Hydrogen as a Fuel and Its Storage for Mobility and Transport

Louis Schlapbach, Guest Editor

Abstract

This brief article describes the content of this issue of *MRS Bulletin* on Hydrogen Storage. Hydrogen is a powerful, clean, synthetic fuel with the inconvenient property of being an ideal gas under ambient conditions. In order to use hydrogen efficiently as a fuel, compacting it for mobile storage is a key issue. As an introduction to the following seven contributions on different storage techniques and their potential, we start with a description of the technical and socioeconomic aspects of the mobility and transport issues involved and present an overview of volumetric and gravimetric storage densities for hydrogen.

Keywords: fuel cells, energy density, hydrogen storage, liquid hydrogen, metal hydrides.

Introduction

In industrialized countries, one-third to one-half of the energy generated annually is used to power buildings and another third is used to move people and goods. As energy is rather inexpensive to produce, the applied energy technologies are often in the low-technology, low-efficiency range. The need to move people and goods continues to increase with increasing globalization, but the sustainability and safety of the energy supply to meet these needs are questionable. In terms of mass and distance, transport by air consumes the most energy, followed by road travel, then rail.

Vehicles can be linked to a power line with a continuous supply of energy (electric trains) or they can carry their energy or produce it on board (trucks, cars, bicyclists, airplanes, submarines). Combustion and electricity are the main methods of supplying energy for mobility and transport. Will this change in the future?

Many other high-energy-density, reversible, and controllable chemical reactions may be considered when shortages in the energy supply or environmental issues force us to look for alternatives. Thermally or electrochemically driven

processes forming oxides, halides, nitrides, carbides, or hydrides with elements and compounds of Periods 1, 2, and 3 should be evaluated.

But we dream of another solution: hydrogen as a synthetic fuel.

Why Hydrogen?

Hydrogen¹⁻³ is the most abundant element in the universe (75 at.%). On Earth, we find it in small quantities in air and, of course, in unlimited amounts but chemically bound in H₂O. It is a nontoxic and highly volatile gas. In the area of energy technology, we find hydrogen (1) in gaseous, liquid, or solid hydrocarbons; (2) in nuclear fission and fusion processes; and (3) as synthetic fuel produced by the dissociation of H₂O or hydrocarbons using primary energy (coal and other fossil fuels, electricity or heat from nuclear or renewable energy, photons).

Hydrogen is the simplest element in the periodic table, with just one electron. As a gas, it exists in the form of molecular H₂, which liquefies at 21 K and solidifies at 14 K. Solid hydrogen is a molecular insulator. Under high pressure, it should transform into an atomic solid, the simplest

metal, and possibly a room-temperature superconductor.

The oxidation of hydrogen, that is, its combustion with air to form water, is a very attractive exothermic chemical reaction, with two main advantages over other processes:

- It is a clean combustion of a nontoxic fuel: no CO₂ is produced, and when lean hydrogen-air mixtures are used, NO_x formation is prevented.
- The delivered energy per mass of hydrogen is very high, due to its electron per nucleon ratio, which is the best of all elements, and the high energy gain per electron.

Why are we not using these advantages of hydrogen for our energy needs today? Because the problems of production and particularly of storage have not yet been solved.

The traditional town gas, made from burning coal and water, contains about 50 vol% hydrogen, so the use of hydrogen as a fuel source is familiar and should not raise undue concerns. Electrolysis would be the appropriate production method for hydrogen, where electric energy is available. Thermochemical cycles have been abandoned, and photochemical production is not a mature technology.

The storage of hydrogen is a crucial issue for mobility and transport applications. Gravimetric and volumetric hydrogen-storage densities are summarized in Figure 1. Safe and comfortable modern cars consume 5–6 kg of fuel per 100 km in a combustion engine; in the future, a hybrid fuel cell/electric motor could reduce the fuel consumption to around 3 kg per 100 km. Taking into account that the energy per mass of hydrogen is 2.5–3 times higher than that of conventional liquid hydrocarbons, we need to store about 8 kg of hydrogen to drive 400 km in the combustion mode (or 4 kg of hydrogen in the fuel-cell mode). Since 1 kg of hydrogen has a volume of 11 m³ at room temperature and atmospheric pressure, a tank with considerable storage space would be needed.

