Modeling and Simulation of Hydrogen Storage Device for Fuel Cell Plant

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Abstract: The article reviews a brief literature on the modeling of hydrogen storage device for fuel cell. Different dimensional approaches in modeling hydrogen absorption/desorption in a metal hydride reactor for use in fuel cell are summarized. Mathematical modeling equations involved are also stated. The effect of various operating parameters such as temperature, concentration, viscosity, thermal conductivity and time on the gas is also verified. The importance of various simulation software with reference to their major functions is also identified. The review concludes on the opportunities and challenges with the use of hydrogen as an alternative renewable energy.

Key words: Modeling, simulation, hydrogen, storage, fuel cell.

Nomenclature

C_p	Specific heat ($JKg^{-1} K^{-1}$)
E	Activation energy (J mol^{-1})
h	Conductance between hydride bed and around fluid ($Wm^{-2} K^{-1}$)
h_o	Conductance between outlet face of the reactor and exterior medium ($Wm^{-2} K^{-1}$)
H	Reactor height (m)
Hgs	Heat coefficient exchange between between solid and gas ($Wm^{-2} K^{-1}$)
H/M	Hydrogen and metal atomic ratio
k	Permeability (m^2)
m	Hydrogen mass absorbed or desorbed ($Kgm^{-3} s^{-1}$)
M	Molecular weight (kg mol^{-1})
P	Pressure (Pa)
R_g	Universal gas constant ($Jmol^{-1} K^{-1}$)
R	Reactor radius (m)
T	Temperature (K)
t	Time (s)
V	Gas velocity (ms^{-1})
ΔH^o	Reaction heat of formation (JKg^{-1})
ε	Porosity

λ	Thermal conductivity ($Wm^{-1} K^{-1}$)
μ	Dynamic viscosity ($Kgm^{-1} s^{-1}$)
ρ	Density (Kgm^{-3})
ω	Average volume (m^3)
a	Absorption
d	Desorption
e	Effective
eq	Equilibrium
f	Cooling or heating fluid
g	Gas
ge	Gas effective
se	Solid effective
s	Solid
ss	saturated
0	Initial
g	Gaseous phase
s	Solid phase
-	Average volume

1. Introduction

Energy demand across the globe has risen sharply over the last decade. Today, fossil-fuel based sources of energy are being depleted at a fast rate because of the ever increasing energy demand and consumption. Additionally, fossil-fuels are contributing to both greenhouse gas emissions and global warming [1]. Due to these hazardous effects, there should be an urgent need for alternative and cleaner fuels which will be toxic free and environmentally friendly. Hydrogen energy appears to be the best alternative for the future to replace the fossil fuels and it already has several industrial applications such as in heat pumps and automotive industries. One of the main problems in large usage of hydrogen in automotive industry is the storage problem. The conventional hydrogen storage methods in use for hydrogen gases are gas compression and liquidification. These storage methods are impractical since the former requires very heavy gas tank and the latter is too expensive to be employed in public vechicles. Storing hydrogen in metal hydride beds as a chemical compound appears to be method of hydrogen storage in the near future, and this has received much interest recently [2]. There are several important challenges to overcome before hydrogen can become a viable fuel. Only 1% of the hydrogen is available as molecular hydrogen gas while the majority is present in the form of water or hydrocarbons. Large scale production of hydrogen from water or chemical compounds in an efficient and clean method remains a challenge. Another challenge is storage volume. Though hydrogen has high energy content by weight (three times that of gasoline), its energy content by volume is only one-tenth that of gasoline. The process is major storage volume problem in automotive and other mobile systems with stringent volume constraint. A third challenge is hydrogen transport to filling stations. Because of the low density of hydrogen,

finding an economical way of transporting large amounts of hydrogen to various locations is quiet illusive. The fourth challenge is the use of hydrogen as a fuel in producing clean energy that would justify its wide acceptance in the market [1]. Hydrogen can be used to produce energy either by burning with oxygen in air (e.g., internal combustion engine), or by electrochemical reaction with oxygen using fuel cells. Fuel cells are both energy efficient and virtually pollution free compared to internal combustion engines. The choice of energy production method is dictated mainly by cost and application. In summary, the advantages of using hydrogen as fuel can be realized only if all the relevant processes, from production to storage to energy generation, are clean and efficient [1].

