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Mechanical behavior of metal hydrides and hydrogen storage containers: A review

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Abstract

This review article delves into the intricate mechanical behaviors exhibited by metal hydrides within hydrogen storage tanks during hydrogen absorption and release processes. The metal's crystal structure undergoes expansion upon hydrogen absorption, leading to the liberation of energy—an exothermic phenomenon. Conversely, during hydrogen release, the metal contracts, necessitating an intake of energy from the surroundings—an endothermic occurrence. These cyclic processes give rise to two significant mechanical implications: firstly, the initiation of a decrepitation mechanism; secondly, the material undergoes rhythmic expansion and contraction, often referred to as "hydride breathing." These dual mechanisms collectively contribute to the escalating strain and stress imposed on the walls of the metal hydride container, thereby impacting its structural integrity. This review delves into the comprehensive landscape of experimental studies, measurement techniques, and modeling approaches employed in analyzing stress and strain within metal hydride hydrogen storage tanks. The report encompasses an exploration of the factors amplifying mechanical stresses within the metal hydride bed, alongside proposed strategies for their mitigation and control. Furthermore, the article concludes by presenting pragmatic and experimental recommendations aimed at the development of secure hydrogen storage tanks grounded in metal hydride technology.

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1. Introduction

Hydrogen is an abundant, non-polluting, and renewable substance that is utilized in numerous diverse sorts of industries. It has a high energy density, which is three times greater than the amount of petroleum (1-3). This means it could be used instead of fossil fuels. Hydrogen can be burned without producing any carbon dioxide emissions (4). It can also be used in hydrogen fuel cells to generate electricity, with water as the only byproduct. As a fuel, hydrogen has a low energy-to-volume ratio under normal conditions (5). However, hydrogen can potentially contain higher energy levels when compressed into a smaller volume, which poses many safety issues for hydrogen storage systems.

Two primary classifications of hydrogen-storing methods exist: physical and chemical hydrogen storage methods (6, 7). Physical methods for storing hydrogen without any chemical change include compressed hydrogen and liquefied hydrogen. In the compressed hydrogen storage method, pressurized gaseous hydrogen is kept in tanks with thin walls under high pressure. However, in this method, the energy stored in compressed hydrogen is lower than an equivalent gasoline quantity (8, 9). Liquid hydrogen tanks store hydrogen at low temperatures to reduce the hydrogen storage pressure. By utilizing this approach, the storage capacity can be increased almost twice as opposed to the compressed hydrogen storage method (7), but its cost is 30% to 40% more (9, 10). In the chemical hydrogen storage methods, hydrogen connects with a storage material through covalent solid/ionic bonds or weak van der Waals forces. Thus, these storage systems include hydrogen storage media and storage containers. These methods can be classified into solid-state hydrogen storage and chemical storage. The solid-state storage method can be recharged on the device using hydrogen, whereas chemical storage materials must be recharged at centralized facilities elsewhere (11). Chalk et al. (12) stated that the success of fuel cell vehicles in the market depends on the ability to store hydrogen as a solid material.

An efficient hydrogen storage system should be capable of storing the most significant quantity of hydrogen per mass and volume unit and have anti-leak capability (13). In the solid-state storage method, the hydrogen release rate, in case of leakage, is much lower than the other methods, reducing the explosion risk. Therefore, the solid-state storage method is more common and covers a wide extent of industrial applications such as fuel cells used to power vehicles (14-17), hydrogen compressors (18), closed thermodynamic frameworks, actuators, heat storage systems, and gas filtration (19-21).

Metal hydrides represent the most efficient method for securely storing hydrogen, showcasing remarkable energy storage capabilities (22). These compounds form through a chemical interaction between hydrogen and metal elements, resulting in a reversible reaction under moderate pressures and temperatures. As a result, metal hydrides demonstrate rapid hydrogen absorption and release capabilities (23). Notably, metal hydrides offer inherent safety advantages over mechanical hydrogen storage techniques, making them highly desirable absorbent materials (24). One of the obstacles associated with the use of metal hydrides to store hydrogen is the metal hydride breathing phenomenon, which is a term that describes the metal hydride's volume change when it absorbs and desorbs hydrogen gas. This phenomenon can cause mechanical stress to the metal hydride container, depending on the material's shape, size, and density (25).

The main objective of this review paper is to better comprehend the hydride breathing phenomenon and the stresses induced in the tanks' structures. In the first part of this review, studies conducted to investigate the metal hydride behavior in breathing phenomenon and measuring the metal hydride bed's volume change are reviewed. The second part reviews the numerical and experimental research to investigate the stress on the tank wall while hydrogen is being absorbed. Finally, the practical rules and experimental and scientific recommendations for the safe design of a metal hydride hydrogen storage tank are stated.

2. Metal hydrides mechanical properties

The origins of metal hydrides can be traced back to the early 19th century; during this period, they were initially perceived as anomalies that did not align with the prevailing scientific theories (26). This perception changed in 1868 when T. Graham made a significant discovery: he found that metallic palladium wires could absorb hydrogen gas, leading to the formation of metal hydrides (27). This breakthrough marked a turning point, sparking increased interest and systematic investigation into metal hydrides by the late 19th century. In a pivotal moment in 1971, E. Muetterties played a key role by editing the first comprehensive book on transition metal hydrides (28). This seminal work placed significant emphasis on the synthesis and stereochemistry of hydride complexes, solidifying the foundation for understanding metal hydrides.