This issue of *MRS Bulletin* is dedicated to various techniques for compact storage of hydrogen gas—classical ones like high pressure or liquefaction as well as sorption on high-surface-area solids or bulk sorption, and newer chemical processes that still suffer from the lack of reversibility. None of the presented solutions is economically competitive yet with today's inexpensive liquid hydrocarbons. However, as we are looking for non-CO₂-producing solutions, we should compare the hydrogen solution with other alternatives.

Following this introduction, Irani describes conventional steel high-pressure

Figure 1. Stored hydrogen by volume and by mass. The graph shows volumetric hydrogen density ρ_v versus gravimetric hydrogen density ρ_m for various materials to store hydrogen by bulk absorption, surface adsorption, or compound formation; dec. is the decomposition temperature, m.p. is melting point, and b.p. is boiling point. The container is not taken into account.

tanks and modern composite and fiber-reinforced aluminum tanks for hydrogen storage. It is noteworthy that Japan recently changed legislation concerning high-pressure gas tanks and now allows them on the roads. A problem with high-pressure tanks is, of course, that they deliver hydrogen at varying pressure instead of the constant pressure needed for transportation typical for applications.

Techniques and advantages of liquid hydrogen and its storage in superinsulated vessels is the topic of the contribution from Wolf. The most extensive tests of this technique so far have been performed using a fleet of city taxicabs in Stuttgart, Germany. The rather high energy loss that results from the liquefaction process is, of course, a concern.

More focused on materials science are the next three articles concerning the reversible process of solution of hydrogen in metallic alloys and metal hydride formation. Bowman and Fultz describe hydride formation through gas-phase absorption. Joubert et al. discuss electrochemical hydride formation for storage of electricity (the Toyota Prius hybrid vehicle uses this

technology). The third article in this group, by Akiba and Okada, describes hydrogen bulk sorption by a new class of metallic alloys showing the highest sorption capacity in the lower-temperature range.

The initial excitement of potential hydrogen storage in carbon nanostructures is over.⁴ A critical description of findings in hydrogen sorption by carbonaceous species and ways to prepare these species on the laboratory scale and in larger amounts is covered in the article by Züttel and Orimo, along with an illustration of efforts to extend the storage range using, for example, carbon magnesium composites.

Finally, as sometimes happens in scientific and technological progress, we look back at existing materials systems to apply them to new technologies, in this case, borates of light metals, alanates (complex aluminum compounds), and others, which have been known for more than 50 years for their high hydrogen content. Until 1996, they were considered far too stable to reach reversibility in an everyday energy process; then it was shown that the individual roles of thermodynamics and kinetics had not been interpreted correctly:

catalysts were developed that brought reversibility close to the levels required. Bogdanović and Sandrock describe this new approach in their article.

What characterizes a good hydrogen-storage unit for mobility and transport applications? Safety, reversibility in a temperature range of the order of 0–100°C, and high hydrogen concentration to provide an empty storage mass in a reasonable proportion to the total mass of the vehicle are required.

The U.S. Department of Energy (DOE) has set a goal that the fuel component should be 6.5 mass% of a total hydrogen-storage system (containment plus fuel). If energy efficiency is a rationale, then a different definition would have to be applied. For cars, the aim is to move a person with some amount of luggage (e.g., 100 kg mass) in a safe and comfortable way and reasonably rapidly from point A to point B. Modern 1000-kg automobiles are capable of doing this. One could set a limit of maximum energy consumption for that transfer (e.g., 1–2 kg of hydrogen per 100 km) and then define that the fuel component of the storage system must reach 5–10%, for example. As long as DOE-supported projects define a “mid-sized car” as a 2500-kg vehicle, the 6.5% target makes no sense and is certainly not innovative.

While progress is being made, an ideal technology for hydrogen storage is not yet available; without it, hydrogen will not become a major fuel for mobility and transport.

For more detailed reviews of this subject, we refer readers to References 5–7.

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Hydrogen Storage: High-Pressure Gas Containment

R.S. Irani

Abstract

This review outlines the prospects for gaseous hydrogen, stored in high-pressure cylinders, as a fuel for automotive applications. Following an initial description of hydrogen embrittlement problems encountered in the past in steel cylinders, the article explores the use of other types of gas cylinders, including the recent examples of hoop-wound and fully wound composites. Central to the article is the concept of the volume of hydrogen transported for the minimum amount of container weight. Finally, the role of international standards in developing safe and efficient cylinders for this application is emphasized.