2. Fuel Cell Modelling

Many researchers have made an investigation on the effect of parameters on the performance of fuel cells. The parameters, which have been investigated, include operating parameters (i.e., temperature, humidity, pressure, flow rate etc.), design parameters (i.e., geometrical parameters etc.), physical parameters (i.e., porous media, porosity, permeability etc.), electrochemical parameters (i.e., specific area of catalyst layer timed by reference electrical density) and other parameters (i.e., CO poisoning etc.).

Further research on modeling of fuel cells is grouped into different dimensional models. These models are grouped into one-dimensional model, two-dimensional model and three-dimensional model.

2.1 Modeling of Fuel Cell Based on One-dimensional Model

Djilali and Lu [3] came up with a non-isothermal model to analyze the cell performance. A staggered grid volume method was used along with a fully implicit iterative solution algorithm with under-relaxation. A standard Thomas algorithm for tri-diagonal matrix inversion was used at each iteration

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step. Pisani and Murgia [4] examined the flooding phenomena wave effect of different agglomerate radii within the catalyst layer. The model domain consisted of a hydrophobic and hydrophilic layer.

The problem was solved using COLSYS that adopted an adaptive method for grid generation and subsequent collocation with B-splines. Nam and Kaviany [5] presented a detailed work on diffusion media. A network model was used to account for material parameters and their dependence on saturation. The problem was mainly solved by the gauss-siedel iteration with slight over-relaxation. The convergence criterion was set to 1×10^{-6}

The work of Yan et al. [6] addressed detailed water and thermal management issues including the impact of various parameters. A control volume finite difference method by Lee, [7] was used (1000 grid points). Linet et al. [8] modeled liquid phase water effect in the porous media using an isothermal cathode side model. It was reported that the flooding situation within the catalyst layer is more severe than in the gas diffusion layer. The results were obtained using a banded matrix solver presented by Liu, [9]. Ziegler et al. [10] presented a dynamic isothermal, two-phase model to stimulate cyclo-voltammograms. An interesting numerical solution method was used here in the stationary solution is calculated using COMSOL Multiphysics [FEMLAB] to provide good initial conditions for the dynamic analysis. The governing equations were transformed into a weak form, connected to various principles, the time dependent solution was calculated using an implicit scheme because of different time scales.

2.2 Modeling of Fuel Cell Based on Two-dimensional Models

Nguyen and white [11] derived a pseudo-2D water and heat management model. The catalyst layer was treated as an interface. Algebraic expression and linear assumption were used for the concentration along the electrode the relationship between the transport number and electro-osmotic drag coefficient was presented. In addition, the current density distribution was analyzed. A Newton-Raphson based iterative solution technique was used to compute cell voltage for along-the-channel calculations, a fourth order Runge-Kutta method was used. Fuller and Newman, [12] used concentrated solution theory to account for diffusive transport through the membrane and showed that adequate heat removal is essential for preventing membrane dehydration and to maintain high performance. Moreover, the effects of dry gas feeds were studied.

Gu et al. [13] developed a non-isothermal along-the-channel model. Current distribution and species concentration along-the-channel were computed using the simple algorithm along with complex iterative procedures. Yi and Nguyen [14] analyzed flow field design and temperature distribution, and liquid water effects at different operating conditions. Further cooling strategies were examined, these model were solved using BAND (J) algorithm. Kulikovsky et al. [15] presented a detailed cathode side sandwich model focusing on catalyst loading and architecture. The governing equations were converted to a finite difference method by the control volume method. An orthogonal non-uniform grid was introduced. A five point's approximation for divergence was derived using the Sharfetter-Gummel relations. The potential were calculated using a derived system of linear-equation solved with a standard symmetrical successive over- relaxation technique. Another dynamic isothermal model is the two-phase model of a cathode gas diffusion layer, which was developed by Natarajan and Nguyen [16]. An empirical function was used to relate capillary pressure and saturation. This study delivered information on the effects of varying operating conditions and design parameters; they also showed that porous media saturation takes place on the order of minutes for the profile to develop. A branded matrix solver was used by Wang et al. [17].