Metal hydrides can be utilized as simple metal, alloy, or combined form. Simple metal hydrides are compounds that contain only one metal and hydrogen in their crystal structure. They have a moderate hydrogen capacity, a high enthalpy of hydride formation, and a high hydrogen desorption temperature (29). Alloy metal hydrides comprise multiple metals and hydrogen in their crystal structure. They are usually formed by intermetallic compounds that absorb and release large amounts of hydrogen gas. These substances have various applications in hydrogen storage, batteries, and catalysis. Their hydrogen absorption/desorption reaction kinetics are high, and their activation energy to initiate the reaction is lower. Due to their high weight and volume capacity in storing hydrogen, complex hydrides have the necessary efficiency to overcome the performance defects of simple and alloyed metals (30) (31).

Alloy hydrides are the most common hydrides as solid absorbent material. They have the general formula of $A_m B_n H_x$ and are made of two metal components, A and B, which combine with hydrogen to create compounds known as hydrides in the form of AH_x and BH_y. In the given formula, component A represents the base metal component that forms a hydride, while component B belongs to the complementary metal component that does not form any hydride or generally acts weakly in this reaction. The most common alloy that can be used in metal hydride (MH) tanks as absorbent media is the metal composition with the formula AB_5. Numerous AB_5 compounds can be obtained by simply substituting elements in either A or B positions. Remarkable gas storage attributes are demonstrated by these compounds, which resist gaseous impurity, stay stable for a long time, and have high volumetric storage density (7). Large groups of researchers have been investigating AB_5 metal hydride problems and applications (32-34). For example, Dong et al. (35) and Jean-Marc Joubert et al. (36) reviewed the widespread application and properties of AB_5-based intermetals in recent tests. Their review covered topics such as crystal structures, thermodynamic properties, hydrogen storage, magnetic properties, electrochemical applications, and novel utilization of AB_5 compounds.

In the early 1980s, a crucial development took place in the field when the mechanical integrity of containers holding hydrides was, for the first time, highlighted by Lynch (37). This awareness arose when he witnessed a vessel's failure during the hydrogenation process. This awareness pertains to the primary stage of the noteworthy development of hydride tank prototypes (the subsequent stage transpired post-2000 when hydrogen technology regained prominence) (16, 17, 38). Hence, when a container has been constructed to contain a considerable quantity of hydride, the assessment of its mechanical durability is approached more carefully. In this approach, it is imperative to consider the phenomena of hydride powder swelling as an effective factor in the mechanical behavior of MH tanks and metal hydride beds.

Furthermore, the metal hydride's volume change due to absorbing hydrogen accompanied by material pulverization (39), also known as decrepitation (40) or disintegration (41, 42) in other investigations, should be considered. This occurrence results from the material's heterogeneous hydration (43). In general, the hydride materials exhibit brittle behavior. Therefore, under high stresses caused by heterogeneous phase change in its structure, the material breaks into smaller particles due to cleavage rupture. Although the crushing of metal hydride particles causes agglomeration, leading to increment mechanical stresses in the hydride bed, the rate at which hydrogen is absorbed will be improved when the particles are crushed into smaller pieces. This is mainly ascribed to the increasing reaction rate constant after the particle's size decreased (44). As a result, this aspect helps to increase a new fresh surface for hydrogen absorption and ultimately improves the storage capacity of the MH tank. Hence, researchers focused on the impact of other factors, such as operating conditions, tank shape, geometry, etc., on the MH tank and metal hydride's mechanical behavior.

The porosity of an MH tank is a parameter that reflects the fraction of void space in the bed of metal hydride and affects various aspects of the tank's performance, such as heat and mass transfer, hydrogen capacity, kinetics of the reaction and permeability of metal hydride bed. It varies according to different circumstances, such as the type and composition of a metal hydride, the shape, geometry, and size of the metal hydride bed's particle, the packing method and density of the metal hydride bed, and the presence of additives or fillers. Reducing the porosity of a metal hydride (MH) tank can result in heightened thermal conductivity (45, 46). In a study conducted by Busqué et al. (47), employing both experimental testing and a COMSOL software model, it was established that a greater porosity in the MH tank leads to reduced charging time and hydrogen storage capacity. Later, in 2019, Lin et al. (48) used a finite element method to simulate heat and mass transfer reaction kinetics and analyzed the impact of particle radius and porosity in an MH tank on its hydrogen absorption ability. The findings suggest that greater porosity holds advantages for the hydrogen absorption process. Mostafavi et al. (49) employed a mathematical model encompassing heat, mass transfer, and reaction kinetics to reveal that an optimal porosity of approximately 0.6 yields favorable results. Interestingly, hydrogen absorption improves as porosity increases, albeit accompanied by an elongated absorption process. This duality highlights a trade-off: while enhancing one aspect, it inadvertently hampers the other. This dilemma underscores the significance of meticulously selecting the porosity parameter during the optimal design of metal hydride tanks.

Gillia (25) presented a set of parameters that are used to define the porosity and geometrical definitions of the MH tanks. To calculate some of these parameters, it is necessary to use a transparent container (50-52), X-ray pictures, neutron radiography pictures (53), or trace the location of a piston by recording the position of the hydride bed surface (54) or eventually utilize impedance techniques to monitor the height of the hydride bed (55).

