputed that the amount of carbon dioxide and other greenhouse gasses in the atmosphere has increased dramatically over the past century. This increase coincides with the increased use of fossil fuels. For these and many other reasons, alternative fuels need to become a larger part of the energy make up of this country and the rest of the world.

Many of today's potential energy alternatives—such as wind, solar, geothermal, hydro, and nuclear—produce only electricity. Storage of this

electricity has been a problem. The development of advanced battery technology to store excess electricity has been disappointing. A better energy carrier is required to convert this electricity to something useful to meet our transportation needs. Hydrogen is the leading candidate. Hydrogen is the most abundant element in the universe, and, when used as a fuel with oxygen, it produces pure water as a byproduct with no pollutants. Very little pure hydrogen exists naturally on earth. Hydrogen on earth is found in the form of water or hydrocarbons such as oil, natural gas, and other organic materials. Most of the hydrogen produced today for industrial uses comes from processing natural gas and other hydrocarbons. In the future most hydrogen will come from electrolysis or photochemical reactions that convert water to its basic parts, hydrogen and oxygen. The hydrogen that is produced can then be used as a fuel in a direct combustion engine or in a battery-like device called a fuel cell.

One problem, however, still remains. How will we safely and efficiently store the hydrogen? One solution is metal hydrides. The same technology that SRS used to safely and efficiently store tritium and the other hydrogen isotopes for over 20 years can now be used to help the nation's and the world's energy problems. The ability of metal hydrides to store hydrogen compactly and at low pressures make them an ideal candidate for hydrogen storage systems on board future vehicles.

Dual-Use Hydrogen Technology

SRTC, funded by the Department of Energy's Office of Energy Efficiency and Renewable Energy (DOE-EERE), has recently developed several metal hydride-based systems for hydrogen vehicular applications. In April 1997, a hydrogen-powered bus called the H2Fuel Bus first rode down the streets of Augusta, Georgia. The H2Fuel Bus was a large-scale, demonstration project that enlisted the combined talents and efforts of commercial companies, academic

institutions, and SRS engineers and scientists. The overall goal of the project was to develop a hydrogen-powered vehicle that employed SRS metal hydride technology. The metal hydride system onboard the H₂Fuel Bus performed better than expected as it safely stored and delivered hydrogen to the bus's internal combustion engine during its testing and operational phases.

Another multi-partnered project, this time using a fuel-cell vehicle, was initiated in 1998. Fuel cells are a promising new technology that functions like a hydrogen-fueled battery. The advantage of a fuel cell is that it can generate power continuously as long as it is supplied with a source of hydrogen. Therefore, the fuel cell itself never loses its charge and does not need to be recharged. The goal of this project was to demonstrate and to eventually commercialize an industrial fuel-cell vehicle. The vehicle could be used in various indoor locations such as airports and warehouses to replace current electric-battery vehicles. The combined fuel cell-metal hydride vehicle can significantly outperform battery-powered vehicles and eliminate the harmful exhaust associated with gasoline-powered vehicles. A large part of the safety and reliability of the system is a result of using the SRTC-developed metal hydride system to store the onboard hydrogen.

Other projects currently underway at SRTC involve the use of fuel cell-metal hydride technology in mining vehicle applications. Increased regulation on the levels of carbon monoxide and diesel exhaust particulate in underground mines has forced the mining industry to pursue both battery and trailing cable electric alternatives. The performance of these alternatives, especially in very deep and long mines, has often been found to be unacceptable. A fuel cell-metal hydride alternative can be the solution by providing the range and power of a diesel vehicle without the harmful exhaust. Safe and low-pressure storage on solid metal hydrides will be the only acceptable hydrogen storage method for underground mines. SRTC is working with several commer-

cial and academic partners in leading this effort toward a clean and efficient alternative vehicle for the mining industry.

The projects described above, along with other similar projects supported by SRS and DOE, have not only made an impact on our nation's energy future but have also helped to maintain the critical skills needed to support our nation's long-term defense mission. SRS scientists and engineers have been able to maintain their 50 years of expertise in the area of hydrogen/tritium storage and separation by continually taking on new challenges and opportunities. SRS is also better able to attract and retain the best scientific talent available by providing them with a stimulating and varied work environment. The benefits of supporting "dual-use", hydrogen/tritium technology at SRS cannot be overstated. Various improvements in recent tritium storage and separations systems at SRS came about as a direct result of technology developed for hydrogen energy applications. This has led to substantial cost savings and improved operations at the SRS Tritium Facilities.

Summary

The use of metal hydrides for storing and handling tritium and hydrogen isotopes has dramatically improved the overall safety and efficiency of the SRS tritium operations. Metal hydrides continue to be used in today's tritium facility to store, separate, purify, and compress hydrogen isotopes. The ability of metal hydrides to compactly store tritium at low pressure greatly reduces the potential for atmospheric releases. The simplicity of metal hydrides also improves the efficiency of many of today's tritium operations and helps lower operating costs. Metal hydride technology can offer similar benefits to future fusion and hydrogen energy systems. With the continued support of SRS and DOE, scientists at SRTC can develop new solutions to future hydrogen technology problems, thus, allowing SRS to maintain its critical tritium and hydrogen expertise well into the next millennium.

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Biography

Theodore Motyka received a BS degree from the Newark College of Engineering in 1973, a MS degree from Cleveland State University in 1975, and a Ph.D. from the University of Colorado in 1979 all in chemical engineering. In 1980 he joined the Savannah River Laboratory as a research engineer. Since 1989, Mr. Motyka has been a manager in the Hydrogen Technology Section of the Savannah River Technology Center supporting the development of tritium and hydrogen storage and separation processes. He and his group have numerous papers and hold several patents in this field. In the last two years, Mr. Motyka has been actively involved in the development and demonstration of hydrogen energy technology.

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