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Analysis of Multi-Corrugated Tube Metal Hydride Based Hydrogen Storage System

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Abstract - *This paper presents simulation and analysis of three-dimensional, transient, coupled heat and mass transfer process in a multi-corrugated reactor filled with metal hydride $MmNi_{6.4}Al_{0.4}$ during hydrogen absorption. The model is validated, the average bed temperature of cylindrical reactor at 30 bar supply pressure showed good agreement with the experimental data reported in the literature. Hydrogen storage performance of the system is numerically analysed by implementation of corrugation, varying the tube pitch layout and operating parameters such as hydrogen supply pressure in the system. Further, the effects of supply hydrogen pressure and cooling fluid temperature are also presented.*

Keywords - *Hydrogen storage reactor, Multi tube corrugated Metal hydride: $MmNi_{6.4}Al_{0.4}$, Coupled heat and mass transfer, CFD*

I. INTRODUCTION

The world requires a lot of energy to produce goods and services. The earth is a very complex and vulnerable system. Our historical development has affected on the fragile natural balance. Now days, the energy consumption is totally based on the limited reserves of fossil fuels like oil, natural gas and coal. Consumption increased for all fuels, reaching record levels for every fuel type except nuclear power; production increased for all fuels except coal. For oil and natural gas, global consumption growth was weaker than production. If the production rate continues at today's level, the estimated reserves of oil in the entire world will end in 52.5 years while the reserves of natural gas will end in 54 years and the reserves of coal will end in 110 years. The consumption of fossil fuels also leads to environmental consequences such as emissions of the climatic gas carbon dioxide (CO_2) and nitrous gases (NO_X). This is an increasing pollution problem in cities and villages and a hazard to health.

Consequently there are two main reasons to investigate other energy carriers than fossil fuels:
1. Limited reserves of fossil fuels on earth.
2. Environmental problems due to consumption of fossil fuels.

The government and environmental protective agency's (EPA) are promoting for new alternative and renewable energy technologies concerning about these issues. Due to decreasing production rate and increasing demand of fossil fuels the renewable energy technologies with solar, biomass, wind energy or nuclear energy sources can be proposed. However, as most of these energy resources are intermittent and, a way of storing energy is required. Also, energy sources like hydrogen, CO_2 are non-polluting or less-polluting fuels are under developing conditions. Therefore, hydrogen energy has much potential to become as an alternative energy to conventional resources. Many researchers have investigated heat and mass transfer studies for different metal hydrides and geometries of reactors [1-11]. This paper focuses on investigation of corrugations provided to the reactor tube.

II. PHYSICAL MODEL OF THE REACTOR

The configuration of a corrugated cylindrical metal hydride reactors in the corrugated tubes are holding by baffles mounted in the shell, is shown in Fig.1. The numerical analysis is carried out using the thermo-physical properties of alloy, hydrogen, and constants used.[12]The inner tubes of the reactors are porous filter. The porous filter works in two ways; it serves as an effective hydrogen passage and distributor, and it prevents the escape of metal hydride particles along with hydrogen gas during desorption. The annular space between porous filter and inner concentric tube is filled with hydriding alloy (MmNi6.4Al0.4). During absorption process, hydrogen is supplied to the reactor through the porous filter. Hydride bed absorbs hydrogen by releasing heat to the cooling fluid which is passing between inner and outer concentric tubes. The hydriding mechanism is assumed to be axisymmetric, i.e. one fourth of the reactor is considered for the analysis in an advanced CFD tool FLUIDNEXUS 1.6.0 with solver FLUENT 16.2.

To simplify the calculations for numerical simulations of hydrogen storage system the following assumptions are made.[13]
1) Heat transfer through the hydride bed is by conduction only
2) The hydrogen gas in the bed is in local thermal equilibrium with the hydride particles
3) The thermo-physical properties of the metal hydride and hydrogen are constant

4) There is no heat transfer between the hydride reactor and the surroundings
5) There is no heat transfer through the porous filter
6) There is no heat transfer between the bed and hydrogen at the inner radius of the inner tube

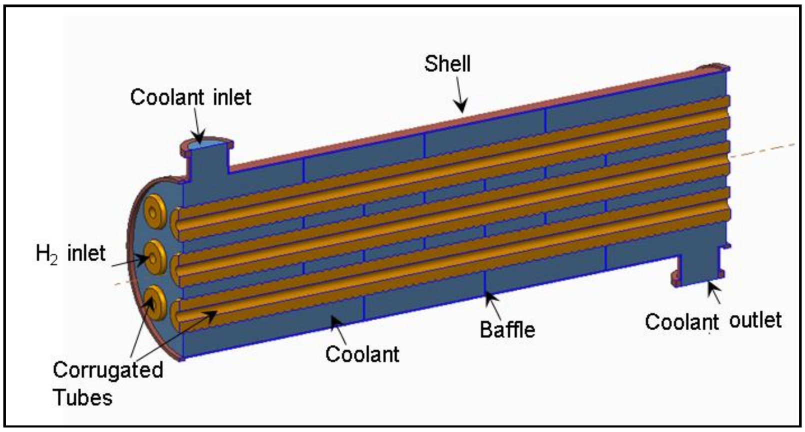

Figure 1.Schematic of Corrugated tubes hydrogen storage reactor.