3. MH tank thermodynamics

In a solid-state storage method, the hydrogen molecules may adhere to the solid surface or break into their constituent atoms, penetrate into the solid body, and take place in its atomic structure (56). The former case is physisorption, in which the kinetic energy of the gas molecules should be reduced by lowering the temperature so that the gas molecules can adhere to a solid surface. While in the latter case, breaking hydrogen into its constituent atoms is endothermic and necessitates energy expenditure. Then, bringing the resulting atoms to rest and forming bonds between them and the solid matter is exothermic and requires cooling the hydrogen adsorbing layer (5, 57).

Metal hydride's high volumetric capacity at low temperatures and pressures makes them superior absorbents. The efficiency of energy storage due to the use of such absorbent materials is approximately 88%. Rechargeable hydrides, which are generally called metal hydrides, are more suitable options when a high storage capacity and fast charging and discharging are required. They are usually made into powder form with the particles' diameter in the range of microns to create a bigger absorbent surface and to accelerate gas penetration (58). Four distinguished classifications of materials excel in solid-state hydrogen storage: carbon and similar materials with large surface areas, H_2O-reactive chemical hydrides, thermochemical hydrides, and rechargeable hydrides (7).

If the enthalpy of the separate formation of these hydrides is equal to ΔH_A and ΔH_B, respectively, the compound enthalpy will be between those two values. In other words, we will have $\Delta H_A < \Delta H_{AB} < \Delta H_B$ (7, 59, 60). The interaction between hydrogen atoms and metal atoms affects the characteristics of these intermetallic compounds. Accordingly, the properties of these hydrides depend on the crystal structure of their compounds.

To create a metal hydride, the hydrogen molecule in the gas phase on the surface of the metal is divided into two hydrogen atoms that penetrate into its atomic structure. This reaction is described by:

$$M(s)+\frac{1}{2}xH_2(g) \Leftrightarrow MH_x(s)+Q \quad (1)$$

where Q (kJ) represents the quantity of heat released during the occurrence of the reaction. An exothermic reaction ($Q > 0$) is observed during the process of storing hydrogen, while in order to release hydrogen from the material, an endothermic process occurs ($Q < 0$). During the absorption process, hydrogen molecules are separated on the surface of the metal matrix and penetrate its crystal structure. This phenomenon causes the metal's crystal lattice to expand and creates strain and defects in the lattice structure (61). In the process of discharge or dehydrogenation, hydrogen atoms separated from the structure of the metal network recombine and form hydrogen molecules.

In charging, the initial pressure within the tank is near the ambient pressure, and then hydrogen gas with elevated pressure is injected into the tank. The motion of hydrogen gas towards the metal hydride's porous region stems from the MH container's pressure gradient. As soon as hydrogen enters the tank, the process of hydrogen absorption by the metal begins. The exothermic hydrogen absorption reaction on the metal's surface engenders an increment in temperature within the MH tank, consequently increasing the internal pressure. This expansion in pressure diminishes the pressure gradient created between the inlet hydrogen gas and the interior of the tank, and the rate of hydrogen entry and its absorption by the metal decreases. In order to sustain the charging process, it is imperative to extract the thermal energy resulting from the hydrogen absorption reaction. The procedure of removing heat typically entails implementing a cooling system installed around the tank structure. This system enhances the absorbed rate and overall quantity of hydrogen absorbed in a shorter time (62, 63).

Likewise, during the discharge phase, the first stage involves the liberation of hydrogen from the metallic compound and then its eventual departure from the container. Primarily, the isolation of hydrogen from the metallic surface occurs. The liberation of hydrogen molecules from the surface of metal induces an increase in the tank's internal pressure, leading to a gradient in pressure between the interior and exterior of the tank. During this process, the hydrogen pressure and its temperature decrease simultaneously. A heating system is necessary to avoid freezing, blockage of the gas discharge channel, and the thermal stresses in the tank container.

It was discovered that pressure and temperature affect the charging and discharging process in MH tanks. Subsequently, the circumstances for the acknowledgment of this process rely upon the structure of the solid phase and its thermodynamic behavior within the condition's hydrogen gas absorption or desorption. The pressure-composition-temperature (PCT) curve is used to describe the thermodynamic behavior of solid hydride formation in such systems. Fig. 1. PCT curves for hydrogen absorption in a typical hydrogen-absorbing metal or alloy at different temperatures. A flat plateau characterizes the coexistence region of the α-and β-phases and ends at the critical temperature (A). The Van't Hoff plot related to the phase transition from metal to metal hydride. The enthalpy and entropy of the hydrogenation reaction can be obtained from the slope and the intercept of this plot, respectively (B). offers additional information to facilitate understanding of the hydrogen absorption process. In this diagram, the α phase occurs in metal when hydrogen is placed on its surface. When an atom of hydrogen penetrates into the metal's crystal structure, the β→α phase change begins and continues until the metal structure is saturated with hydrogen and reaches the full β-phase (64). Under ideal conditions, the hydrogen absorption/desorption reaction's phase change occurs at constant pressure. Fig. 1. PCT curves for hydrogen absorption in a typical hydrogen-absorbing metal or alloy at different temperatures. A flat plateau characterizes the coexistence region of the α-and β-phases and ends at the critical temperature (A). The Van't Hoff plot related to the phase transition from metal to metal hydride. The enthalpy and entropy of the hydrogenation reaction can be obtained from the slope and the intercept of this plot, respectively (B).A shows that as the temperature rises, so does the pressure in the phase change region until it reaches the critical temperature T_c. Once the temperature surpasses Tc, the plateau region vanishes, and the α-phase continuously converts to the β-phase. The extent of the plateau region directly impacts the quantity of hydrogen that can be stored reversibly with a small pressure variation.

