Study of coupled heat and mass transfer during absorption of hydrogen in $\text{MmNi}_{4.6}\text{Al}_{0.4}$ based hydrogen storage device

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Abstract. A two-dimensional numerical analysis of coupled heat and mass transfer processes in a cylindrical metal hydride reactor containing $\text{MmNi}_{4.6}\text{Al}_{0.4}$ is presented. To understand the hydrogen absorption mechanism the governing equations for energy, momentum and mass conservation and reaction kinetic equations are solved simultaneously using the finite volume method (FVM). Performance studies on $\text{MmNi}_{4.6}\text{Al}_{0.4}$ based hydrogen storage device are carried out by varying the hydrogen supply pressure, absorption (cooling fluid) temperature, overall heat transfer coefficient and hydride bed thickness. Effect of convection terms in the energy equation on hydrogen storage performance is found to be negligible. The results obtained from the computer simulation showed good agreement with the available experimental data. At the supply conditions of 30 bar and 298 K, $\text{MmNi}_{4.6}\text{Al}_{0.4}$ stores about 1.28 wt%, which is very close to the experimental value of 1.3 wt%. Overall high heat transfer coefficients are found to reduce the absorption time significantly.

Keywords. Hydrogen storage; metal hydride; coupled heat and mass transfer.

1. Introduction

A mathematical model of coupled heat and mass transfer in metal hydride reaction bed is particularly used to assist in interpreting the absorption and desorption rates of hydrogen, to design the hydrogen storage device, and to assist in system optimization. Various investigators attempted to study such coupled heat and mass transfer phenomena encountered in the hydride bed, particularly focused on the improvement of absorption and desorption rates. Ram Gopal & Srinivasa Murthy (1992; 1993) investigated the effects of operating parameters such as supply and delivery pressures, and cold and hot fluid temperatures on hydriding and dehydriding characteristics of the metal hydride beds of cylindrical and rectangular configurations using one-dimensional transient heat conduction model in dimensionless form. Later, they carried out the experimental studies on hydrogen storage device filled with

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MmNi_{4.7}Al_{0.3} for different supply pressures (Ram Gopal & Srinivasa Murthy 1995). But their numerical models are limited to thin bed with negligible pressure drop across the bed. Mayer *et al* (1987) concluded from their numerical investigation that an excellent heat and mass transfer inside the reaction bed, good thermal contact of the bed material to the bed wall, and high external heat transfer coefficients were the important parameters to make the hydrogen absorption/desorption reaction efficient. Guo & Sung (1999) revealed from their numerical predictions that the effect of insertion of aluminum sheet in the metal hydride beds augments the heat transfer and reaction rate significantly. Jemni and his group predicted the dynamic heat and mass transfer characteristics in metal hydride bed during both absorption and desorption processes using two-dimensional model considering the variation of gas pressure inside the hydride bed (Faouzi *et al* (2004a; 2004b) and also the effect of radiative heat transfer (Askri *et al* 2003). The major observations from their models were the faster absorption rate at higher supply pressures, negligible convection effect, and the significant radiative heat transfer only for Mg based alloys. Ha *et al* (2004) developed a two-dimensional mathematical model for unsteady heat and mass transfer in metal hydride bed during the hydriding process occurring in the bed. Their results showed that the whole process occurring in the bed is governed and controlled by the heat transfer from the bed to the surrounding cooling fluid.

Muthukumar *et al* (2005; 2007) reported the hydrogen storage performances of MmNi_{4.6}Fe_{0.4}, MmNi_{4.6}Al_{0.4} and Mg₂Ni at different supply pressures. They observed that the hydrogen storage capacities of all the three alloys increase with supply pressure. Misch metal based alloys demand lower absorption temperatures of below 20°C for storing maximum hydrogen while, Mg₂Ni demands minimum of 300°C for storing maximum storage capacity of about 3.6 wt% (Muthukumar *et al* 2007) at 20 bar supply pressure. The demand of higher overall heat transfer coefficients (for accompanying the rapid heat removal) were necessitated at higher supply pressure. Recently, Marty *et al* (2006) and Phate *et al* (2007) presented a numerical approach for the prediction of heat and mass transfer in a cylindrical hydride reactor during the absorption of hydrogen using a commercial code FLUENT.

