INVESTIGATION OF HYDROGEN STORAGE IN IDEAL HPR INNER MATRIX MICROSTRUCTURE USING FINITE ELEMENT ANALYSIS

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Babu Gopalan

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by

BABU GOPALAN

has been approved for

the Department of Mechanical Engineering

and the Russ College of Engineering and Technology by

Hajrudin Pasic

Professor of Mechanical Engineering

Dennis Irwin

Dean, Russ College of Engineering and Technology

ABSTRACT

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Studies have proven hydrogen gas as a highly efficient, renewable and alternative energy source and it is expected to serve as a common fuel for all mobile and stationary applications. However, currently the on-board storage difficulties prevent the practical usage of hydrogen in automotive applications. A more efficient and innovative method of hydrogen storage for automotive fuel cell application is to compress hydrogen in minute hollow spherical bubbles incorporating the Hydrostatic Pressure Retainment (HPR) technology. In a HPR vessel, the material properties and the inner matrix structure are two critical design parameters that determine the hydrogen mass efficiency. The focus of this study is devoted to investigating the performance characteristics of one configuration; spherically shaped bubbles homogenously arranged in a simple cubic inner matrix packing structure for a HPR vessel, using Finite Element Analysis.

Approved:

Hajrudin Pasic

Professor of Mechanical Engineering

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CHAPTER 1 INTRODUCTION

This chapter introduces the concept of Hydrostatic Pressure Retainment (HPR) vessel technology for onboard hydrogen storage and summarizes the objectives of this thesis. A general outline of the thesis is also given.

1.1 Overview

Presently, compressed hydrogen storage tanks for automotive application uses a void volume cylindrical vessel. At ambient conditions (300 K, 0.1 MPa), the energy density of hydrogen is very low (0.0107 kJ/m³) when compared to gasoline (0.0316 kJ/m³). To increase the energy density hydrogen has to be pressurized to high levels such as 69 MPa (10,000 psi). Therefore hydrogen storage tanks require thick-walled cylinders to withstand high pressure ranges desirable for automotive purpose.

It is estimated that automotive applications require vessels to withstand pressures around 34.5 MPa (5000 psi) to achieve a minimum driving range of 160-241 km (100-150 miles) [1]. The U.S Department of Energy has estimated a storage capacity of 5-6 kg of hydrogen for a passenger car to achieve a range of 480-560 km (300-350 miles) [5]. This requires much higher levels of compression. Prototypes of fuel cell vehicles are continuously being developed and tested by major automotive companies. These vehicles use void volume vessels where the outer wall acts as the structural support and all of the pressure induced stresses are carried by this outer wall. This necessitates thick walls and adds to the weight of the vessel. In order to reduce the total weight of the vessel the focus is on advanced light weight materials that can withstand high pressures and at the same time contribute towards weight-reduction. A recent development is the Honda FCX vehicle. This vehicle stores 3.75 kg of hydrogen at a pressure of 34.5 MPa (5000 psi) in two large cylindrical tanks with a combined volume of 156.6 liters [2]. This configuration achieves a driving range of about 430 km (270 miles) [2]. The outer wall in this tank is constructed with three layers, an aluminum liner, a carbon fiber layer and a glass fiber layer [2]. These tanks are placed in the rear end of the car between the two wheels. Higher driving ranges require tanks to withstand pressures up to 69 MPa (10,000 psi) [1]. Advancement for improved hydrogen storage tanks is continuously being made.

1.2 HPR definition

HPR technology is aimed at developing improved hydrogen storage tanks for automotive applications. A HPR vessel involves compressing hydrogen gas within small spherical bubbles. The bubbles are arranged within a solid mass similar to a foam-like structure. By compressing the gas within the small bubbles, a near- hydrostatic (tri-axial) tension is induced in the structural material between the adjacent bubbles. The shape of the bubbles and the bubble packing arrangement within the solid mass are referred to as the inner matrix structure. The bubbles containing the gaseous hydrogen can be of any shape, geometry (i.e., spherical bubble, polyhedral) and can take any bubble packing arrangement. When the bubbles are arranged to simulate a simple cubic, body centered cubic or a face centered cubic packing structure, the resulting inner matrix structures are referred to as ideal HPR inner matrix structures. The ideal HPR inner matrix structures contribute towards homogeneity and uniformity in the bubble arrangement and are preferred over random bubble arrangement in a HPR vessel. The structural material between the adjacent bubbles acts as a network of load bearing struts. Through proper configuration of the inner matrix structure, nearly all of the pressure-induced stresses can be retained within the inner matrix resulting in significant structural efficiency advantages. This inner matrix structure is then attached to a conformable outer surface for the purpose of carrying a portion of the pressure induced stresses. This approach is referred to as "Hydrostatic Pressure Retainment".

1.3 Spherical bubbles for HPR inner matrix

An important factor in designing an HPR vessel is to maximize the occurrence of near-hydrostatic tension within the inner matrix structure. In 2004, Robert J. Setlock proposed that spherical bubbles of uniform size maximize the occurrence of tri-axial tension within the HPR inner matrix [3]. Earlier in 2002, FEA were conducted on simple models with one spherical bubble and multiple bubbles [4]. The results of this study confirmed regions of near-hydrostatic tension in the structural material between the bubbles. Also, it was observed that the spherical bubbles are surrounded by a circumferential region where the stress was much higher due to induced shear stresses. Similar observations were made in models containing multiple spherical bubbles. This region was defined as the angular distortion zone [3]. Figure 1.1 and Figure 1.2 show the von Mises stress plot results from previous studies performed by Robert J. Setlock.



Figure 1-1 Angular distortion zone for in a single spherical bubble [3]

The angular distortion zone in Figure 1-1 shows a circumferential region surrounding the spherical bubble where the stress is much higher than in the outside the zone.



Figure 1-2 Interacting angular distortion zones in multiple bubbles [3]

Figure 1-2 is a case of multiple spherical bubbles. It shows circumferential regions of high von Mises stress (15 MPa) surrounding each spherical bubble (angular distortion zone) similar to the case of one bubble. Regions farther from the angular distortion zone

are in a state of near-hydrostatic tension. The von Mises stress in this region is typically negligible and measures between 0.10 MPa and 0.13 MPa (near-hydrostatic tension). The maximum von Mises stresses (18 MPa) occur in the region where the angular distortion zone of one bubble interacts with the angular distortion zone of the surrounding bubbles (interacting angular distortion zones) [3].

Based on the FEA results and his algebraic analysis Robert J. Setlock developed a general relationship between a bubble sphere radius and the angular distortion zone surrounding that spherical bubble. He concluded that the radius of the angular distortion zone (R_{ad_zone}) surrounding a spherical bubble can be related to that bubble sphere radius (R_{bubble}) by the equation given in Equation 1.1.

$$R_{ad_zone} = 1.2247 R_{bubble}$$
 1.1 [3].

These results indicated that bubble packing arrangement and the spacing between bubbles are significant parameters in maximizing the occurrence of near tri-axial tension within HPR inner-matrix. Nevertheless, application of the HPR concept for compressed hydrogen storage is further investigated in this thesis.

This thesis has the objective of analyzing the performance characteristics of a simple cubic HPR vessel for the application of compressed hydrogen storage through FEA and simple calculations. Spherical bubbles of uniform size are considered for the bubble geometry. The vessel model analyzed in this study is cube shaped. When the bubbles within the vessel are packed to simulate a simple cubic structure it is referred to as simple cubic HPR vessel. The vessel geometry and CAD modeling are discussed in chapter 4.

Practical usage of HPR vessels requires bubble interconnectivity or open cell inner matrix structure for free flow of hydrogen gas between the bubbles. Simple cubic inner matrix vessel investigated in this thesis is a closed cell structure. Hence it necessitates the use of hydrogen permeable structural material for the inner matrix. Metals are not considered as a suitable option for HPR vessels due to their non-permeable nature. However, hydrogen permeable polymeric foams that exhibit microstructures that are close to ideal structures are best suited for HPR vessel applications. [6].

1.4 Mass efficiency for compressed storage tanks

The aim of this thesis is to study the feasibility of simple cubic inner matrix HPR vessel for compressed hydrogen storage. For this purpose, hydrogen mass efficiency is chosen as a reference parameter. Throughout this study, hydrogen mass efficiency refers to the ratio between the mass of hydrogen stored in the HPR vessel and the total mass of the hydrogen-filled vessel. The U.S Department of Energy has set a short term target (2005-2010) of 6 % mass efficiency and 62 kg H₂/m³ for an acceptable driving range of 560 km [5]. The long-term target (2010-2015) is set for 9 % mass efficiency and 85 kg H₂/m³ [5]. The current storage systems offer mass efficiencies between 3% and 4.5% [5].