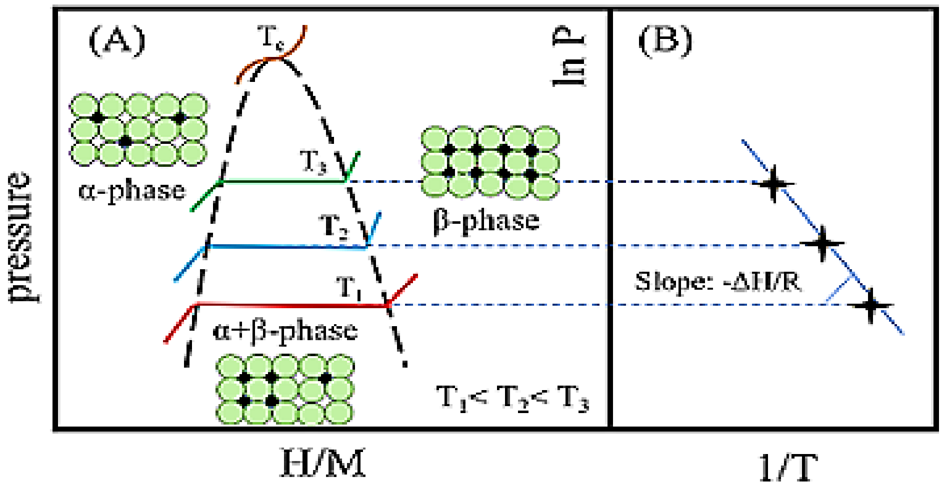

Fig. 1. PCT curves for hydrogen absorption in a typical hydrogen-absorbing metal or alloy at different temperatures. A flat plateau characterizes the coexistence region of the α-and β-phases and ends at the critical temperature (A). The Van't Hoff plot related to the phase transition from metal to metal hydride. The enthalpy and entropy of the hydrogenation reaction can be obtained from the slope and the intercept of this plot, respectively (B).

The rate of hydrogen diffusion into and out of the material can be influenced by the size, shape, and morphology of the metal hydride particles and the surface area available for hydrogen adsorption. Optimization of these factors can lead to improved hydrogen storage performance. In general, smaller metal hydride particles have a greater surface area per unit mass than larger particles, which can increase the rate of hydrogen adsorption and desorption (65, 66). This can result in a shift to the left in the absorption and desorption curves on the PCT diagram, meaning that the material can both absorb and desorb hydrogen at lower pressures and temperatures. The equilibrium pressure of the metal hydride, denoted by P_{eq}, refers in MH tanks to the pressure of the material phase change between solid and hydride phases during hydrogen absorption/desorption reaction in its ideal state (67). This equilibrium pressure depends on the changes in enthalpy and entropy during the formation or deformation of hydrides. The Van't Hoff equation expresses this parameter as a function of temperature.

A hydrogen storage tank incorporating hydrides is essentially a thermal machine designed to handle heat flux. Various sources examine techniques to optimize MH tank operations by controlling their heat. These techniques solve the heat energy equation conservation with an energy source term linked to the enthalpy of the reaction applied to the porous hydride material. Two recent reviews have summarized these studies well (68, 69). The introduced models are often effective in enabling engineers to design efficient and optimized systems for thermal performance (16, 69-75).

4. Strength and Durability of MH Tanks

The efficiency of charging refers to the ratio of the quantity of hydrogen stored in the tank to the amount of hydrogen that could theoretically be stored in it based on the MH material properties (76). The four main metal hydride bed characteristics that affect the quality of hydrogen absorption in MH tanks are porosity, thermal conductivity, absorption rate constant, and activation energy (47, 49, 77-81).

The absorption rate constant is a concept that was introduced by researchers who studied the kinetics of the hydrogen absorption process within metal hydride beds. It is based on various elements, including the type and composition of metal hydride, the temperature and pressure of hydrogen gas, the particle size and metal hydride bed's porosity, and the presence of catalysts or additives (49, 78, 79). The activation energy of an MH tank is a variable that defines the energy extent for absorption and desorption reactions of hydrogen in a metal hydride bed. The metal compound's capacity to absorb hydrogen decreases proportionally to its activation energy for the absorption reaction (47). The thermal conductivity of an MH tank demonstrates the ability of the tank to transfer heat between the metal hydride bed and the surroundings. This component is influenced by factors such as the type and composition of metal hydride, the temperature and pressure of hydrogen gas, the particle size and porosity of metal hydride bed, the presence of additives or fillers, and the design and configuration of the tank (81-83).