In view of the above, it is observed that most of the reported numerical studies have not considered the effects of plateau slope and hysteresis on the pressure-concentration-temperature characteristic curve (PCT) of the metal hydride alloys (Ram Gopal & Srinivasa Murthy (1992; 1993); Guo & Sung 1999; Mat & Kaplan 2001; Dogan *et al* 2004; Faouzi *et al* (2004a; 2004b); Askri *et al* 2003; Marty *et al* 2006; Phate *et al* 2007), which are essential for modelling the dynamic behaviour of absorption and desorption processes. Even though there has been enough literature cited on the heat and mass transfer aspects of hydride bed, still their experimental validations are scarce (Ram Gopal & Srinivasa Murthy (1992; 1993); Askri *et al* 2003; Isselhorst 1995; Ha *et al* 2004; Guo & Sung 1999; Marty *et al* 2006; Phate *et al* 2007). Further, most of the reported studies are concentrated about the predictions of concentration, temperature and pressure profiles variation in the hydride bed and very few parametric studies on hydrogen reactor are reported (Ram Gopal and Srinivasa Murthy (1992, 1995)). Hence, one can conclude that still there is a need of rigorous heat and mass transfer studies in hydride bed considering the effects of plateau slope and hysteresis of PCT, and various operating and hydride bed related parameters on the hydrogen storage capacity. Therefore, the present study aims at developing a two-dimensional mathematical model for cylindrical metal hydride reactor considering the effects of convection in the energy equation and also plateau slope and hysteresis in PCT using a generalized reaction kinetic equation for predicting the influences of various operating parameters and reaction bed thickness on the hydrogen storage performance.

Figure 1. Physical model of hydrogen storage device.

2. Heat and mass transfer analysis

The study has been performed by considering an annular cylindrical metal hydride reaction bed as shown in figure 1. Metal hydride alloy fills the space between the filter (inner wall of hydride bed) and the inner concentric tube of the reactor. Hydrogen is supplied into the bed radially through a porous filter. The heat transfer fluid flows spirally through the space between inner and outer concentric tubes of the reactor.

The following assumptions are made in the heat and mass transfer analysis.

- (i) The medium is in local thermal equilibrium ($T_s = T_g = T$). It means that the conduction in solid and gas phases takes place in parallel and there is no net heat transfer from one phase to another.
- (ii) Effect of radiative heat transfer is negligible. This assumption is valid for all Mm, La, Zr and Ti based alloys whose minimum absorption temperatures are well below 30°C.
- (iii) The gas phase is ideal from thermodynamic point of view.
- (iv) Only mass transfer and no heat transfer takes place through the porous filter.
- (v) The thermal conductivity and specific heat of the hydride bed are assumed to be constant. This assumption underestimates the bed performance slightly, because in actual case the effective thermal conductivity varies with bed pressure and hydrogen concentration.
- (vi) The other thermo-chemical properties such as enthalpy of formation, entropy of formation and activation energy of the metal hydride are independent of temperature and pressure.

The governing equations for heat and mass transfer with chemical reaction in metal hydride bed are described as follows:

2.1 Energy equation

The energy equation in two-dimensional (r and z axis) coordinate is given by

$$\begin{aligned}
 (\rho C_p)_e \frac{\partial T}{\partial t} = & \frac{1}{r} \frac{\partial}{\partial r} \left(r k_e \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left(r k_e \frac{\partial T}{\partial z} \right) - \rho_g^g C_{pg} V_{gr} \frac{\partial T}{\partial r} \\
 & - \rho_g^g C_{pg} V_{gz} \frac{\partial T}{\partial z} - m \left[\frac{\Delta H}{M_g} - T(C_{pg} - C_{ps}) \right].
 \end{aligned} \quad (1)$$