Mennola [18] used an isothermal, along the channel model to account for two-phase flow and transport within the cathode side gas channel and gas diffusion layer. Both phases were classified by a threshold current density. A two-phase multi component mixture model was applied here in; it was found that capillary is the dominant mechanism for water transport. Inside the two-phase zone of a hydrophilic structure, emphasis was put on a smooth transient between the two phases. The catalyst layer was modeled using a reactive boundary layer. Ziegler et al. [19] presented a single-phase model of a planar, self-breathing fuel cell to account for cell performance. A strong non-uniformity of the current density was found.

FEMLAB (COMSOL Multiphysics) was used on a computational domain with normalized coordinates. For the charge balance, a non-linear solver based on an affine invariant form of the damped Newton-Raphson method was used. A simplified along the channel model (1 + 1 dimension with 1,048 grid points) was used to account for water management by Thoben and Siebke [20], CO and counter flow were considered for several operating conditions. The catalyst layer was treated as a boundary and an effective water drag coefficient was assumed. ODEs (ordinary differential equations) were solved iteratively using MATLAB/SIMULINK within 10 iterations (convergence error less than 10%). Water management was analyzed by Shimpalee, [21] with an emphasis on gas diffusion layer material properties, modeling result matched with experimental data.

2.3 Modeling of a Fuel Cell Based on Three Dimensional-Models

In 1999 and 2000, Shimpalee et al. [22] presented multi dimensional fuel cell models, they solved the complete Navier-Stoke equation within the gas channel. Several physical properties and operating conditions were studied; they considered the liquid water as a component of the gas mixture transported by

convection due to total gas pressure and diffusion by density gradients. FLUENT and simple algorithm was used to solve for the conservation equations. Both studies used 190 OOO-mesh element.

Li et al. [23] analyzed flow field behaviour using the simple algorithm and a complex solution procedure. Lee et al. [24] presented a-non-isothermal two-phase model coupled to experimental investigations. In this model, membrane electrode assembly was not included into the computational domain but rather simplified as an interface without thickness. A· good agreement with experimental investigation was found and emphasis was put on the current density distribution and spatial membrane conductivity distribution. Nguyen et al. [25] presented a model including the gas channels, all-major conservation equations were included based using a single-phase approach. The model consisted of 351, 000 mesh element and was solved using CFX. The set of algebraic was solved with a full fieldstone's method for the momentum and scalar transport equation, AMG was used for the pressure and energy equation and a FORTRAN-based VTC (voltage-to-current) algorithm for accurate calculation of the local activation over potential and current distribution. Depending on the operating voltage 6,000-8,000 iterations were required to achieve convergence. Meng and Wang [26] came up with an isothermal, two- phase model based on a multiphase mixture formulation and investigated for the liquid water behaviour and cathode flooding.

Sivertsen and Djilali [27] presented a single-phase model. FLUENT was used to solve the problem on a linus cluster with dual 2000 + AMD Athlon processor. The numerical results matched with experimental result at low-to- medium current densities. Lum and McGuirk [28] presented an isothermal model, solving for a minimum of species to reduce computational costs. They used an in-house CFD code fuel 3D together with the simple algorithm on co-located grids. The problem was solved on a 24-processor origin 2000 computer (CC 1×10^{-6}).

3. Previous Modeling and Simulation on Metal Hydride Storage Tank

Developing effective solid-state hydrogen storage for transportation is crucial for the success of the hydrogen economy, besides energy density and system cost requirements. One important requirement by transportation application is fast kinetics of hydrogen storage i.e. quick hydriding and dehydriding. Efficient release of hydrogen gas in metal—hydride reactor should met the need of fast load variation. In order to build and satisfy such hydrogen systems, understanding of physics of the transport process coupled with reaction kinetics is very important. Such transport processes include hydrogen mass flow in the hydride bed, heat transfer within the bed and local hydrogen absorption rate. Previous studies on modeling of hydrogen storage by several groups can be summarized. Zehner and Schlunder [30] presented a model based on a one-dimensional heat flow model for conduction through a packed bed of spherical particles where they assumed point contact for particles in the direction of heat flow.