Beyond the thermal effect, a second design aspect is to consider the safe design of tanks considering the successive hydride particle volume expansion during the tank charging process. The volume expansion creates a chain of forces between the hydride bed particles. The amount of these forces depends on the friction coefficient between the particles. This chain of forces between the hydride particles and the hydrogen gas inlet pressure exerts pressure on the tank's wall and causes mechanical stresses. Although this phenomenon can affect the lifespan of the tanks, little research has been done on this topic compared to the thermal issue of metal hydride hydrogen storage tanks (25).

In this review paper, rechargeable hydrides, especially metal hydride and metal hydride composite, are considered absorbent materials in the solid-state method of hydrogen storage. Most studies analyzing the behavior of these materials in metal hydride-based tanks have focused on the macroscopic structure, which can be classified into three categories: (1) heat and mass transfer, (2) pressure and temperature gradient, and (3) stress and strain measure. The mechanical behavior of MH tanks is related to the stress and strain induced inside metal hydride beds; therefore, this type of study will be reviewed in the following sections.

MH tanks are devices that use metal hydrides to store hydrogen gas uniquely. They can store hydrogen at high density and low pressure and usually consist of a metal hydride bed, a heat exchanger, and a pres sure regulator. The tank body is an aluminum alloy or stainless-steel cylinder in horizontal or vertical positions. Their inside area can be divided into (a) the absorbent media region, which is a porous material; (b) the buffer zone where hydrogen accumulates and waits to either enter the porous region or exit the tank; and (c) the inlet and outlet of hydrogen gas from/to the storage tank (Fig. 2. Simplified schematic of the cylindrical metal hydride tank with external heat transfer structure and three main areas: porous region, buffer area, and hydrogen gas inlet/outlet.) (57).

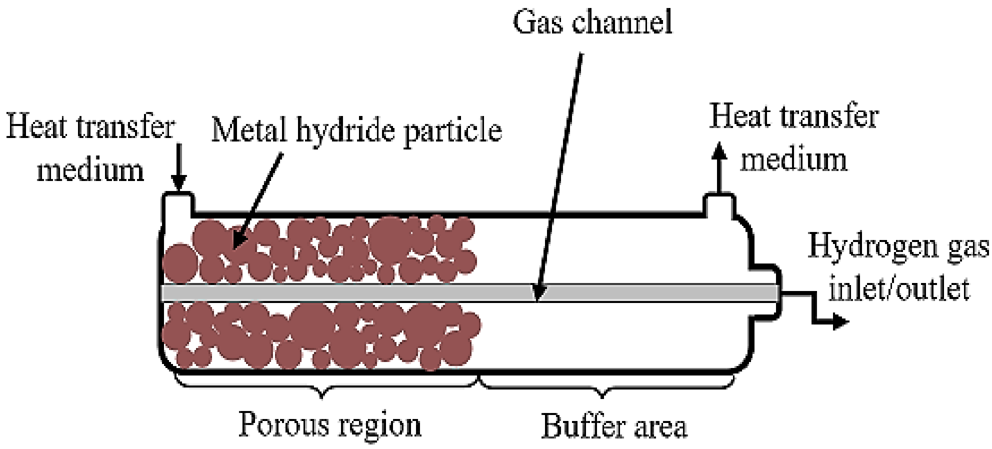

Fig. 2. Simplified schematic of the cylindrical metal hydride tank with external heat transfer structure and three main areas: porous region, buffer area, and hydrogen gas inlet/outlet.

The initial application of MH tanks for hydrogen storage is unclear, as different types of MH materials and applications have been developed over the years. However, many industrial prototypes were made in the 1970s and 1980s. In recent years, numerous researchers have investigated the subject of metal hydrides. Fig. 3. Number of studies published from 2018 to 2022 in the fields of metal hydride, metal hydride tanks, and metal hydride mechanical properties using Google Scholar database. shows the number of research studies that have been retrieved from the Google Scholar database using the keywords “Metal hydride,” “Metal hydride mechanical property,” and “Metal hydride tank.” The studies from 2023 include those up to 2023/04/05. It is clear that the rate of the studies in the two fields of “Metal hydride” and “Metal hydride mechanical properties” have a somewhat constant rate. However, in the “Metal hydride tanks” field, an increasing trend in the number of studies by year can be readily discerned.

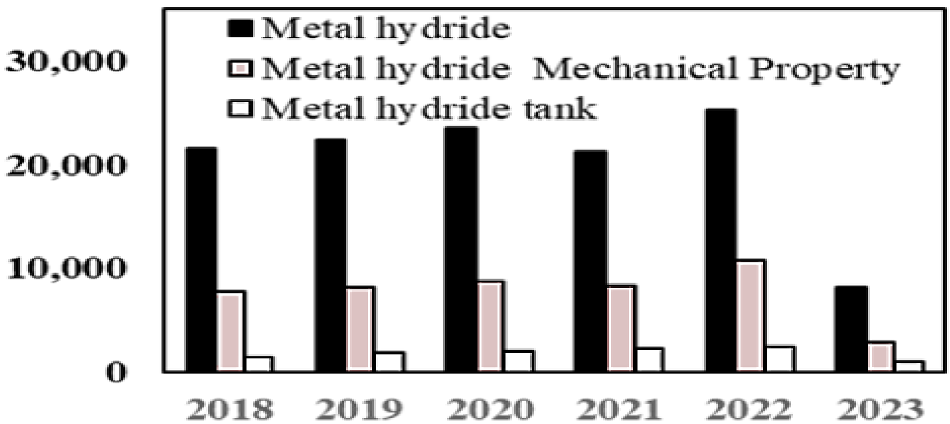

Fig. 3. Number of studies published from 2018 to 2022 in the fields of metal hydride, metal hydride tanks, and metal hydride mechanical properties using Google Scholar database.