Using FEA tools and simple calculations it is possible to numerically determine hydrogen mass efficiency for the HPR vessel. Thus the feasibility of simple cubic HPR vessel for compressed hydrogen storage applications can be systematically studied with reference to mass efficiency.

1.5 HPR vessel advantages

HPR pressure vessels offer three major advantages over traditional hydrogen storage vessels.

HPR vessels need not be cylindrical or spherical in shape. Each bubble within the vessel acts as an independent spherical vessel. Since the gas is compressed within very small bubbles packed within the inner matrix structure, for larger tank volumes the tank can take more flexible shapes or geometries. Also the pressure induced stress is well retained within the structural material in-between the bubbles. Therefore, for large tank volumes the outer wall shape does not affect much on the stress distribution within the inner matrix [3]. Therefore the shape of HPR vessels can be customized to fit available space in the automobile. This adds more flexibility and versatility for on-board storage purposes.

By the definition of mass efficiency, reduced tank weights contribute significantly to higher mass efficiencies. HPR vessels offer weight savings when compared to conventional cylindrical and spherical pressure vessels [3]. Since a large portion of material within the inner-matrix is subjected to near-hydrostatic tension, the structural material in a HPR vessel is more efficiently utilized [3]. Robert J. Setlock developed a general relationship to compare the volume of structural material required to fabricate a traditional cylindrical pressure vessel and the volume of structural material required to fabricate a cylindrical HPR inner-matrix vessel, both having the same load bearing capacity. It was theoretically proved that the HPR inner matrix vessel needs only half the volume of the total structural material required to fabricate a traditional cylindrical pressure vessel, both having identical vessel configurations [3]. This contributes to significant weight savings in the overall HPR vessel.

Finally, HPR vessels are safer than the traditional cylindrical pressure vessels. In HPR vessels, the pressure-induced stress is distributed throughout the entire volume of the inner matrix within the HPR pressure vessel, rather than concentrated on the outer surface only. Since the load carrying material is evenly distributed throughout the inner matrix, it is safeguarded from the external surroundings [3]. In case of an accidental rupture, it follows that the new lightweight HPR vessels will feature a characteristic of non-catastrophic failure when compared with traditional vessels where the entire stress is carried on the outer wall. Also, the structural material in between the surrounding bubbles is pressurized on all sides contributing to a pressurized fuel tank. This reinforces the strength of the structural material within the inner matrix. [6].

1.6 Objectives

This section lists the objectives of this thesis. A brief explanation is given, stating its substance.

• Analyze the stress distribution within a simple cubic HPR vessel.

Studying the stress distribution within a simple cubic inner matrix is essential for two reasons. Firstly, the theoretical assumption of pure hydrostatic tension in a HPR structure can be verified only through FEA. Although FEA for simple models with one bubble and multiple bubble cases confirmed regions of near-hydrostatic tension, its true nature when extended to larger models needs to be investigated. Secondly, throughout this study hydrogen mass efficiency is calculated through a numerical FEM approach. Since the mass efficiency is a function of bubble size, the effect of bubble size on the stress distribution within the simple HPR structure needs to be analyzed.

• Derive an optimized bubble sphere size range for a simple cubic HPR vessel.

The hydrogen mass efficiency for a simple cubic HPR vessel is indirectly dependent on the bubble spacing. The total mass of hydrogen stored in the simple cubic HPR vessel depends on the maximum workable pressure (pressure to which hydrogen is compressed) and the useful hydrogen storage volume within the inner matrix. Bubble size has an effect on both these factors. Hence, the relationship between the bubble size and the hydrogen mass efficiency must be investigated to derive an optimum bubble size range that will contribute to maximum hydrogen mass efficiency.

The optimum bubble size derived from this study is not limited only to simple cubic HPR vessel geometry investigated in this study. Rather, it should aid in reasonably predicting an optimum bubble size range for a simple cubic HPR vessel of any given contour and large volume. To achieve this purpose, the optimum bubble sphere radius is defined as a percentage of half the distance between the centers of two closest bubbles lying on the same edge or bubble sphere touching radius. By fixing the bubble centre to centre distance, it is possible calculate an optimized bubble size range through the relationship developed from this analysis.

• Perform wall thickness optimization for the simple cubic HPR vessel model for maximum hydrogen mass efficiency.

From the definition of hydrogen mass efficiency, it is implied that reducing the structural mass of the HPR vessel contributes to higher mass efficiency. To achieve this, the wall thickness is reduced in steps from all the faces of the vessel. FEA is performed

after each step and the hydrogen mass efficiency is recalculated. This analysis is intended to maximize the hydrogen mass efficiency in the vessel by optimizing the outer wall thickness on all faces of the simple cubic HPR vessel model.

• Perform a material comparison to identify desired material properties suitable for simple cubic HPR vessels.

Material selection is an important parameter that influences the hydrogen mass efficiency. This study is important to identify desirable material properties that can help in further narrowing the search for the right material. Type 305 stainless steel and epoxy/carbon fiber composite are chosen for comparison. To make a meaningful comparison between the materials, the analysis is performed on the vessel configuration sharing the same optimized geometry (optimized bubble size and optimized wall thickness). It was already stated that metals are not a workable option for simple cubic HPR vessel due to their non-permeable nature. However a metal is chosen in this study for comparison and to gain an understanding on material properties that are suitable for achieving high hydrogen mass efficiencies. This study will help in narrowing the search for advanced material properties that would yield higher mass efficiencies for a simple cubic HPR vessel. This thesis report is organized in the following format.

CHAPTER 2: briefly describes the primary methods of on-board hydrogen storage.

CHAPTER 3: introduces the idea of metal foams. The application of foam structure in on-board hydrogen storage through HPR technology is discussed.

CHAPTER 4: This chapter briefly describes simple cubic bubble packing structure. It is followed by a brief introduction to two material candidates used in this study for investigating the mass efficiency of the vessel. Finally, a review of FEA tools and preprocessing steps on the vessel for analysis is presented.

CHAPTER 5: This chapter presents the results of this thesis. A detailed discussion on stress distribution within the simple cubic HPR vessel, bubble size optimization, wall thickness optimization and desired material properties for HPR structure are presented.

CHAPTER 6: This chapter presents the conclusions of this study. Recommendations for near future work are provided towards the end.

CHAPTER 2 HYDROGEN STORAGE FOR AUTOMOTIVE APPLICATIONS

The success of using hydrogen as an alternative fuel for driving vehicles depends on the proper distribution and storage of hydrogen. Hydrogen fuel exhibits the highest energy content compared to its weight and a very low energy content compared to its volume. Complete commercialization of hydrogen powered vehicles require the need for storage systems that can contain sufficient hydrogen onboard a car to match up with the conventional gasoline-powered vehicles. Hydrogen storage systems should account for inherent safety as well as high volumetric and gravimetric efficiency. The three principal technologies available for storing hydrogen are compressed storage, cryogenic storage and hydride storage. These methods are discussed in this chapter.

2.1 Compressed hydrogen storage

Compressed hydrogen storage offers the least complex method of storing hydrogen. Hydrogen has one of the highest energy density (120 MJ/kg) of all fuels. But the energy content of hydrogen per volume is very low (0.0107 kJ/m³) compared to all fuels at ambient conditions (300 K, 101 kPa). At ambient temperature and pressure (300 K, 101 kPa), 1 kg of hydrogen gas occupies 11 m³. Consequently, hydrogen storage implies the reduction of an enormous volume of hydrogen gas. This is accomplished by compressing the gas inside pressure vessels. Hydrogen gas stored at ambient temperature avoids costly and bulky thermal insulation.

Three main types of hydrogen storage tanks are [12]

- 1. Steel tanks
- 2. Aluminum core encased in fiber glass (composite)
- 3. Plastic core encased in fiber glass (composite)

Storage of hydrogen as a compressed gas in metallic pressure vessels contribute significantly to the total weight of the system. This leads to low gravimetric efficiency. Hence, composite polymer pressure vessels in combination with increased working pressure up to 68.94 MPa (10,000 psi) are used to achieve high volumetric and gravimetric efficiency.

The hydrogen gravimetric density in relation to its different working pressures and temperature is important to compare different hydrogen high pressure storage systems. To achieve high hydrogen mass efficiency, the hydrogen gas density in the vessel must be significantly higher than the weight and volume of the vessel. The basis for the hydrogen density calculation is the universal gas law for an ideal gas and is given in Equation 2.1.

$$Pv = RT$$
 2.1

P is absolute pressure, v is specific volume, T is absolute temperature and R is the specific gas constant. For an ideal gas, the pressure-volume deviation or the compressibility factor, k equals 1. But at higher pressures, the compressibility factor, k is slightly less than unity. At high pressures, the intermolecular distance between the gas

molecules become smaller. This increases the intermolecular force of attraction between the molecules accounted by Vander Walls equation given in Equation 2.2.

$$[P + \frac{an^2}{v^2}](v - nb) = nRT$$
 2.2 [7]

a and *b* are Vander Walls constants (different for different gases). Vander Walls equation compensates for the deviation of real gases from ideal gas at high pressures [7]. For real gases at high pressures, the adjoining molecules exert a higher force of attraction between one another. This will result in reduced interaction of the molecules with the vessel walls [7]. Therefore the working pressure exerted by any real gas under these conditions will be less than the pressure observed for an ideal gas [7]. At higher pressures k is less than unity (~ 0.9987) and this is taken into account in the equation for a real gas given in Equation 2.3.

$$\rho = \frac{P}{RTk} \qquad 2.3 [7]$$

The Beattie Bridgman equation [8] is an improvement over Vander Walls equation. This equation predicts the non-linearity in the amount of hydrogen that can be stored at higher pressures with increasing pressure. This equation is given in Equation 2.4.