The effective bed thermal conductivity of the bed k_e is expressed as

$$k_e = \varepsilon k_g + (1 - \varepsilon) k_s. \quad (2)$$

The effective heat capacity $(\rho C_p)_e$ is given by

$$(\rho C_p)_e = \varepsilon \rho_g C_{pg} + (1 - \varepsilon) \rho_s C_{ps}. \quad (3)$$

2.2 Momentum equation

The gas velocity can be expressed using Darcy's law. By neglecting the gravitational effect

$$\bar{V}_g = -\frac{\lambda}{\mu_g} \overrightarrow{\text{grad}}(\bar{P}_g^g). \quad (4)$$

2.3 Hydrogen mass balance

$$\varepsilon \frac{\partial \rho_g}{\partial t} + \text{div}(\rho_g V_g) = -m. \quad (5)$$

Assuming hydrogen is ideal gas $\left(\rho_g = \frac{P_g M_g}{RT_g}\right)$ and considering Darcy's law, the mass conservation equation of hydrogen becomes

$$\frac{\varepsilon M_g}{RT_g} \frac{\partial P_g}{\partial t} + \frac{\varepsilon M_g P_g}{R} \frac{\partial}{\partial t} \left(\frac{1}{\partial T} \right) - \frac{\lambda}{v_g r} \frac{\partial}{\partial r} \frac{r \partial P_g}{\partial r} - \frac{\lambda}{v_g} \frac{\partial^2 P_g}{\partial z^2} = -m. \quad (6)$$

2.4 Mass balance for solid phase

During hydriding and dehydriding, the metal hydride alloy cracks into small particles and becomes fine powder. The mass conservation equation of solid phase is given by

$$(1 - \varepsilon) \frac{\partial \rho_s}{\partial t} = m. \quad (7)$$

2.5 Reaction kinetic equation

The hydrogen mass absorbed per unit time and unit volume m , is given by (Mayer *et al* 1987)

$$m = C_a \exp\left(\frac{E_a}{RT_s}\right) \ln\left(\frac{P_g}{P_{eq}}\right) (\rho_{ss} - \rho_s), \quad (8)$$

where C_a is reaction rate constant (s^{-1}) and E_a is activation energy (J/mol H_2).

Number of hydrogen mole absorbed is given by (Ram Gopal and Srinivasa Murthy 1992)

$$N_{H_2} = \frac{1000 \times m_{\text{alloy}} \times N_{\text{alloy}}}{2 \times M_{\text{alloy}}} \frac{\delta X}{\delta t}, \quad (9)$$

where, m_{alloy} is the mass of metal hydride alloy and N_{alloy} is the number of metal atoms per mole of alloy. X is the hydride concentration defined by the ratio number of hydrogen atoms to number of metal atoms per mole of alloy. Hydride equilibrium pressure (P_{eq}) is calculated using the Van't Hoff relation (Nashizaki *et al* 1983).

$$\ln P_{eq} = \frac{\Delta H}{RT} - \frac{\Delta S}{R} + (\phi + \phi_0) \tan \left[\pi \left(\frac{X}{X_m} - \frac{1}{2} \right) \right] + \frac{\beta}{2}, \quad (10)$$

where, ϕ and β are the plateau flatness and plateau hysteresis factors and X_m is the maximum concentration.

2.6 Initial conditions

Initially, bed temperature, gas pressure and hydride density in the reaction bed are assumed to be uniform through out the reactor

$$T_s = T_g = T_0; \quad P_g = P_0; \quad \rho_g = \rho_0. \quad (11)$$

2.7 Boundary conditions

Assuming walls are impervious and adiabatic

$$\frac{\delta P(0, r)}{\delta z} = 0; \quad \frac{\delta T(0, r)}{\delta z} = 0; \quad \frac{\delta P(Z, r)}{\delta z} = 0; \quad \frac{\delta T(Z, r)}{\delta z} = 0. \quad (12)$$