The operation of MH tanks is evaluated based on cost, efficiency, durability, and safety (7, 84, 85). The critical factor that plays a significant role in increasing the safety of MH tanks is having a thorough knowledge of their strength and durability. The reaction enthalpy, hysteresis effect, and tank design can affect the stress magnitude and distribution.

The reaction enthalpy of the MH material refers to the quantity of heat that is absorbed or desorbed during hydrogenation or dehydrogenation. At a given temperature, the difference between the absorption and desorption pressures is known as the hysteresis effect. The reaction enthalpy and the hysteresis effect can cause thermal expansion or pressure gradients inside the tank, affecting the distribution and magnitude of the stress in the MH tank (59, 85-88). Moreover, during the charge and discharge operation of the MH tanks, the cyclic expansion and contraction of metal hydride particles take place, which can limit the performance of hydrogen storage tanks due to the stress created between the hydride particles and the tank's wall. Tarasov et al.'s (89) review of the metal hydride hydrogen storage systems stated that the solid fraction of the tank is limited to about 55% due to the stresses that the metal hydride applies to the MH tank structure.

The primary design parameters of the MH tank containers are the slenderness ratio, thickness, shape, position, and solid fraction. The slenderness ratio of a tank refers to the proportion of its length to its diameter. This parameter is typically employed to characterize storage tanks that are oriented vertically. As it determines the available volume and surface area for the MH bed, it can influence the tank's hydrogen storage capacity and efficiency. The higher the slenderness ratio, the more uneven the tank's heat and mass exchange interior, which can cause thermal and mechanical stress on the tank container (25, 35, 76). The solid fraction of the MH bed is determined by dividing its solid volume by its total volume. The higher the solid fraction, the more compact and dense the MH bed, which can cause higher mechanical stress on the tank container (31, 77, 90).

4.1. Experimental studies

The initial stage in comprehending an MH tank's stress distribution and structural integrity involves measuring the volume alteration within the hydride bed. This is accomplished by discerning the volume fluctuation of the metal hydride crystals (91-93). Usually, researchers use dilatometers to measure the change in the hydride bed volume (94). A dilatometer is a scientific device that measures alterations in volume resulting from physical or chemical reactions (95). It measures the volume change in ambient pressure while no constraint is applied to the samples. Dilatometers can be categorized into various types according to the measurement method, such as capacitance, connecting rod, and optical dilatometer.

Some researchers have used the optical dilatometer to measure the metal hydride expansion with the aim of developing optical hydrogen sensors. Optical hydrogen sensors are devices that use the optical properties of metal hydrides to detect the presence and concentration of hydrogen gas. These sensors provide an appealing solution for hydrogen detection across diverse conditions and possess a notable safety edge compared to alternative detection approaches. Notably, they eliminate the necessity for electrical leads in close proximity to the sensing region (96). Briki et al. (97) used a dilatometer to measure the change in volume of $LaNi_5$ compacted powder samples throughout the cycles of hydrogen absorption and desorption. Chen et al. (98) published a review article on optical hydrogen sensors based on metal hydrides. The researchers conducted a comprehensive examination of advancements in optical hydrogen sensors utilizing metal hydrides, employing various sensing methodologies. These encompassed the gasochromic effect, micro-mirror, fiber grating, interferometer, evanescent field, SPR (Surface Plasmon Resonance), and LSPR

(Localized Surface Plasmon Resonance). The efficacy of these optical hydrogen sensors is contingent upon the sensing materials, which dictate the interaction between the sensor and hydrogen gas. Consequently, further enhancements to these sensors hinge on the ongoing exploration and evolution of novel sensing materials.

The height change of the metal hydride bed could specify the volume change of metal hydride particles. One of the simplest methods to measure hydride bed height is to install a light piston on the hydride bed surface (21, 54, 99-101). A spring mechanism is employed in tandem with the piston to maintain consistent contact with the hydride bed surface. This spring mechanism permits the piston to shift in response to variations in the height of the hydride bed brought about by the volumetric expansion of its particles. In this way, the extent of the piston's displacement serves as an indicator of alterations in the volume of the metal hydride bed. However, utilizing the piston and spring system comes with certain limitations. One issue arises from the unevenness of the free surface force within the hydride bed. This disparity leads to fluctuations, which can be mitigated by extending and stiffening the spring. Additionally, this remedy is ineffective in curtailing significant oscillations (54). Another drawback stems from the need for several cycles to establish a level surface for the hydride powder bed. This introduces inaccuracies in the initial measurement of the hydride bed's volume. Consequently, Matsushita et al. (52) put forth a distinct approach for measuring hydride bed height. According to this method, the hydride material is located in a transparent container, and its surface is free. Therefore, the average bed height can also be calculated for non-flat surfaces. Afterward, Ribeiro et al. (55) introduced a smart method to analyze changes in hydride bed volume and porosity at the macroscopic scale. This technique provides valuable data on the changing packing patterns of powder hydride beds as a function of hydrogen concentration.