21
$$p = [\frac{R.T.(1-\varepsilon)}{v^2}][v-B] - \frac{A}{v^2}$$
2.4 [8]

A, B and ε are Beattie Bridgman constants. The non-linearity of hydrogen gas at higher pressures is shown in Figure 2-1.



Higher density becomes increasingly difficult to attain with higher pressure. This trend is observed in Figure 2-1. It follows from the graph that hydrogen gas density at 68.94 MPa (10,000 psi) is 2/3 that of an ideal gas. Assuming technical feasibility and if pressure is doubled to 138 MPa (20,000 psi), the gas density increases by only 50% [9]. Hence 68.94 MPa (10,000 psi) is a useful maximum pressure for compressed hydrogen [9]. High pressure storage units are composed of tanks, outer shell, protective foam and gas control system, all in an integrated package [10]. To respond to variations in load gas flow control valves are positioned between the storage tank and the fuel cell. Figure 2-2 and Figure 2-3 show pictures of integrated high pressure hydrogen storage tanks.



Figure 2-2 Compressed hydrogen gas integrated storage system [10]



Figure 2-3 Composite hydrogen tanks [10]

2.2 Metal hydride storage

In a metal hydride storage system, hydrogen is stored in the form of metal hydrides. A hydride is a binary compound (chemical compound composed of only two elements) formed by the union of hydrogen and other elements. Metal hydrides are alloys that absorb and store large amounts of hydrogen by bonding with hydrogen and forming hydrides. Hydrogen storage in a metallic material involves the dissociation of hydrogen into its atoms. These hydrogen atoms are absorbed into the solid metal structure. This is schematically shown in Figure 2-4. The metallic hydrides are housed within a tank and are mounted onboard vehicles. Later, the hydrogen is released from the metal hydride tank by providing heat to the tank with the help of an integrated heating system. The chemical reaction between hydrogen and the metal alloy is given in Equation 2.5.

$$2M + XH_2 \leftrightarrow 2MH_r + Heat$$
 2.5 [11]

M is a metal or metal alloy.



Figure 2-4 Metal hydride formations [12]

Metallic hydrogen storage devices consist of hydrogen gas, the solid metal and an intermediate membrane. In-flowing hydrogen gas is adsorbed into the intermediate membrane and is split into its constituent atoms at the intermediate membrane. The atoms are then absorbed into the metal crystal structure. Here, the orientation of the hydrogen atoms in particular pattern with the metal atoms results in the formation of metallic hydride. In this process, flow of hydrogen gas from the external metal surface to the inner metal crystal structure directly controls the rate of formation and decomposition of metal hydrides. For a given volume, more hydrogen atoms can be packed into some metal hydrides than into the same volume of liquid hydrogen. This variation is dependent on the characteristic property of the material and the particular positioning of the hydrogen atoms with the metal atoms.

Reversible and Irreversible Hydrides

Hydrides are classified as reversible or irreversible hydrides. These are stored in a solid form or in a water-based solution depending on their classification. Reversible hydrides are solid-alloys or inter-metallic compounds. They release hydrogen gas under specific pressures and temperatures. The chemical reaction between a metal and hydrogen in the formation of a metallic hydride is given in Equation 2.6.

$$2M + XH_2 \leftrightarrow 2MH_r + HEAT$$
 2.6 [11]

M is a metal or an alloy.

In a reversible reaction, the absorbed hydrogen is released by the decomposition or transformation of its compound by extreme heat. This process is called Pyrolysis reaction. After the complete usage of hydrogen, reversible hydrides are refueled through a supply of pure hydrogen at a filling station to the original alloy.

Irreversible hydrides are compounds that undergo reactions with other reagents such as water, producing hydrogen and byproducts. The chemical reaction in the formation of an irreversible hydride is given in Equation 2.7.

$$MH_X + XH_2O \rightarrow M(OH)_X + XH_2 \qquad 2.7 [11]$$

M is a metal or an alloy and x is its valence. The reaction given in Equation 2.7 is termed as a hydrolysis reaction and is irreversible. After the hydride has released its hydrogen, the byproduct remains in the fuel tank. Unlike the reversible metallic hydrides, the hydrogen generation reaction is irreversible in an onboard vehicle under simple pressure and temperature changes. Recharging the tank with hydrogen requires the byproduct to be chemically regenerated under specific conditions. This is usually done at a central site or a chemical processing tank.

Metal hydride formation is an exothermic process. Heat is released while the metal powders absorb hydrogen to form metal hydrides. Between 10-25 % of the heating value of hydrogen gets discharged and this depends upon the hydride used [13]. Consequently, removal of heat from the metal particles results in efficient and quick recharging of the hydride bed. But this reduction in temperature quickly cools the hydride

bed and results in the stoppage of hydrogen flow. Hence, heat is added to the metallic bed to release gaseous hydrogen. This exothermic process is represented in Figure 2-5.



Figure 2-5 Absorption - desorption process in the formation of metallic hydrides [14]

This requires provisions for a heat exchanger in a practical hydride storage bed. The steam exhaust and waste heat generated from the fuel cell are transported back to the hydride bed as shown in Figure 2-6.



Figure 2-6 Schematic diagram of a hydrogen storage using metallic hydride [13]

Metallic hydride storage systems are highly compact in storing hydrogen and enables high volumetric energy density i.e., contains a large amount of hydrogen in a small volume. A metal hydride tank might carry 5 kg of hydrogen in one-third the volume of a 34.47 MPa (5,000 psi) tank. Conversely, weight capacities of the metallic hydride tanks are usually hindered by the use of heavy metals leading to maximum values below 1.5% by weight. For example, a robust tank carrying 6 kg of hydrogen is estimated to weigh more than 300 kg. However, significant improvement in weight capacities close to 3.6% is achieved by using light elements such as Magnesium [15]. The advantages and drawbacks of metallic hydride tanks over compressed storage systems are briefly discussed next.

Summary

1. In general, compressed hydrogen tanks are much lighter than equivalent metallic hydride tanks.

2. Hydrogen supply from a metallic hydride bed for an automotive fuel cell application is more complex than a compressed gas storage cylinder. This is due to the heat accompanied with hydriding / dehydriding, and the requirement of an internal heat exchange system for the discharge of hydrogen gas.

3. Compressed hydrogen storage methods require significant levels of compression on the order of 20 -35 MPa (3000-5000 psi). Energy loss is associated with high compression, adding to the cost and maintenance of high-pressure compressors. In contrast, metallic hydride compressors are charged at relatively low pressures of 0.3-3 MPa (43.5 – 435 psi).

4. Both compressed hydrogen and metallic hydride storage methods require pressure/flow control devices between the storage tank and the fuel cell to respond to load variations [see Figure 2-6 above].

5. Compressed hydrogen storage is associated with high levels of pneumatic energy and quick hydrogen loss. Hence, in case of an accidental rupture, a compressed hydrogen tank presents a potential safety hazard. In contrast, metallic hydride tanks operate at near atmospheric pressures and are self-limiting in case of a tank rupture due to the endothermic nature of hydrogen.

2.3 Liquid hydrogen storage

The change of state from hydrogen gas to liquid is accomplished by pressurizing hydrogen gas at its boiling point [38.15 K (-235 C)]. Consequently, storage of liquid hydrogen in onboard vehicles necessitates expending energy to pressurize the hydrogen. At the temperature of liquid hydrogen, many materials in contact with it become brittle and contract from their dimensions at room temperature. Also, the exposure of liquid hydrogen in an atmosphere of air will condense the oxygen in the air into the liquid hydrogen, presenting a high risk of explosion. For the above reasons, liquid hydrogen containment tanks are designed differently than the usual simple envelope used for storing fuels such as gasoline.