Woodside and Messmer [31] suggested a model which was a combination of series and parallel distributions they used an aggregated of conductive particles saturated with conducting electrolyte to derived a modified resistor model equation to predict the effect thermal conductivity. The group of Jenni

and Nasrallal has published a number of articles (Jemni and Nasrallah [32]) describing heat and mass transfer at the outer surface, where hydrogen leaves and enters at the top of the reactor. They used their model to study the validity of the simplifying assumptions used by various authors (like the assumption of thermal equilibrium between gas and solid) and the effect of different physical modeling input parameters such as thermal conductivity extended the analysis to three dimensions for a cylindrical geometry and verified their model using interactive data. The assumption of thermal equilibrium between gas and solid and the importance of heat correction were analyzed by Nakagawa et al. [33] for a cylindrical reactor where hydrogen was introduced at the top of the reactor.

They also considered the influence of variable physical properties such as heat capacity as a junction of reacted fraction. Their results indicated that both the assumption of thermal equilibrium and neglecting the convection term in the energy equation influenced the total amount of hydrogen desorbed form the reactor. Kuznetsor et al. [34] presented analytic criteria for the validity of some of the most commonly applied assumptions, together with an analytic solution of an idealized heat and mass transfer situation.

Gambini [35] presented a simplified zero dimensional model assuming no temperature gradient
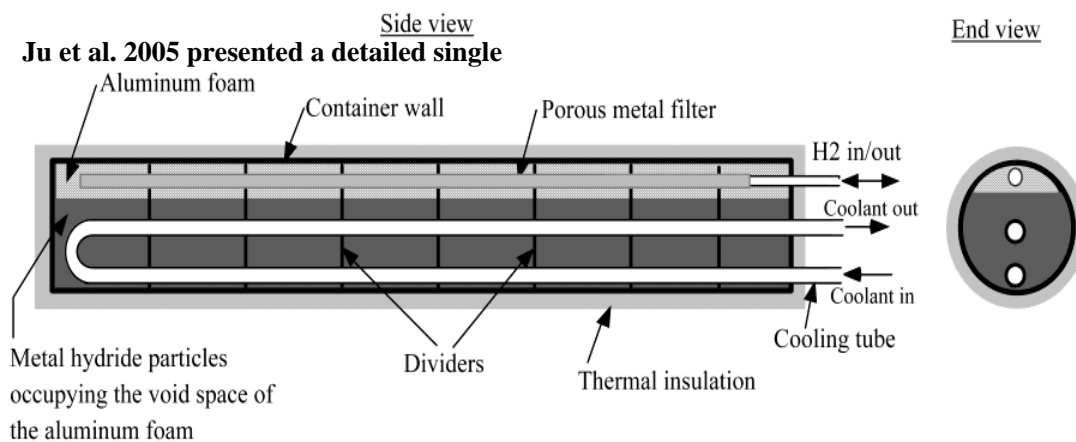
Ju et al. 2005 presented a detailed single

Fig. 1   Metal hydride storage tank (Gadre, S. A. et al. [29]).

in the metal hydride (i.e., a stirred reactor type) which was used to study the transient behaviour of two coupled metal hydride reactors. Gopal and Murthy [36] development a one dimensional heat transfer model based on the experiments carried out on a cylindrical metal—hydride reactor filled with $M_mNi_{4.5}Al_{0.5}$. Shmalkov et al. [37] proposed a theoretical model of heat and mass transfer in metal hydride hydrogen gas impurities system. Their model makes it possible to describe the sorption processes taking into account the convective transfer in the metal-hydride modules for gas mixtures containing impurities that are inactive or weakly active to the hydride forming material.

Jemni and Nasrallah [32] developed two mathematical models, one based on separated solid and gaseous phases as a mixture. The effect of several parameters on the overall hydrogen storage efficiency was illustrated. A transport equation for both gas and solid were solved, and the results were compared with a continuum model, which consider metal and a gas as a continuous model. They have shown that solving the problem with a single phase like model does not significantly change the result, they also justify that the convection is insignificant in the hydriding process. Jemni and Nasrallah [32] presented an experimental approach to determine the reaction kinetics, equilibrium conditions and transport properties in $LaNi_5$-H_2 system.

Mat and Co- workers et al. [38] carried out a detailed analysis and studied the parameters affecting the hydriding process, and extended the analysis the analysis to three dimensional cases. Askri et al. [39] extended the previous studies by investigating the effect of radioactive heat transfer on sorption processes. Kaplan [40] extended the work of Mat, [41] and investigated numerically the hydrogen storage process in a 2D metal—hydride bed including the full momentum balance equation which is considered to be important when large pressure gradients exist in the system. A simulation of a metal hydride–hydrogen storage device with embedded filters and cooling tubes

is treated by Mohan et al. [42]. Results of the simulation confirm the importance of bed thickness as a major parameter controlling the rate of hydrogen absorption at all locations within the bed. They observed that the heat exchanger tube diameters have marginal influence on hydriding time at higher values.