Within the metal hydride bed, while undergoing the hydrogen absorption process, the particles experience not only volume expansion but also thermal expansion. This latter expansion results from the rise in temperature induced by the exothermic nature of the process. Volumetric and thermal expansion of the metal hydride bed, along with the hydrogen gas inlet pressure, exert pressure on the tank wall. In this regard, measuring or analyzing the stress and strain exerted on the MH tank container is another significant stride in improving the MH tank's design. The volume expansion of metal hydride represents the local normal and tangential stress components introduced to the tank wall from the hydride bed, respectively. The friction between the hydride bed particles and the tank wall causes tangential stress on the wall. So far, the value of this component has not been measured in studies, most likely due to its insignificant value compared to normal stress (25). The amount of normal stress on the wall of the tank is usually measured indirectly by installing a strain gauge on the outer surface. By measuring the strain and knowing the material's behavior, the normal stress on the wall of the tank can be calculated.

Lynch (37), in 1980, was the first to study the mechanical strength of an MH tank container. He experimented with a long horizontal copper tube containing metal hydride. He observed that the long copper tube failed only after a few hydrogen absorption and desorption cycles. To prevent this defect, he suggested double layering of the tank. Because the exact wall thickness of the internal hydride tank and the quantity of solid absorbent material contained within it remain unknown, it was not feasible to determine the critical solid fraction value within the scope of the study. Au et al. (102) were the next to investigate the strength of a long horizontal MH tank. They equipped the MH tank with a strain gauge to study the expansion of the hydride bed effect on the MH tank container and used a simple composite bed of hydrogen storage alloy and aluminum fiber to decrease the strain in the storage vessel. They concluded that improving the hydride bed's thermal conductivity can decrease the strain resulting from volume expansion. As stated in this test, for the solid fraction higher than 0.45, there is a risk for high stresses. Au et al. (102) categorized the methods to reduce tank damage due to metal hydride breathing into:

- Strengthening the container walls
- Double-wall containers
- Lubricating hydride powder bed with non-volatile oil
- Loosely packing the powdery materials in the container
- Splitting the hydride bed into smaller sections to stop the powder from shifting
- Porous metallic matrix hydrides
- Composite materials

According to the experiment carried out by McKillip (103) in 1992, after several cycles of absorption and desorption, the stress value reaches a stable level, and the more the solid fraction, the more the amount of strain in the tank wall (104, 105). The results of Estochen's experiments in 2005 (106) and 2017 (107) showed that the stress value increases as a function of the hydrogen absorption rate, and its maximum value occurs exactly after the activation cycle. Consequently, the stress level undergoes a rapid and pronounced increase throughout the absorption process, followed by a swift reduction to zero upon discharge. In 2008, by comparing two tubes containing vertical and horizontal MH tank containers, Qin et al. (108) found that during the expansion of the hydride bed, the horizontal tube tends to bend, while the vertical tube expands in the radial direction.

Subsequently, in a later study by Duan et al. (109), an investigation was conducted into strain variations throughout hydrogen absorption and desorption cycles in a horizontal tank housing TiFeMn alloy. This research involved the attachment of ten strain gauges to the tank's surface to gauge its deformation. The findings from this inquiry demonstrated that deformation becomes noticeable after the activation phase, with a considerable magnitude of strain, and the distribution of this strain does not remain consistent along the length of the cylinder. The uneven introduction of hydrogen into the cylinder, coupled with the formation of gaps at the cylinder's ends, results in the maximum strain occurring at the center of the tank. Kubo et al. (80) presented a new hydrogen absorber method to form the MH powders with polymer materials and reduce the strain in the MH tank. They also increased the filling of MH tanks more than conventional MH tanks. Lototskyy et al. (110) also suggested using one wt % of expanded natural graphite (ENG) to reduce the stresses on the MH tank walls due to the metal hydride bed expansion.

In summary, exceeding the critical solid fraction by loading excessive hydride into horizontal MH tanks results in elevated stresses, permanent deformations, and a decrease in safety. When designing and evaluating such tanks, it is prudent to prioritize the solid fraction during the absorption phase, given the abundance of absorbent material at that point. The specific value of the solid fraction is contingent on various factors, including the nature of the hydride particles, their composition, and operating conditions such as temperature, pressure, and loading parameters. The studies discussed in this section highlight that the stress imparted to the MH tank is influenced by the number of absorption/desorption cycles in the process.

Within vertical MH tanks, the critical strength parameter is the height. This attribute is encapsulated by the concept of the slenderness ratio, which is interconnected with the friction existing between the hydride particles and the walls of the MH tank. The first study to investigate the stress in vertical MH tanks was done by Kawamura (111) in 1981. Afterward, Wang et al. (112) observed that the metal hydride bed's density can increase the container's strain and deformation. They advocated for imposing restrictions on the extent of container filling. Additionally, they employed a metal hydride composite to avert the disintegration of the metal hydride material. In vertical MH tanks, the increase in stress in the hydride bed leads to a decrease in the hydrogen absorption capacity (113), and the amount of strain changes according to the value of the solid fraction (114, 115). According to Lin's studies published in 2011 (116) and 2012 (117), the existence of the central porous tube in the tank improves the hydrogen storage capacity and the absorption reaction rate. In addition, the amount of tank wall deformation is also reduced. These experiments proved that the cellularization of the hydride bed volume reduces the amount of stress on the wall and prevents the densification of hydride particles after crushing. Recently, Borzenko et al. (118) tested two horizontal and vertical tanks with the same conditions and material. The results of this research showed that in the vertical state, the compressive stress of the hydride material powder is higher than in the horizontal state, and the equilibrium pressure in the vertical state amid the hydrogen absorption process is lower than in the horizontal state, which means that this type of tank has a higher storage capacity. In summary, in vertical tanks, the maximum stress occurs at the bottom of the tank, and the amount of stress changes according to the slenderness ratio. In such tanks, the relationship between the solid fraction and the evolution of stress change is not clearly defined and depends on the type of material.