In a hydrogen molecule, each of the two hydrogen atoms holds one proton. Each proton carries a charge and the two protons take a spin in the same direction (referred to as orthohydrogen molecules) or in the opposite direction (referred to as parahydrogen molecules). The direction of the spin depends on the charge on the proton (Figure 2-9). Thus depending on the spin of the protons the hydrogen molecules exist as either orthohydrogen molecules or parahydrogen molecules. In the state of thermal equilibrium at room temperature, the dihydrogen contains 25 % of parahydrogen (nuclear singlet state) and 75 % of orthoghydrogen (nuclear triplet state). At the temperature of the boiling point [38.15 K (-235 C)] the equilibrium orthohydrogen concentration is 0.21% and the parahydrogen concentration is 99.79 %.



Parahydrogen molecule- Nuclear spins anti-parallel (opposite direction)



Figure 2-7 Hydrogen molecular form - orthohydrogen / parahydrogen

Liquefaction of hydrogen is an energy consuming process and requires compression and expansion of hydrogen in multiple phases. During the liquifaction process, hydrogen is converted from its "ortho" to "para" form. Orthohydrogen, is unstable at liquid hydrogen temperature and transforms to "para" hydrogen over time. This ortho-para conversion liberates heat and results in hydrogen evaporation within the storage vessel. This effect is counteracted with the help of a reactive cooling tower which removes the heat during the "ortho" to "para" hydrogen conversion.

Liquid hydrogen is stored in cryogenic storage systems and is highly-insulated to prevent heat leakage. Because of its extremely cold temperature, the equipment is designed and manufactured with a material suitable for extremely low temperature operation. A major concern associated with storing hydrogen at cryogenic temperatures is the heat leakage from the surrounding objects into the tank leading to the evaporation. Also, the seals of the container used for storing the propellants loose their ability to maintain a proper sealing at very low temperatures. To overcome these disadvantages, double layered walls (a dewar) with a vacuum region in between are used to reduce the undesirable heat leakage that occurs in liquid hydrogen storage.

Dewar's are non-pressurized vessels and are used for storing liquid hydrogen as a propulsion fuel in aerospace applications. A representation of a dewar vessel is shown in Figure 2-10. The design of a dewar minimizes the heat loss by three modes: conduction, convection and radiation. Liquid hydrogen is stored in an inner tank and it is encased by a vacuum to minimize the heat transfer by conduction and convection. The major portion of heat leak into the flask is in the form of thermal radiation. To reduce the heat transfer by radiation, a thin wall on the other side is maintained at the temperature of liquid nitrogen [77 K (-196.15 C)]. This shields the walls of the liquid hydrogen container and is insulated from the outermost shell of the dewar by another vacuum space. The reduction of the external temperature from 300 to 77 K reduces the contribution of heat gain by a factor of about 250 [16]. The dewar's are equipped with an array of valves, vacuum pumps and a dust cap at the outlet, for the gas produced from the vaporized liquid to escape.



Figure 2-8 Liquid hydrogen storage in dewar [17]

Another method of storing liquid hydrogen is the compressed liquid cylinders. These are highly pressurized spherical or cylindrical pressure vessels and are equipped with safety relief valves and rupture discs to protect the cylinder from excessive pressure build-up. A major drawback of liquefied hydrogen storage is the hydrogen boil.

Boil off

At extreme low temperature, liquid hydrogen vaporizes and escapes from the tank over time. Vapors contained within the tank increases the tank pressure and the pressure within the tank increases with increase in temperature. This pressure build-up increases the weight of the tank. To offset this, pressure relief valves are provided to avoid the tank from exploding. However, the usage of pressure relief valves to reduce the pressure inside the tank results in some propellant leakage from the tank. This is accounted to boiloff loss. Boil off rate is controlled by the amount of heat leakage and the quantity of liquid hydrogen in storage tanks. Hydrogen storage tanks are protected from heat to prevent the boiling off gases. The amount of heat that can leak into a storage vessel is related to its surface area. Thus reducing the surface area of the storage tank slows down the boil-off loss. Also, a hydrogen storage vessel completely poured out of liquid hydrogen contributes to boil-off losses. This warms up the storage tanks appreciably above 20 K and there is a large amount of boil-off when liquid hydrogen is reintroduced into the warm tank, again chilling it down to liquid hydrogen temperature. Hence, some amount of liquid hydrogen is routinely left in the tank to maintain a low temperature.

CHAPTER 3 TECHNICAL DESCRIPTION

This chapter introduces the general idea of metal foams. The application of a foam structure in on-board hydrogen storage through HPR technology is discussed.

3.1 Solid foam

Foam is a substance that is generally formed by trapping many gas bubbles in a liquid or solid. Foams are classified as liquid foams and solid foams.

Solid foam is a dispersion of gas in a solid. Solid foams are cellular materials and are made from a framework of solid material surrounding the gas-filled voids (bubbles). This makes solid foams extremely light. The low density of solid foams combined with other physical properties such as low thermal conductivity, makes them ideal for various applications. Examples of cellular material that are common in nature are wood, cork, coral and bone.

Solid foams have a hardened structure and essentially do not change over time unless put into extreme loading conditions. The choice of the parent material and the foam structure are the two important variables that determine the final foam properties. Polyurethane and polystyrene are examples of solid foams products used for cushioning, packaging and insulation. Polymeric foams are widely used for their low weight, sound and shock absorbing properties [18].

3.2 Metal foam

Metal foams originate from the solidification of liquid foams and have a restricted morphology. They have a complex structure and consist of a network of thin

plateau borders. The very low densities combined with their novel physical, mechanical, thermal, electrical and acoustic properties of metal foams finds potential applications in sound insulation, heat exchangers, filters and catalyst carriers. The open-celled, interconnected structure of metal foam is very similar to that of the cancellous bone material found in the human body. Figure 3-1 shows this structural similarity.



Figure 3-1 a) Cancellous bone



Figure 3-1 b) Metal foam sample

Figure 3-1 Structural similarities between cancellous bone and metal foam [19]

The properties of metal foam and other cellular structures depend upon the properties of the parent material, the relative density and cell topology (open or closed cell). Structural and load bearing applications use closed cells (Figure 3-2 a) while functional applications such as filtration, damping and compressed gas storage require bubble interconnectivity and necessitates open structures. (Figure 3-2 b).





Figure 3-2 a) Closed cell structureFigure 3-2 b) Open cell structureFigure 3-2 Metal foam structure [20]

In a closed cell metal foam each cell is sealed from its adjacent cells with the metal distributed in cell faces and the plateau borders. Closed cell structure provides high stiffness, strength and high impact force absorption characteristics in comparison with the actual parent material [20].

Open cell foam consists of ligaments that form a network of interconnected cells. The cells are randomly oriented and are mostly homogenous in size and shape. This depends on the manufacturing method used to create the metal foam precursor material. Open cell foams are not as stiff or as strong, as closed cell foams. But due to their ability in free passage of fluids they find wide usage in multifunctional load supporting and heat dissipation applications [20].

The strength of metallic foam is similar or slightly lower than the solid material of the same weight. An important characteristic property of the metal foam is the isotropic nature of their structural material. Under external loading the material structure behaves the same way on every load-bearing axis. Another unique property of metal foam is its bending stress as a function of mass moment of inertia of the material. Due to
distribution of structural material over larger volume, the overall mass moment of inertia is higher in metal foam [21]. This contributes to higher bending stress and strength for a foam structure than for same weight of solid metal [21]. This high strength combined with the low density makes it useful as a load bearing component in automotive and aerospace applications.

The density is an important factor in metal foam and it determines the mechanical properties. Elastic modulus, tensile strength and compressive strength increases with increasing density (Figure 3-3) [21].



Figure 3-3 Deformation behavior of various aluminum foams under compressive load - engineering stress vs engineering strain [21]

The characteristic stress strain curve of metal foam is divided into three stages. These can be observed in Figure 3-3. For small strains (< 1-2%>) and at the beginning of deformation, the foam deforms elastically with a linear increase of stress. For higher deformations up to 60-80 % strain, the stress is almost constant with

deformation in the plateau region. Due to this long plateau region foams are capable of withstanding high impact energy without exceeding a given stress value [22]. Finally, the deformation reaches the densification strain. At this stage, the cell walls crush together and the foam densifies. This deformation behavior is typical of all kinds of solid foams. Hence, metal foams are excellent energy absorbers due to their deformation mechanism and are used as bumpers in cars, trams, railcars etc. [23].

3.3 Hydrostatic Pressure Retainment storage vessel

HPR technology offers a great potential in hydrogen storage for automotive fuel systems by potentially reducing the weight of the hydrogen storage tanks by as much as 40% [3]. Currently, hydrogen storage on a vehicle is accomplished by a void volume vessel that is constructed in a cylinder shape. In these vessels, all of the pressure induced stresses are carried by an outer wall covering the vessel which comprises the structure. For a thin-walled cylindrical pressure vessel, we observe that the distribution of normal stresses on a plane perpendicular to the surface of the vessel is essentially uniform throughout the thickness of the vessel. The stress in the radial direction is typically negligible for a thin walled cylinder as represented in Figure 3-4.