Hydrogen is supplied radially to the hydride bed through a porous filter. Assuming only mass transfer and no heat transfer takes place across porous filter

$$\frac{\delta T(z, r_i)}{\delta r} = 0; \quad P(z, r_i) = P_0. \quad (13)$$

The reaction heat is removed from the hydride bed using a cooling fluid with temperature T_f and assuming walls are impervious

$$-k_e \frac{\delta T(z, r_o)}{\delta r} = U(T - T_f); \quad \frac{\delta P(z, r_o)}{\delta r} = 0. \quad (14)$$

3. Numerical procedure

The resulting system of equations is discretized using the finite volume method. Alternate direction implicit (ADI) scheme with tri-diagonal matrix algorithm (TDMA) is used for solving the set of partial and ordinary differential equations. Velocity terms are handled using staggered grids to catch the heat transfer across the control volume by convection effectively. Boundaries are solved using the half control volume method. A cell independence test has been carried out by analysing the effect of grid number on the average bed temperature with time, which is illustrated in figure 2. A good agreement is observed between 104×104 and 161×161 . Hence, the analysis is carried out using a minimum grid size of 104×104 .

4. Results and discussion

For the validation purpose, a cylindrical reactor of 27 mm internal diameter, 3 mm wall thickness and 450 mm length containing $\text{MmNi}_{4.6}\text{Al}_{0.4}$ (Muthukumar *et al* 2005) is chosen for the present analysis. Table 1 lists the thermo-physical properties of $\text{MmNi}_{4.6}\text{Al}_{0.4}$ and constants used in the modelling. Considering the effect of insertion of copper matrix, an effective thermal conductivity of the hydride is taken as 1.6 W/m-K. Table 2 provides the list of parameters and their ranges used in the analysis.

At the supply condition of 20 bar and 25°C absorption temperature, it is observed from figure 3 that for first few seconds of the reaction, the amount of hydrogen absorbed close to the porous filter (0.5 mm from filter) is more due to high reaction rate accompanied by larger pressure difference between supply pressure and hydride equilibrium pressure ($P_s - P_{eq}$).

Figure 2. Cell independence test.

Table 1. Details of thermo physical properties of $\text{MmNi}_{4.6}\text{Al}_{0.4}$ and constants used in the model.

Properties of $\text{MmNi}_{4.6}\text{Al}_{0.4}$	
Density of metal	8400 kg/m ³
Specific heat of metal (C_{ps})	419 J/kg-K
Effective thermal conductivity of metal (including copper additive) (k_s)	1.6 W/m-K
Porosity (ε)	0.5
Effective density of the hydride at saturation (ρ_{ss})	4259 kg/m ³
Effective density of hydride (ρ_s)	4200 kg/m ³
Activation energy (E_a)	21170 J/mol H ₂
Permeability (λ)	10 ⁻⁸
ΔS	107.2 J/mol H ₂ -K
ΔH	28000 J/mol H ₂
Properties of hydrogen	
Thermal conductivity of hydrogen (k_g)	0.1272 W/m-K
Specific heat hydrogen (C_{pg})	14283 J/kg-K
Density of hydrogen (ρ_g)	0.0838 kg/m ³
Constants used	
Universal gas constant (R)	8.314 J/mol-K
Reaction constant (σ)	75 s ⁻¹
Slope factor (φ_s)	0.35
Constant (φ_o)	0.15
Hysteresis factor (φ)	0.2

Table 2. List of parameters and their operating range.

S. No.	Operating parameter	Range of parameters		
1.	Supply pressure (P_s), bar	10	20	30
2.	Cooling fluid temperature (T_f), °C	15	20	25
3.	Overall heat transfer coefficient (U), W/m ² -K	750	1000	1250
4.	Bed thickness ($r_o - r_i$), mm	7.5	12.5	17.5

Note: 1. Effective thermal conductivity of the bed is fixed at 1.6 W/m-K

2. Numbers in italic bold show the values around which other parameters are varied.

Later, due to rapid heat generation close to the porous filter, the rate of reaction drops significantly (due to fall in pressure difference ($P_s - P_{eq}$)) and becomes negligible till the hydride equilibrium pressure falls below a certain value. However, the region close to the convection boundary starts to absorb hydrogen at relatively faster rate and reaches the saturation state much before the net absorption comes to an end. Due to the effective heat transfer from the hydride bed to the cooling fluid, the rise in bed temperature close to the convection boundary region is lower, resulting in larger driving potential ($P_s - P_{eq}$) for hydrogen absorption.