According to earlier studies, due to the difficulty and cost of certain measurement methods, strain gauges are, as a rule, utilized to measure stress. In addition to this method, it is possible to install in-situ systems to measure desired parameters in metal hydride-based hydrogen storage tanks. One of the common methods to check the volume change of a hydride bed is X-ray diffraction (XRD) (119). This method is based on applying X-ray radiation to the sample and analyzing its diffraction or reflection pattern, which provides information related to the crystallography of the sample. The detection of the crystal phase, the size and shape of the crystal grain, the distance between the crystal layers, determining the orientation and position of the crystal, measuring the crystallinity percentage of the sample, the composition of crystal atoms, and their structure are among the things which can be determined with this analysis. Therefore, it is a suitable method to monitor the changes in phase structures during hydrogen absorption and desorption reactions in storage tanks. Since 1990, multiple studies have been conducted to understand the hydrogen storage process using this specific method. Measuring the strain in the metal crystal lattice during hydrogen absorption was achieved for the first time using the X-ray diffraction method by Nakamura in 2000 (120). There are various approaches available to determine the extent of stress present in the hydrogen storage tank, for example, the use of stress-sensitive foils, which can detect the position of the maximum stress due to the intensity of the red color of the foil, or the use of an in-situ force measuring cell which is embedded in the storage tank itself (87, 121, 122).

4.2. Analysis and simulation studies

From an engineering point of view, it is fundamental to supply a model to depict the alteration of the hydride bed and its interaction with the tank. The development of phenomenological laws is used to achieve this goal, assuming the powder substance is a continuous medium. These laws are the elastoplastic compressive laws, including the DPC and Cam-Clay models. The DPC model simulates the powder compaction of hard and dry particles, and the Cam-Clay model is used for soft particles (123). These models are executed by computer programs called finite element codes, such as ANSYS, CASt3M, ABAQUS, and COMSOL (124).

In 2013, Charlas (125) simulated and calculated the change in volume of the hydride bed by using Abaqus software. This study used the DPC model because it was more compatible with the hydride powder made of hard and dry particles. The introduced model predicts an increment within the volume of the hydride bed after passing one cycle, while in the experimental sample tested for this material, a decrease in the volume of the hydride bed was observed. The inappropriateness of the evolutionary laws of DPC model parameters causes this contradiction. These parameters were developed to model uniform loading during compression, which is very different from the problem of the hydride breathing phenomenon. Some studies modeled the hydride bed volume change with phenomenological laws, assuming the pure elastic deformation of the granular medium (126-131). In these articles, modeling was done using the analytical and finite element simulation methods with Ansys and Comsol software. The assumption of pure elastic deformation of the hydride bed oversimplifies the problem. It cannot model the change in the hydride bed density when it absorbs or releases hydrogen. Charlas et al. (132) modeled the deformation of the hydride bed using the discrete element method and compared it with the experimental results. In that research, the design parameters such as contact hardness, friction between particles and the wall, and the shape of the particles were obtained through die compaction and rotating disc tests. The results from the simulation method matched well with the results from the actual experiment. However, this model does not fully predict all cases and has some shortcomings. In this method, the rapid increase of the solid fraction at the beginning of the absorption process is not considered. This increase is caused by the sudden change in morphology and particle size due to crushing at the beginning of the absorption process. Another flaw in this model is the unreal value of the solid fraction because the assumption of the spherical shape of the particle leads to the assumption of the wrong initial solid fraction for the tank.

Conclusion

The present report aims to exhibit an integrated view of how the hydride bed acts and behaves mechanically in hydrogen storage tanks and extract the rules for its proper design. Therefore, the results obtained from previous studies in this area were expressed and compared. In general, parameters such as solid fraction, slenderness ratio, number of cycles, thermal conductivity coefficient of metal, type of material, hydrogen gas inlet pressure, thickness of hydride bed, geometry and shape of the tank, the temperature gradient of hydride bed, amount of absorbed hydrogen, crushing of particles and also their density and accumulation are effective on the amount of stress created in the hydride bed. In order to reduce and control the stress caused by the volumetric and thermal expansion of the hydride particles, solutions such as strengthening the tank wall, double layering the tank wall, applying more empty space between the hydride particles, dividing the hydride bed into smaller units, and using lubricants can be used. Based on the reviewed research, a comprehensive and accurate model for predicting the mechanical behavior of the hydride bed of hydrogen storage tanks has not been presented yet. The digital image correlation (DIC) optical technique, which is non-destructive, highly accurate, and designed to measure deformation from micro to macro scale or acoustic emission test, can be used to achieve this objective. Another interesting point would be improving the strength of the MH tank's structure by applying composite material to the MH tank's walls.

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