Figure 3-4 Bi-axial stress in thin-walled cylindrical pressure vessel [3]

Thus when pressure is applied within the vessel, the structural material is subjected to a bi-axial or planar stress where the stress in the hoop direction (σ_1) is always twice the stress in the longitudinal direction (σ_2). In thick walled cylinders the stress in the radial direction is not negligible and a tri-axial state of stress exists. However, due to the inherent geometry limitations the stress in the outer wall is not very uniformly distributed over the structural material. An approach that can satisfy tri-axial loading (hydrostatic pressure) with nearly equal stress can add to efficient use of the structural material. This requires more than a void volume vessel to equally distribute the pressure induced stresses over the structural material.

An HPR vessel consists of compressing gas inside small spherical-shaped bubbles arranged within a solid mass. The arrangement pattern of the spherical bubbles within the solid material is referred to as the inner matrix structure. By compressing the gas inside small spherical bubbles within the inner matrix structure, a tri-axial state tension is induced within the structural material between the adjacent bubbles. This approach is referred to as Hydrostatic Pressure Retainment. Thus in an HPR vessel, the inner-matrix structure acts as a primary load bearing component, and this inner-matrix structure is attached to a conformable outer surface. Unlike traditional pressure vessels, the stresses in an HPR vessel is distributed within the inner-matrix and significantly lower stress is transferred to the outside wall. This favors flexible tank design and the HPR vessels can assume and take any confirmable shape (irregular shapes) to fit the automotive chassis. Since nearly all of the pressure induced stress is retained within the inner matrix, the conformable outer shell encasing the inner matrix can utilize light weight materials. Designing the outer shell for optimized thickness will greatly reduces the weight of the vessel thereby contributing for high hydrogen mass efficiency.

In an HPR vessel, the inner matrix may be of any shape, geometry or configuration (collectively referred to as the "matrix microstructure"). However, different inner matrix microstructures produce different performance results. If the inner matrix microstructure is configured properly, all or nearly all of the pressure induced stress can be transferred to the inner matrix, resulting in significant structural material efficiency advantages. Tri-axial loading requires symmetry and homogeneity to be the preferred characteristics of inner-matrix microstructure. With a simple cubic HPR vessel (spherical bubbles arranged in simple cubic structure), is subjected to high internal pressure load, the material structure tend to behave the same way on every load-bearing axis. Thus a complete state of tri-axial stress is induced within the inner matrix as shown in Figure 3-5.



Figure 3-5 Representation of Tri-axial hydrostatic stresses in inner matrix [3]

Figure 3-5 represents the tri-axial tension in the structural material between the bubbles. The HPR technique, as the name suggests is intended to maximize the

occurrence of tri-axial tension in the inner matrix material. Hence, for HPR vessels, spherical shaped bubbles of uniform size are considered for reasons of symmetry and homogeneity. It also results in a minimized development of internal shears because of the near equal omni-directional characteristic of the stress. In addition, structural performance will be completely unconstrained by the exterior physical shape.



Figure 3-6 Conformable HPR hydrogen tank [3]

The HPR storage vessels offer additional advantages over conventional cylindrical vessels. All existing high-pressure retainment systems are also prone to catastrophic "explosive" failures, when failures do occur. But, in the HPR vessel, the pressure-induced stress is distributed throughout the entire volume of the inner matrix within the HPR pressure vessel, rather than concentrated on the outer surface only. It follows that the new lightweight HPR vessels will feature a characteristic of non-catastrophic failure (if a failure would occur). Also flexible vessel shapes can overcome completely the volumetric inefficiencies caused by the configuration limitations of current spherical and

cylindrical/spherical systems. In a direct comparison with all other parameters left equal; same size and shape, same pressures, same materials, same maximum allowable stress (including factors of safety), HPR demonstrates a potential for superior material efficiency [3].

CHAPTER 4 SIMPLE CUBIC HPR VESSEL MODELING

This chapter briefly describes simple cubic structure for the HPR inner matrix. It is followed by a brief introduction to two material candidates used in this study for investigating the mass efficiency of the vessel. Finally, a review of FEA tools and preprocessing analysis is presented.

4.1 Simple cubic structure description

Simple cubic structure is represented by simple packing of one bubble at each corner in a unit cube of structural material. When the void spherical bubbles within the HPR vessel are homogenously arranged to represent a simple cubic structure, the resulting matrix is called as simple cubic inner matrix structure or simple cubic HPR vessel.

The mass of hydrogen that can be stored in a simple cubic HPR vessel is dependent on the volume occupied by the spheres or useful storage volume. In studying a simple cubic HPR vessel for its hydrogen mass efficiency it is important to define packing density. The packing density for a simple cubic structure is given by the fraction of the cube's volume that is occupied by spheres. Packing density for a simple cubic unit cell is discussed next. A representation of simple cubic unit cell is given in Figure 4-1.



Figure 4-1 Simple cubic unit cell [3]

The unit cell for the simple cubic structure contains one-eighth portions of eight corner bubbles to make one complete bubble in each unit cell. If the radius of the bubble is r_T , the edge length (a) of the cube equals two times r_T (Figure 4-1). Hence, the volume of the cube is $(2r_T)^3$. The packing density (ρ) for a simple cubic unit cell is given in Equation 4.1.

$$\rho = \frac{V_{sphere}}{V_{cube}} \tag{4.1}$$

 V_{sphere} is total volume of the spheres in the cube and V_{cube} is the volume of the cube. By substitution, Equation 4.1 reduces to Equation 4.2.

$$\rho = \frac{(4/3\Pi r_T^{\ 3})}{(2r_T)^3} \tag{4.2}$$

 $\rho = 0.524$

In other words, 52.4% of the total volume of the unit cube is occupied by the void spheres. However, in a simple cubic HPR vessel the bubbles do not touch each other $(r_T < \frac{a}{2})$ and also additional structural volume (V_{cube}) is needed for the outer wall. Therefore the packing density for a simple cubic HPR vessel is always less than 52.4%.

Therefore the packing density for a simple cubic first vessel is arways less than 52.4%.

The simple cubic HPR vessel model and its packing density for various bubble configurations are discussed next.

4.2 Simple cubic HPR vessel modeling

The CAD model of the simple cubic HPR vessel analyzed in this study is shown in Figure 4-2. The vessel is modeled out of a solid cube with sides measuring 440 mm. Spherical shaped symmetric bubbles are cut inside the solid cube to simulate a simple cubic packing arrangement. The modeling procedure is briefly discussed in this section. The solid cube is parted into four planes, all parallel to each other and to one face side of the cube. The first plane is positioned at a distance of 70 mm from one face of the solid cube. The second plane is positioned at a distance of 100 mm from the first plane. The other two planes are positioned such that the distance between any two adjacent planes measure 100 mm. Sixteen complete spherical bubbles are cut symmetrically in each of the four planes to reproduce a 4x4x4 matrix pattern on each plane. The bubbles are patterned in such a way that the centre to centre distance between any two adjoining bubbles measured 100 mm. This arrangement leads to a simple cubic inner matrix structure within the solid cube and referred to as a 3x3x3 inner matrix structure. Thus the inner matrix structure is homogenously distributed with sixty four equal sized bubbles with their center to center distance between the adjoining bubbles measuring 100 mm. Hence the bubble sphere touching radius for this structure is 50 mm.



Figure 4-2 2D view of 3x3x3 simple cubic HPR vessel

From Figure 4-2, it can be observed that simple cubic HPR vessel is a closed cell structure. For the inner matrix structure, hydrogen permeable polymeric foam is best suited to allow the flow of gas between the bubbles. The outer tank wall can utilize a light weight non-permeable solid material.

For the vessel geometry in Figure 4-2, the useful hydrogen storage volume (total volume occupied by bubbles) for six different bubble sphere radii is calculated as a fraction of total vessel volume. This is tabulated in Table 4-1. It is important to study

this relationship as the mass of hydrogen that can be compressed in the vessel is directly dependent on the useful storage volume. Hence optimizing the bubble size is an important factor in maximizing the hydrogen mass efficiency.

Bubble sphere radius	Packing density $\rho = \frac{V_{sphere}}{V}$
mm	, cube
40.0	20.14 %
42.5	24.16 %
45.0	28.68 %
47.5	33.73 %
49.0	37.03 %
51.0	41.69 %

Table 4-1 Packing density for the HPR vessel for different bubble sphere radius



Figure 4-3 Relationship between bubble sphere size radius and packing density for the HPR vessel

Figure 4-3 shows that the packing density for the simple cubic HPR vessel model varies between 42 % and 20 % for a bubble sphere radius ranging between 51 mm and 40 mm respectively. This was well expected as the maximum packing density for a simple cubic unit cell is 52.4 %. The materials that are used in this study for modeling the HPR vessel are discussed next.