Figures 4 and 5 together reveal that the low temperature regions in the hydride bed ensure high rate of absorption and vice-versa. It is observed from figure 5 that in the beginning of the absorption process the bed temperature increases sharply, reaches its maximum and then decreases gradually, and becomes equal to the cooling fluid temperature at the end of the absorption process. Due to poor thermal conductivity of the hydride bed, the bed is not able to transfer the complete heat of absorption during the initial rapid reaction. Hence, the excess heat is stored in the bed itself, resulting in sudden rise in bed temperature. Later, the bed temperature decreases due to fall in the reaction rate and increase in heat transfer from the bed to the cooling fluid.

Figure 3. Variation of hydride concentration with time for first 40 s.

Figure 4. Variation of hydride concentration with time.

It is observed from figure 6 that the equilibrium pressure increases sharply at the initial stage of absorption due to the rise in bed temperature. Later, due to heat transfer to next layers, the equilibrium pressure of the region close to filter ($r = 0.5 \text{ mm}$) decreases gradually after attaining its maximum value till about 150 s. After 150 s, due to formation of hydride the equilibrium pressure is again found to increase and becomes equal to the supply pressure at the end of the absorption process. While the regions closer to the convective boundary experience continuous rise in the equilibrium pressure due to the formation of hydride. It is

Figure 5. Variation of bed temperature with time.

Figure 6. Variation of equilibrium pressure with time.

observed that the absorption of hydrogen continues till the existence of pressure difference between supply pressure and hydride equilibrium pressure ($P_s - P_{eq}$). It is clear from figures 4 to 6 that the regions closer to the convective boundary attain the saturation stage much before the estimated absorption time due to effective heat transfer from hydride bed to the cooling bed. Hence, one can conclude that for accompanying faster absorption rate, the rise in bed temperature should be minimized by improving the heat removal rate from the hydride bed.

4.1 Effect of supply pressure

An attempt was made for validating the present numerical results with the experimental data of similar reactor geometry under same operating conditions. Figure 7 shows that the numerical

Figure 7. Effect of supply pressure on hydrogen storage capacity.

Figure 8. Effect of cooling fluid temperature on average bed temperature.

results are in good agreement with the experimental data reported by Muthukumar *et al* (2005). It is also observed from figure 7 that the maximum storage capacity of about 1.3 wt % is obtained at the supply conditions of 30 bar and 298 K. The effect of supply pressure on the hydrogen storage capacity is more predominant for the supply pressures of above 10 bar. This is due to the large slope of the PCT characteristic of the alloy; higher supply pressures increase the storage capacity significantly.

4.2 Effect of cooling fluid/absorption temperature

Figures 8 and 9 together reveal that at lower cooling fluid temperatures, the hydrogen absorption proceeds at a faster rate due to the availability of larger driving potential for mass transfer. At low absorption temperature, the equilibrium pressure (P_{eq}) which is the function of bed temperature is lower, resulting in larger pressure difference ($P_s - P_{eq}$). In addition, at lower absorption temperatures, the temperature difference ($T - T_f$) is also higher, leading to a faster heat removal during the hydriding reaction. Hence, at lower absorption temperatures the hydride absorbs more hydrogen with shorter reaction time. For a given supply pressure, hydrogen storage capacity is also found to increase significantly at lower absorption temperature due to prevailing lesser plateau slope.

4.3 Effect of overall heat transfer coefficient

By definition, the overall heat transfer resistance ($1/U$) combines the resistances offered by the reactor wall and the heat transfer fluid, while the contact resistance between the hydride particle and wall is negligible. Different overall heat transfer coefficients are obtained by varying the heat transfer fluid flow rates. Figures 10 and 11 illustrate that the increase of overall heat transfer coefficients up to about $1000 \text{ W/m}^2 - \text{K}$ accelerate the reaction by accompanying fast heat removal rate and beyond which either supply pressure or bed effective thermal conductivity controls the reaction rate.