Mellouli et al. [43] experimentally examined the hydrogen sorption in a metal hydride tank equipped with an internal heat exchanger. They found that the charge/discharge times of the tank are considerably reduced. Jemni and Nasrallah [32] presented a theoretical study of the mass and heat transfer dynamics in a metal hydride reactor. In subsequent papers they validated their assumptions as well as presented experimental approaches to determine the reaction kinetics, equilibrium condition and transport properties in a $LaNi_5$-H_2 system. Nakagawa et al. [33] presented a 2-D model for the transient heat and mass transfer within a metal hydride bed. Kikkinedes et al. [34] presented a detailed mathematical model which included both the axial and radial dimensions of the storage bed. Apart from the heat and mass transfer effects a cooling medium for the system was modeled and optimized by them.

In the one dimensional model used by Mayer et al. [35], the gas and the solid temperature are assumed to be equal and that gas pressure is constant in the reactor. The model used by Choi et al. [36] takes into account the gas pressure variation in the reactor. The three dimensional model used by Jenni and Nasrallah, [32] takes into account the hydrogen flow and assumes that solid and gas are in thermal equilibrium, they conducted an experimental and numerical study to determine the effective thermal conductivity, the equilibrium pressure and reaction kinetics.

Demircan [37] experimentally examined the hydrogen absorption in two $LaNi_5$-H_2 reactors. They found that hydriding time mainly depends on the successful heat removal from the bed, and a bed geometry that provides more heat transfer area significantly reduces hydriding time. Boser [38] was

the first to investigate the hydrogen sorption by activated LaNi$_5$, they used pressure variation in a constant volumetric apparatus. Boser performed his experiment in the 273-363 k temperature range, limited their observation at the low temperature of 195 K, to slow down the reaction rate. They were able to developed different arguments from their experimental data to justify the choice of a possible mechanism, controlling the hydride precipitation. Thus, Boser proposed the phase transformation as a regulating step, limited mass transfer limitations through crocks within the particles or in the channels were advanced by eliminating all the chemical processes which were considered to be too fast.

Choi and Mills [39] used one-dimensional mathematical model to investigate several effects including thermal conductivity, bed thickness, charge pressure on performance of the hydride bed and optimum bed size for heat pump applications. They found that the optimum bed size should be around 10-20 mm for a 3 min absorption time. Nakagawa et al. [40] carried out studies on hydriding process with a two dimensional model .they observed that the convection initially enhances the hydriding processes and has an adverse effect at later times.

4. Software for Modelling

4.1 Cfd Software for Modeling

Various computational fluid dynamic software for fuel cell modeling and simulation are available such as FLUENT, COMSOL, Multi physics [FEMLAB], STAR-CD, CFD-ACE+, NA DIGEST FDEM, OPENFOAM, SCILAB. This software can produce the following functions:

• Solver set up (e.g., flexibility, relative error tolerance, damping factor, preconditioned);

• Speed and computational accuracy;

• Complexity (e.g., possibility to tune and add source code by hand, to understand the algorithms, unmasked subroutines and coding);

• Parallel computing possibilities (e.g.,

Communication protocol);

• Interface to other software (e.g., import/export, CAD-interface);

• Time set up (e.g., fixed, variable, real-time);

• GUI (graphical user interface) for interpretation and visualization of the modeling results;

• Documentation and information resources for ready-to-run models and tool boxes.

• Commercial or open-source code.

4.2 Modeling Equations

The Equations shown below illustrate previous modeling equations used. Two-dimensional non-stationary mathematical model of the metal hydride reactor for hydrogen storage consists of mass, momentum and energy balance equations together with convectional equation for pressure and temperature.