4.3 Materials studied in the HPR vessel model

Material properties and the inner matrix structure are key parameters that affect the mass efficiency of an HPR vessel. In a preliminary effort to find the right choice of material, the simple cubic HPR vessel matrix structure is analyzed with two material candidates: 1) Type 305 stainless steel and 2) Epoxy/carbon fiber composite. These materials are chosen to demonstrate and compare the hydrogen mass efficiency that can be accomplished in an HPR vessel. The key properties of these materials are shown in Table 4-2. The values displayed in Table 4-2 refer to the average values taken from the literature and may vary depending on the grade of the epoxy resin and the percentage content of the carbon fiber.

	Type 305 Steel	Epoxy/carbon fiber composite
Density (kg/m^3)	8000	1570
	260	345 (1)
Yield tensile strength (MPa)	(ultimate tensile strength= 585)	(ultimate tensile strength= 810)
Elastic modulus (GPa)	193	190
Poisson's ratio	0.31	0.30

Table 4-2 Key material properties [24, 25]

⁽¹⁾ Assumed value capable to withstand 69 MPa (10,000 psi) of compressed hydrogen in the HPR vessel.

Stainless steel metal foams are one of the recently developed ultra lightweight materials. These foams have very low densities with high energy absorption capacities and therefore have widespread applications in the manufacturing of ultra lightweight structural components. Hollow metal spheres and three dimensional sintered shapes produced from stainless steel have potential applications in lightweight automotive parts, thermal protection systems for aerospace structures, biomaterials, and high-temperature thermal insulation.

Composite materials are favored where high strength to weight ratio and high stiffness to weight ratio are key factors in design. The composite material used in this study is epoxy/carbon fiber composite. The carbon fibers are bonded together in epoxy resin matrix to form successive layers of lamination. Epoxy/carbon fiber composite has excellent stiffness and strength compared to steel. This makes it more suitable than most of the metals in various structural applications. A comparison of mechanical properties between 305 stainless steel and epoxy/carbon fiber composite is given in Figure 4-4, 4-5 and 4-6. It is observed that epoxy/carbon fiber composite is 1.5 times stronger and 5 times lighter than the Type 305 stainless steel.



Figure 4-4 Yield strength comparison between steel and composite [24, 25]



Figure 4-5 Elastic modulus comparison between steel and composite [24, 25]



Figure 4-6 Density comparison between steel and composite [24, 25]

4.4 FEA tools and pre-processing

Although various solid modeling and FEA packages are available, the software tools used for this research were Solid Edge and MSC Patran with Nastran interface.

Solid Edge:

Solid Edge from unigraphics is 3D-CAD software and has ability to sketch drawings, model and assemble engineering parts. Solid Edge Version 16 was used throughout this research for modeling the HPR vessels.

MSC Patran:

MSC Patran is a general purpose Computer Aided Engineering simulation tool. It is a finite element modeler and is used to perform a variety of CAD/CAE tasks including modeling, meshing and post processing for FEM solvers NASTRAN, ANSYS, ABAQUS, LS-DYNA and MARC. It is a preprocessor that links engineering design, analysis and results evaluation in a single application.

MSC Nastran:

MSC Nastran is finite element analysis software to determine displacement, strain, stress, vibration, and temperature response with various material properties, applied loads and boundary conditions of a structure. MSC Nastran is used throughout this research in studying the structural behavior of the inner matrix model.

In studying the HPR vessel model, the hydrogen pressure applied inside the bubbles is assumed to be constant over a relatively long period of time. Hence, a linear static structural analysis simulation is performed. Firstly, the simple cubic HPR vessel is simulated for a pressure load of 3.45 MPa (500 psi). The pressure load can be applied either on the solid model (i.e., the bubble surface) or directly on the FEA model (nodes and elements). In this study, the pressure loads are applied to the bubble surfaces as there are fewer entities to choose. This is represented in Figure 4-7. Also, solid model loads are independent of the mesh and this facilitates in changing the mesh density without the need to reapply the loads. Regardless of whether the loads were applied to the bubble surfaces or the FEA model, the solver expects all loads to be in terms of the Finite Element Model. Hence, solid model loads are automatically transferred to the underlying nodes and elements during solution.



Figure 4-7 Pressure load applied to the bubble surfaces within the inner matrix

Finite Element models were generated using Patran (version 5). The HPR vessel model is uniformly meshed with a Tet 4 element. The representative solid models for analysis contained about 140,000 to 250,000 elements and 40,000 to 60,000 nodes.

The structural behavior (von Mises stress) inside the HPR vessel was studied by constraining the center node of the solid model in the x, y, and z direction. The displacement constraint of the center node is a direction-dependent nodal quantity and is interpreted in the global co-ordinate system. The centre node constraint is shown in Figure 4-8.



Figure 4-8 Displacement constraint in the center node of the HPR vessel

For each bubble configuration, the right optimum mesh density (global edge length) is chosen by performing multiple simulations with increasing mesh density, until the observed von Mises stress values had little deviation for two subsequent increasing mesh densities. The HPR vessel is modeled as a linear elastic material with two different materials discussed under section 4.3. FEA simulations were performed with progressively increasing bubble size. Multiple simulations were performed on the HPR vessel model with six different bubble configurations to quantify the optimum bubble size. A representative finite element model is shown in Figure 4-9. The results are discussed in chapter 5.



Figure 4-9 Wire-frame mesh model of simple cubic HPR vessel



Figure 4-10 Finite element model of simple cubic HPR vessel

CHAPTER 5 RESULTS AND DISCUSSION

This chapter presents the results of this thesis. A detailed discussion on stress distribution within the simple cubic HPR vessel, bubble size optimization, wall thickness optimization and desired material properties for HPR structure are presented. Bubble size optimization and wall thickness optimization are performed with reference to 305 stainless steel to obtain an optimum vessel configuration. The optimum vessel configuration is then simulated and compared for its mass efficiency with reference to steel and a composite to study desired material properties suitable for simple cubic HPR vessels.

5.1 Analysis of stress distribution within the simple cubic inner matrix

3D CAD models of the simple cubic HPR vessels are developed by patterning void spherical bubbles within a solid cube. A linear static structural approach (FEA) is taken to study the stress distribution within the hydrogen-filled inner matrix structure.

A single simple cubic unit cell was first modeled and analyzed. The unit cell for the simple cubic structure contains one-eighth portions of eight corner bubbles to make one complete bubble in each unit cell. A pressure load of 0.7 MPa (100 psi) is applied to the surfaces of the eight corner bubbles. The CAD model is shown in Figure 5-1. The FEA stress contour is shown in Figure 5-2.



Figure 5-1 CAD model of a simple cubic unit cell



Figure 5-2 Von Mises stress on a simple cubic unit cell

The FEA showed identical patterns of high stress regions on all faces of unit cubes aligned with the edges. The von Mises stress inside this region was considerably low. However, this model did not show any evidence of near-hydrostatic stress state. By HPR definition, to induce any tri-axial tension within the structure, the inner matrix has to satisfy homogeneity and symmetry with respect to bubble distribution and bubble shape. A simple cubic unit cell structure does not meet these requirements and these results were expected. Next, a 3x3x3 simple cubic inner matrix structure was modeled and analyzed for 3.5 MPa (500 psi) pressure load. The CAD model with no outer wall is shown in Figure 5-3. The von Mises stress results (in psi) for 10% of the applied load (50 psi) are shown in Figure 5-4.





Figure 5-3 CAD model of 3x3x3 simple cubic inner matrix







Interacting angular distortion zone (=~ 220 psi)



The following observations were made for the 3x3x3 inner matrix. The FEA results showed the occurrence of near-hydrostatic stress ($\sigma_{\text{von Mises}} = \sim 0.20$ MPa) in the structural material furthest from the bubble. The von Mises stress in this region was negligible compared to the maximum stress value ($\sigma_{\text{von Mises}} = \sim 1.5$ MPa). Each bubble was surrounded by a circumferential region or angular distortion zone. The von Mises stress in the angular distortion zone ($\sigma_{\text{von Mises}} = \sim 0.7$ MPa) was much higher than outside the zone due to induced shear stress. Also the maximum von Mises stress ($\sigma_{\text{von Mises}} = \sim 1.5$ MPa) was observed in the thin structural plateau between two adjoining bubbles. These are the zones where one or more angular distortion zones interfere with each other.

The vessel section was investigated for principal stresses (σ_1 , σ_2 and σ_3) in the region between the bubbles. Approximate values for σ_1 , σ_2 and σ_3 were observed to be 0.27 MPa, 0.20 MPa and 0.06 MPa respectively. These indicate that pure hydrostatic state of stress is an ideal case ($\sigma_1 = \sigma_2 = \sigma_3$). Nevertheless, within the near-hydrostatic region, the principal stress values are consistently closer to each other resulting in significantly lower von Mises stress in the near-hydrostatic region between the bubbles.