Figure 9. Effect of cooling fluid temperature on hydrogen storage capacity.

4.4 Effect of hydride bed thickness

Effect of hydride bed thicknesses on average bed temperature and hydrogen storage capacity are illustrated in figures 12 and 13. Different bed thicknesses are obtained by keeping the filter radius and volume of the reactor as constant and by varying the outer radius r_o . It is observed that the higher bed thicknesses offer larger resistance to heat transfer resulting in slower reaction and large cycle time. For better heat and mass transfer characteristics, hydride bed thickness should be kept as minimum (below 10 mm).

Figure 10. Effect of overall heat transfer coefficient on average bed temperature.

Figure 11. Effect of overall heat transfer coefficient on hydrogen storage capacity.

4.5 *Effect of convection terms*

Figure 14 shows the variation of bed temperature at different radial position without considering the effect of convection terms in the energy equation. It is observed that figure 14 almost resembles with figure 5, revealing that the effect of convective term on the net heat transfer is negligible. However, there is a slight deviation of temperature profile of the layer closer to the porous filter ($r = 0.5$ mm) for first 20 s. This can be more clearly explained from the variation of hydrogen velocity across the bed. It is clear from Fig. 15 that the gas velocity has

Figure 12. Effect of bed thickness on average bed temperature.

Figure 13. Effect of bed thickness on hydrogen storage capacity.

the significant value only for the initial period (up to 20 s) and becomes negligible during rest of the absorption process. Hence, one can conclude that the effect of convection is significant only during the initial stage of absorption and later, the hydrogen transfer occurs mainly by diffusion. Figure 16 further reveals that the convection effect does not have any significant contribution to the rate of hydrogen absorption.

Figure 14. Variation of bed temperature with time (without convection effect).

Figure 15. Variation of radial velocity of H_2 with time.

5. Conclusions

Two-dimensional numerical model dealing with the coupled heat and mass transfer processes in a cylindrical hydride bed is developed using the finite volume method, considering the plateau slope and hysteresis effects of PCT of the metal hydride alloy. The effect of the convection terms in the energy equation is found to be negligible. At the start of the absorption process, rapid hydrogen absorption close to the filter is observed. The analysis reveals that

Figure 16. Effect of convection terms on hydrogen storage capacity.

the hydrogen absorption is a function of heat transfer rate and driving force ($P_s - P_{eq}$). A maximum storage capacity of about 1.3 wt% is achieved at supply pressure of 30 bar and 298 K. Increase of overall heat transfer coefficient up to about 1000 W/m² – K reduce the reaction time significantly. Thin beds are found to be better both in terms of higher absorption rate and good heat transfer characteristics.

List of symbols

C_p	Specific heat capacity, J/kg-K
k	Thermal conductivity, W/m-K
m	Hydrogen mass absorbed, kg/m ³ -s
m_{alloy}	Mass of alloy, kg
M	Molecular weight, kg/kmole
N_{H_2}	Number of hydrogen moles
N_{alloy}	Number of metal atoms per mole of alloy
P	Pressure, N/m ²
r	Radius, m
R	Universal gas constant, J/mole-K
T	Temperature, K or °C
t	Time, s
U	Overall heat transfer coefficient, W/m ² -K
V	Velocity of gas, m/s
X	Concentration, H/M ratio
z	Axial length, m

Greek letter

β	Plateau hysteresis factor
ε	Porosity
λ	Permeability, m ²
μ	Dynamic viscosity, kg/m-s
ν	Kinematic viscosity, m ² /s
ρ	Density, kg/m ³
ϕ, ϕ_0	Plateau flatness factors
ΔH	Enthalpy of absorption, J/mol H ₂
ΔS	Entropy of formation, J/mol H ₂ -K

Subscripts

e	Effective
eq	Equilibrium
f	Cooling fluid
g	Gas
i	Inner
m	Maximum
o	Outer
s	Solid, supply
ss	Saturated
0	Initial

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