Keywords: carbon-glass hybrids, composite cylinders, hydrogen embrittlement, hydrogen storage, International Organization for Standardization (ISO), liquid-metal embrittlement, ultrasonic testing.

Introduction

Hydrogen has been used for several decades as a fuel, albeit primarily in its cryogenic (liquid) form in the space program. The role of hydrogen as a fuel for transportation is now gaining importance because of its clean by-products and good performance when used in internal-combustion engines.

The use of gas cylinders for the storage of hydrogen fuel for transportation is a relatively recent application. This article aims, initially, to map the development of cylinder materials and the associated technology relevant to the containment of gaseous hydrogen. Based on this platform of best operating practice, the rest of the article then concentrates on the specific application of certain types of materials and construction methods for such storage containers. Safety apart, the single most important technical factor for this application is the relative weight of the container to that of the hydrogen being contained. This concept is developed fully in the article.

In the Beginning

The first known large-scale use of stored compressed hydrogen was in the early 1880s, when British expeditionary troops

used hydrogen, transported in wrought-iron welded vessels, to inflate war balloons in Asia and Africa. These vessels weighed in excess of 500 kg, which reflected the thick cylinder walls necessitated by the low tensile strength of the wrought iron (about 350 MPa); their gas capacity was only 25 m³. This represented a technical efficiency for the vessels of just 0.05 m³/kg, as measured by the volume of hydrogen carried per unit weight of the container.

Subsequently, cylinder technology has moved rapidly, with advances in both the materials of construction and manufacturing technology.

Early Material Developments

With major developments in steelmaking technology in the early to mid-20th century, the technical efficiency of hydrogen cylinders rapidly increased. The earlier high/low carbon steels were followed by carbon manganese steels in the 1940s, only to be replaced by low-alloy steels with additions of about 1% Cr and 0.2% Mo in the mid-1960s. At each step, the technical efficiency of the resulting cylinders rose, so that by the time the Cr-Mo steel cylinders were being employed, the value had risen from the original 0.05 m³/kg to almost

0.13 m³/kg (i.e., a 160% increase in the carrying capacity of the hydrogen container). The exact value varied, depending on which design code was in force locally for the construction of the cylinder shell.

Hydrogen Embrittlement

In the late 1970s, progress in technical efficiencies for hydrogen vessels came to a sudden halt. The culprit was the familiar hydrogen embrittlement mechanism, well known in other applications, which until that time had not reared its head in the area of hydrogen transport. The mechanism for the more than 100 failures that occurred was even more puzzling, since only vessels that experienced frequent (i.e., several times per week) hydrogen filling/discharging operations were affected; vessels that were only filled a few times a year did not fail. In other words, a fatigue-failure mechanism was in operation. Although the pressure vessels had undergone a strict testing regime at the prototype stage, which included a fatigue test, the fatigue test conditions stipulated only a hydraulic test and not a pneumatic one, let alone testing with hydrogen. This omission was identified by Pressouyre, who recommended that test methods should reproduce as closely as possible the in-service conditions, since micro-structural characteristics within a steel that may be acceptable for one gas service may be unsuitable for other gases.¹

The failures, which occurred throughout Western Europe, triggered a number of significant research investigations, many of them funded by national governments in light of the potential public-safety risks generated by the leakage of hydrogen.

By the early 1980s, results from these independent studies attributed the main cause of the failures to the high tensile strength of the steels. It was therefore significant that the failures were concentrated in Western Europe, where the quest for high technical efficiencies had resulted in the use of high-tensile steels, in contrast to other parts of the world, notably North America, where "softer" steels were in use.

In parallel with the concept of high tensile strength, the role of localized stress was also considered to be important, since many of the cylinder failures were in the so-called knuckle area of the cylinder (the region of the cylinder where the uniform parallel wall gives way to the thicker base area of the cylinder). In cases where there was an abrupt change in the profile, possibly coupled with minor manufacturing blemishes on the internal surface, this local stress concentrator became a failure-initiation site.