Similar results were observed for a larger 8x8x8 inner matrix structure at a pressure load of 3.45 MPa (500 psi). Regions of maximum von Mises stress were difficult to represent in Figure 5-5 due to the complexity of the model.



Angular distortion zone $(\sigma = ~ 6 \text{ MPa})$ Near-hydrostatic stress ($\sigma = ~ 0.13 \text{ MPa}$)Figure 5-5 Von Mises stress distribution in a 8x8x8 simple cubic inner matrix

These results were consistent with the simple models of the one bubble case and multiple bubble case discussed under section 1.3. This confirms that simple cubic inner matrix structure induces near-hydrostatic stress between the bubbles, thus efficiently utilizing the structural material.

5.2 Effect of bubble size on stress within the inner matrix

The FEA results confirmed the occurrence of maximum von Mises stress in the region between two bubbles. The HPR vessel model was analyzed for maximum von Mises stress by progressively increasing the bubble sphere radius. A graph was plotted between bubble sphere radius and the maximum von Mises stress. It is shown in Figure 5-6. The steep rise in stress is due to interference between the angular distortion zones of the surrounding bubbles and can be explained as follows.



Figure 5-6 Effect of bubble sphere radius on the maximum von Mises stress (between two adjacent spheres) in a simple cubic HPR vessel - Type 305 stainless steel

When the bubbles are subjected to a compressive load, the section of material in-between two bubbles (consider 2D cut plane) are loaded in tension. As the bubble size increases, the bubbles get closer to each other and bubble spacing decreases. This means that there is less structural material between any two bubbles and this result in the maximum stress concentration in the region between the bubbles.

With increasing bubble size the angular distortion effect causes a steep rise in stress. The angular distortion zone is responsible for high induced resolved shear stress near the surface of the bubble. This can be clearly observed in the shear stress fringe plot shown in Figure 5-7. When pressure is applied within the bubbles of the inner matrix, there is a progressive increase in von Mises stress with increasing bubble size. As the bubbles get closer to each other, the angular distortion zone (circumferential region surrounding the bubble) of one bubble interacts with the angular distortion zone of the surrounding bubbles. This interaction between the angular distortion zones of the

adjoining bubbles causes the maximum von Mises stress to occur in the region between the bubbles.



 $\tau = 110 \text{ psi}$ (Max induced resolved shear stress)





Figure 5-7 shows the shear stress fringe plot indicating the maximum induced resolved shear stress occurring in the thin plateau between the bubbles. The near-hydrostatic region is free of shear stress.

5.3 Optimum bubble sphere radius for maximum H₂ mass efficiency

The inner matrix structure and the material properties are two key parameters that affect the hydrogen mass efficiency. Hence the change in mass efficiency with respect to varying bubble size and material properties are studied in the subsequent sections.

Hydrogen mass efficiency is calculated by the following method. To compute the hydrogen mass efficiency, the maximum working pressure (maximum pressure the hydrogen gas can be compressed) for different bubble sizes needs to be evaluated first. To achieve this, the engineering FEA for the HPR vessel model is explored at a randomly

chosen low pressure of 3.45 MPa (500 psi). The FEA is repeated for six different bubble configurations. The von Mises stress results obtained for six different bubble configurations at 3.45 MPa (500 psi) of compressed hydrogen are then extrapolated on a linear scale to calculate the maximum working pressure in the HPR vessel model. The FEA results are shown in Table 5-1.

Bubble	Applied	Type 305 stainless steel		Epoxy/carbon fiber composite		
sphere	pressure					
radius		Max	Max	Max	Max	
		von Mises	Pressure	von Mises	Pressure	
(mm)	MPa (psi)	MPa (psi)	MPa (psi)	MPa (psi)	MPa (psi)	
	_	_	_	_	_	
40.0	3.45 (500)	4.65 (675)	193 (27925)	4.65 (675)	255 (37065)	
42.5	3.45 (500)	6.17 (896)	145 (21037)	6.17 (896)	192 (27923)	
45.0	3.45 (500)	9.80 (1420)	91 (13274)	9.80 (1420)	121 (17619)	
47.5	3.45 (500)	13.8 (2000)	65 (9425)	13.8 (2000)	86 (12510)	
49.0	3.45 (500)	16.7 (2420)	54 (7789)	16.7 (2420)	71 (10338)	
51.0	3.45 (500)	36.0 (5220)	25 (3611)	36.0 (5220)	33 (4792)	

Table 5-1 FEA results for six different bubble sizes with two material candidates

The FEA results and the angular distortion zone effects confirm that with increasing bubble size, the maximum von Mises stress is concentrated in the region between the bubbles (discussed under section 5.2). This results in reduced load bearing strength in the region between the bubbles as the bubbles get closer to each other. It follows that the maximum pressure to which the hydrogen gas could be compressed into the vessel model without structural failure decreases with increase in bubble size. This trend is noticed in Table 5-1.

From Table 5-1, it is observed that epoxy/carbon fiber composite can withstand pressures as high as 255 MPa. However, in chapter 2 it was discussed that higher hydrogen densities become increasingly difficult to attain at higher pressures due to their

large deviations from the ideal gas. Hence for epoxy/carbon fiber composite, 68.94 MPa (10,000 psi) is taken as the maximum useful pressure for compressed hydrogen. The relation that is used to calculate the maximum working pressure at different bubble size is given in Equation 5.1.

$$P_{\max} = P_{applied} \frac{\sigma_{YS}}{\sigma_{\max_V.M}}$$
 5.1

 P_{max} is maximum working pressure, $P_{applied}$ is applied pressure load (i.e., 3.45 MPa), σ_{ys} is material yield strength, $\sigma_{\max_v M}$ is von Mises stress. The useful hydrogen storage volume (vol_{H_2}) within the HPR vessel for different bubble configurations is estimated using the relation given in Table 4.1. The density of hydrogen (ρ_{H_2}) at different working pressures (P_{\max}) is computed using the Beattie Bridgeman plot for hydrogen (discussed under section 2.1). By knowing the density of hydrogen (ρ_{H_2}) and the useful hydrogen storage volume (vol_{H_2}) within the HPR vessel, we can easily calculate the mass of hydrogen stored in the vessel (M_{H_2}) . From the structural volume of the HPR vessel (vol_{vessel}) and the material density, we can calculate the mass of the vessel (M_{vessel}) . Hydrogen mass efficiency $(Mass \eta)$ is given in Equation 5.2.

65

Mass
$$\eta = \frac{M_{H_2}}{M_{vessel} + M_{H_2}}$$
 5.2

Table 5-2 shows the calculated hydrogen mass efficiency values for six different bubble configurations. The primary goal is to derive an optimum bubble sphere size range for the simple cubic inner matrix vessel that would yield maximum hydrogen mass efficiency. The bubble optimization study was done with respect to 305 stainless steel. But irrespective of the material used, the optimum bubble sphere is assumed to be the same for epoxy/carbon fiber composite. Optimum bubble size is a function of geometry of the inner matrix structure. It is not a material property. Hence, the hydrogen mass efficiency value for epoxy/carbon fiber composite was examined only for the optimized vessel configuration (optimized bubble size and optimized wall thickness) and is presented under section 5.5. Table 5-2 shows the hydrogen mass efficiency value for different bubble configurations with reference to Type 305 stainless steel.

Bubble size	Total storage volume		Total M_{H_2}	Total $M_{\tan k}$	H ₂ mass efficiency	
(mm)	m ³	liter	кg	къ	%	
42.5	0.020	20	0.497	517	0.10	
45.0	0.024	24	0.943	486	0.19	
47.5	0.028	28	0.901	452	0.20	
49.0	0.031	31	0.866	429	0.20	
51.0	0.049	49	0.548	397	0.14	

Table 5-2 Hydrogen mass efficiency for six different bubbleconfigurations - Type 305 stainless steel



Figure 5-8 Hydrogen mass efficiency for six different bubble configurations - Type 305 stainless steel

It can be observed from Figure 5-8 that this optimized bubble sphere radius (45 mm to 49 mm) is specific to the 3x3x3 vessel geometry shown in Figure 4-2. But the bubble sphere optimization analysis is intended to reasonably predict an optimum bubble size range for a vessel of any given geometry but not limited to one that is being investigated in this study. Hence, the optimum bubble size is defined as a percentage of half the distance between the centers of two closest bubbles lying on the same edge or bubble sphere touching radius. From Figure 4-2 it can be noted that the bubble sphere touching radius is 50 mm. It follows that the optimum bubble sphere radius for any simple cubic inner matrix vessel may range between 90 % (45 mm/50 mm) and 98 % (49 mm/50 mm) of bubble sphere touching radius. For a vessel of any given geometry, by fixing the bubble centre to centre distance, we can reasonably predict a range for optimum bubble sphere radius through the relationship developed from this analysis.