4.2.1 Mass Balance

The mass conservation equation of the solid metal hydride:

$$(1 - \varepsilon)\frac{\partial \rho_s}{\partial t} = m \qquad (1)$$

Mass balance equation for hydrogen gas:

$$\varepsilon \frac{\partial \rho_g}{\partial t} + \nabla (\rho_g u_g) = m \qquad (2)$$

where ρ_g denotes the density of the hydrogen gas in the reactor during the desorption process. Hydrogen density is determined using the perfect gas law. Velocity of hydrogen gas (u_g) is calculated using the Darcy's law [32].

$$u_g = \frac{K}{\mu_g} \nabla P_g \qquad (3)$$

where K is permeability, μg is gas viscosity, Pg is gas pressure.

Distribution of hydride powder particles by size and shape makes widening and smoothing of adsorption/desorption curves. Complex mechanism of hydrogen reaction with metal hydride is not well known, and macrokinetics expressions are obtained from experimental data for each particular metal hydride powder.

Equation for hydrogen kinetics (hydrogen mass desorbed, m, per unit time and per unit volume) is given by Dhaoua et al. 2007):

$$m = C_d \exp\left(-\frac{E_d}{R_g T}\right) \frac{P_g - P_{eq}}{P_{eq}} \rho_s \quad (4)$$

For the LaNi$_5$-Hydrogen system $C_d = 9.57$ L/s; $E_d = 16.420$ KJ/mol of H$_2$.

Equilibrium pressure P_{eq} can be calculated from modified van't Hoff equation (Nishizaki T., et al.).

$$\ln\left(P_{eq}\right) = \frac{\Delta H}{RT} - \frac{\Delta S}{R} +$$

$$(\phi - \varphi_0)\tan\left[\pi\left(\frac{C}{C_m} - \frac{1}{2}\right)\right] + \frac{\beta}{2} \quad (5)$$

where, φ (0.35) and β (0.25) are the plateau flatness factor and the plateau hysteresis factor.

4.2.2 Momentum Balance

Momentum balance equation includes Darcy's term to account for momentum transfer due to pressure gradient in the metal hydride porous media:

$$\frac{\partial(\rho_g \varepsilon)}{\partial t} + \nabla\left(-\rho_g \frac{K}{\mu_g} \nabla P_g\right) = 0 \quad (6)$$

where:

Permeability K and porosity ε are related by equation:

$$K = C_k \cdot d_p^2 \left(\frac{\varepsilon}{1-\varepsilon}\right)^2 \quad (7)$$

where d_p is metal hydride particle diameter and constant $C_k = 2.37 \times 10^{-3}$.

4.2.3 Energy Balance

Energy balance equation, describing temperature evolution of the hydrogen—porous bed system:

$$\frac{d}{dt}\left(\varepsilon\rho_g C_{pg} T + (1-\varepsilon)\rho_s C_{ps} T - \varepsilon\frac{\rho_g RT}{M_{H_2}}\right) +$$

$$\nabla\left(\rho_g C_{pg} uT - \lambda_e \nabla T\right) + (1-\varepsilon)(-\Delta H)\rho_s m = 0 \quad (8)$$

Effective thermal conductivity of metal hydride bed traditionally expressed as:

$$\lambda_e = \varepsilon\lambda_g + (1-\varepsilon)\lambda_s \quad (9)$$

4.2.4 Current Research Works

This current Research works on modeling of hydrogen storage in a metal hydride reactor is considering a cylindrical reactor tank containing hydrogen gas and metal hydride aimed at achieving the following set objectives:

• To model and simulate the absorption and desorption rate of hydrogen stored in a metal-hydride storage tank;

• To select the fast, efficient solver that will be use for the simulation of hydrogen gas in metal hydride storage units;

• Study the interaction between a metal hydride storage units and a fuel cell through simulation;

• To develop a mathematical model in order to simulate heat and mass transfer in a packed bed reactor with metal hydride as a material for hydrogen absorption and desorption;

• To use software COMSOL MULTIPHYSICS to reduce, solve and model PDE (partial differential equations) to process modeling equations;

• To determine the effect of the following variables on the stored hydrogen gas in a metal hydride reactor tanks through modeling and simulation. The variables are pressure, concentration, temperature, thermal conductivity and porosity.

5. Conclusions

Hydrogen gas can be modeled effectively, an optimal design is also important for achieving fast refueling of hydride storage tanks. Achieved results on previous studies laid emphasis on absorption process mostly but desorption of hydrogen in a metal hydride storage tank is also very crucial for the optimum yield and performance of hydrogen fuel cell.

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