5.4 Wall thickness optimization for maximum H₂ mass efficiency

By the definition of hydrogen mass efficiency, reduced tank weights contribute significantly to higher mass efficiencies. For a vessel of any given geometry and inner matrix structure, the overall mass efficiency can be increased by optimizing its outer wall thickness without affecting the stress distribution within the inner matrix. By minimizing the outer wall to its optimized thickness, the overall weight of the HPR vessel is reduced, thus contributing to increased hydrogen mass efficiency. This study was performed only with steel as the primary goal of this analysis was to obtain an optimized vessel configuration that could be studied for mass efficiency comparison between steel and composite to classify material properties suitable for simple cubic HPR vessels. Hence it is required to arrive at standard optimized vessel geometry for comparing both the steel and epoxy/carbon fiber composite. Since 305 stainless steel has a very high density with low yield strength compared to epoxy/carbon fiber composite (discussed under section 4.3) an optimized vessel configuration obtained with steel can also be applied to epoxy/carbon fiber composite for hydrogen mass efficiency comparison. These are discussed in the next section.

The wall thickness optimization process was performed for the HPR vessel model with the initial geometry (discussed under section 4.2). A bubble size (48.5 mm) within the optimized range was chosen for the inner matrix structure. The outer wall thickness was reduced in progressive steps, by a few millimeters on each of the six faces of the HPR vessel. FEA was performed at each step to study the stress distribution pattern on the outer wall surface.

This procedure is repeated until the maximum von Mises stress equaled that of internal stress. This can be observed in Figure 5-9.



Figure 5-9 Wall thickness optimization showing the stress distribution on the outer wall - Type 305 stainless steel

The results showed that by reducing the wall thickness by 4.5 mm on all six faces of the HPR vessel model, the mass of the tank decreased by nearly 10 %. Hydrogen mass efficiency increased about 9 %.

The wall optimization result is shown in Table 5-3.

Tank wall thickness reduced in steps	Distance from wall edge to bubble surface on one side	Mass of tank	H ₂ mass efficiency	
(mm)	(mm)	(kg)	%	
440	21.5	437	0.203	
436	19.5	418	0.210	
432	17.5	400	0.220	
431 (optimized thickness) ⁽¹⁾	17	396	0.220	
428	15.5	391	0.223	

Table 5-3 Hydrogen mass efficiencies with varying wallthickness - Type 305 stainless steel

⁽¹⁾ Maximum hydrogen mass efficiency is obtained at this wall thickness

5.5 Material comparison for a simple cubic HPR vessel

To make a meaningful comparison between the materials the analysis should be performed on the same vessel configuration sharing the optimized vessel geometry. Hence, the optimized vessel configuration was simulated for 5 kg of hydrogen with 305 stainless steel and epoxy/carbon fiber composite. A mass of 5 kg of hydrogen was taken as reference since U.S Department of Energy has estimated a storage capacity of 5-6 kg of hydrogen for a passenger car to achieve a range of 480-560 km (300-350 miles) [5]. The hydrogen mass efficiency is calculated and presented in Table 5-4.

	Optimized bubble size	Max H ₂ pressure	Mass of tank	Mass of hydrogen	H ₂ mass efficiency
	mm	MPa (psi)	kg	kg	%
Type 305 stainless steel	48.5	54 (7789)	2267	5	0.22
Epoxy/carbon fiber composite	48.5	69 (10,000)	242	5	2.02

Table 5-4 Mass efficiency comparison between a metal and a composite for 5 kg hydrogen storage in a simple cubic HPR vessel

The results indicate that for storing 5 kg of hydrogen in a simple cubic HPR vessel, epoxy/carbon fiber composite offers nine times higher hydrogen mass efficiency than the Type 305 stainless steel structure by reducing the overall weight of the HPR vessel. It follows that for storing the same mass of hydrogen, a composite material can yield higher mass efficiencies than the metals, simultaneously reducing the weight of the vessel greatly.

CHAPTER 6 CONCLUSION AND FUTURE WORK

The study provides understanding on the structural behavior of the simple cubic HPR vessel for compressed hydrogen storage. The following conclusions are based on the analysis results discussed in chapter 5.

Although theoretical assumption of pure hydrostatic stress is an ideal case, FEA verified that the structural material in a simple cubic HPR vessel is subjected to near-hydrostatic state of stress. This was observed in simple cubic arrangement of 3x3x3 inner matrix and 8x8x8 extended matrices (Figure 5-4 and Figure 5-5). The von Mises stress in sections of material within the simple cubic inner matrix is negligible when compared to the maximum von Mises stress. Each spherical bubble within the simple cubic inner matrix is surrounded by a region of very high stress or angular distortion zone. The maximum von Mises stress occurs in the region between the bubbles where angular distortion zone of one bubble interferes with the angular distortion zones of the surrounding bubbles. This confirms that the pressure induced stress in the simple cubic HPR vessel is evenly distributed within the inner matrix structure. The outer wall is not subjected to very high stress and can utilize light weight material. These results from the simple cubic inner matrix HPR vessel infer that the structural material is more efficiently used and can result in weight and cost savings for hydrogen storage applications.

Bubble spacing within the simple cubic HPR vessel is an important parameter and effects on the hydrogen mass efficiency (Figure 5-8). For a simple cubic inner matrix structure the optimum bubble sphere radius can be predicted to lie anywhere between 90% and 98% of the bubble sphere touching radius. This relationship can be applied to

estimate an optimum bubble size range for any large volume simple cubic HPR vessel with varying outer wall shape by fixing the center to center distance between the bubbles.

Steel is non-permeable to hydrogen and a simple cubic HPR vessel fabricated with steel requires bubble interconnectivity for passage of hydrogen. It can be observed from Figure 5-8 that at 102 % of bubble sphere touching radius, the vessel inner matrix takes an open cell structure and offers a hydrogen mass efficiency around 0.13 %. Although this is outside the optimum bubble sphere range, it is a practical configuration as it allows free flow of hydrogen gas between the bubbles. It can be concluded that 102 % of bubble sphere touching radius is a practical and usable arrangement for a simple cubic HPR vessel fabricated with Type 305 steel as the parent material.

The pressure induced stress in a simple cubic HPR vessel is distributed within the inner matrix with a low portion of the stress being transferred to the outer wall. Hence, optimizing the wall thickness for a HPR vessel yields increased mass efficiency by reducing the overall weight of the vessel.

A simple cubic HPR vessel needs hydrogen permeable material with higher yield strength and lower density when compared to Type 305 stainless steel. Hence steel does not seem to be a very suitable candidate for HPR vessels. Epoxy/carbon fiber composite, with its very low density (1570 kg/m³) and high yield strength (345 MPa) offers mass efficiency around 2.02 %. This is still low when compared to the short term (2005-2010) target of 6 % hydrogen mass efficiency set by U.S Department of Energy. Following the method adopted in the study, neither epoxy/carbon fiber composite nor steel meets the required targets for simple cubic HPR vessel.
Epoxy/carbon fiber composite exhibits nine times higher hydrogen mass efficiency than Type 305 stainless steel for same vessel volume. It concludes that hydrogen permeable materials with a combination of very low density and higher yield strength are best suitable for simple cubic HPR vessels. With a suitable inner matrix structure and material choice, an HPR vessel is expected to be capable of meeting the demands of hydrogen storage systems.

Recommendations

As a direct follow-up upon the conclusion of this study, the following recommendations are provided for further investigation on HPR vessels for hydrogen storage.

Through FEA, this study supports the feasibility of a simple cubic HPR vessel for hydrogen storage applications. However, different inner matrix structures offer different hydrogen mass efficiencies. Extended study of the HPR vessels by means of BCC and FCC inner matrix structures are expected to yield higher hydrogen mass efficiencies due to their higher packing density. However, his needs be investigated and the mass efficiency compared with different inner matrix structures. This will help to determine the best possible inner matrix structure for HPR vessels.

Actual cellular materials which offer a combination of low density with higher yield strengths are more suitable for HPR vessels. Hydrogen-permeable polymers having microstructures close to ideal HPR structures and superior material properties need to be investigated. With extended research and industrial support it will be possible to determine the right material with low cost. After achieving the right inner matrix structure and the material candidate through previous efforts, the CAD model of the actual foam microstructure with complex tank geometry could be studied. The analysis should go beyond the static-structural analysis considered in this study to account for thermal loads, hydrogen permeability, potential chemical reactions and vessel refueling ability. Dynamic crash modeling simulations could be performed to study the catastrophic failure behavior of HPR vessels. Actual vessel prototypes should be built and experimentally tested. Finally, research on different foam manufacturing technologies will help in identifying suitable manufacturing method for HPR vessels.

This study has analytically supported the feasibility of HPR vessel as a potential application for compressed hydrogen storage. However successful implementation of HPR vessels requires more research and experimental verification. The results of this study are intended to serve as a starting point for further research and development of this technology.

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