III. MATHEMATICAL FORMULATION OF HYDROGEN STORAGE SYSTEM

3.1. Problem Formulation

The coupled heat and mass transfer, hydrogen flow and the absorption in storage system are modelled by using. A transport equation in the general form is given by [13]:

$$\frac{\partial \rho \emptyset}{\partial t} + \nabla \cdot (\rho \vec{V} \emptyset) = \nabla \cdot (\Gamma \nabla \emptyset) + S_h \qquad (1)$$

The energy equation for the hydride bed in two-dimensional cylindrical coordinate system is given by:

$$(\rho C_p)_e \frac{\partial T}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\lambda_e \frac{\partial T}{\partial r}\right) + \frac{\partial}{\partial z}\left(\lambda_e \frac{\partial T}{\partial z}\right) + \frac{1}{\Theta}\frac{\partial}{\partial k}\left(\Theta\lambda_e \frac{\partial T}{\partial \Theta}\right) - (\rho C_{pg} V_{gr})\frac{\partial T}{\partial r}(\rho C_{pg} V_{gz})\frac{\partial T}{\partial z} - (\rho C_{pg} V_{gk})\frac{\partial T}{\partial \Theta} - \rho(1-\varepsilon)\frac{\Delta H}{M_{H_2}}\frac{\partial x}{\partial t} \qquad (2)$$

The effective heat capacity $(\rho C_p)e$ of the hydride bed is given as [13]:

$$(\rho C_p)_e = \varepsilon(\rho C_p)_g + (1-\varepsilon)(\rho C_p)_m \qquad (3)$$

The effective thermal conductivity of bed given as [2]:

$$\lambda_e = \varepsilon\lambda_g + (1-\varepsilon)\lambda_m \qquad (4)$$

The density of metal hydride bed is given by [2]:

$$\rho_{mb} = \rho_m(1-\varepsilon) \qquad (5)$$

The last term of the right-hand side of the energy equation (Eq.(2)) represents the heat release during absorption (W/m3), which is a function of hydride bed temperature, hydride equilibrium and hydrogen supply pressures, and hydrogen concentration. The generalized reaction kinetic equation (used for wide range of hydrogen absorbing alloys) is given by [13]:

$$\frac{\partial x}{\partial t} = \sigma \frac{(P_g - P_{eq})}{P_{eq}} \frac{(x - x_f)}{(x_i - x_f)} \exp\left(\frac{-E_a}{R_u T}\right) \qquad (6)$$

Where, ‘x’ is the hydrogen concentration defined as the quantity obtained by dividing the number of hydrogen atoms absorbed by number of metal atoms per mole of alloy (H/M).

The equilibrium pressure (P_{eq}) is estimated by using Van’t Hoff equation [13]

$$P_{eq} = \exp\left[\frac{\Delta S}{R_u} - \frac{\Delta H}{R_u T} + (\psi_s + \psi_o) \times \tan\left(\pi\left(\frac{x}{x_f}\right) + \frac{\psi}{2}\right)\right] 10^5 (7)$$

Where, P_{eq} is in N/m^2.

The continuity equation includes a sink term to take the amount of hydrogen absorption into account and the resulting equation per unit volume (kg/m^3 s) is given by [13]:

$$\varepsilon\frac{\partial \rho_g}{\partial t} + \nabla\cdot\left(\rho_g \vec{V}\right) = \Delta\rho(1-\varepsilon)\frac{\partial x}{\partial t} \qquad (8)$$

Where, the gas density ρ_g is taken from ideal gas equation

$$(\rho_g = P_g M_{H_2}/R_u T) \quad \text{and} \Delta\rho = \rho_{ss} - \rho_s \qquad (9)$$

Where, ρ_{ss} is the hydride density at the end of the absorption process and ρ_s is the hydride density at any given time 't'. The right hand side term of Eq. (8) represents the hydrogen consumption during the absorption.

3.2. Initial and Boundary Condition

Initially (t = 0), the bed temperature, hydride equilibrium pressure and hydride density are assumed to be uniform throughout the reactor.[2]

$$\rho_m(z, r, \Theta, 0) = \rho_0;$$

$$T_m(z, r, \Theta, 0) = T_g(z, r, \Theta, 0) = T_0;$$

$$P_g(z, r, \Theta, 0) = P_0; \qquad (10)$$

Along the porous wall:

$$\frac{\partial T}{\partial t}(z, r, 0, t) = 0; \text{and} \frac{\partial T}{\partial t}(0, r, 90^o, t) = 0; \quad (11)$$

Along the left face, where z = 0, ϴ = 0 and z = 0, ϴ = 90° adiabatic and impervious conditions [13]:

$$\frac{\partial T}{\partial z}(0, r, 0, t) = 0; \text{and} \frac{\partial T}{\partial z}(0{,}0, 90^o, t) = 0; \quad (12)$$

Along the right face, where z = Z, ϴ = 0 and z = Z, ϴ = 90° adiabatic and impervious conditions give.[13]

Along the top boundary, heat released by the bed during absorption of hydrogen is transferred to the cooling fluid. Here the entire bed is assumed to be a heat exchanger system as shown in Fig.1.

The above equation allows two possibilities for analysis:

1. The evaluation of the performance of the hydride bed with established design of heat exchanger.
2. The optimal design of heat exchanger by analyzing the influence of different heat exchanger parameters such as overall heat transfer coefficient and heat transfer area on performance of the hydride bed.

In the present work, the design parameters of the heat exchanger such as mass flow rate of the cooling fluid, over all heat transfer coefficient are fixed and varying the convection heat transfer area, the performance of the hydrogen storage device is evaluated.

3.3. Method of solution

A CFD analysis of coupled heat and mass transfer in corrugated metal hydride beds is carried out using commercial solver FLUENT 16.2. The present analysis is carried out by hooking a user-define function to FLUENT 16.2. Heat source term is solved, the rate of reaction i.e. absorption rate of hydrogen concentration is modelled by using scalar equation and the pressure equilibrium equation are hooked. The user-define function is compiled in the FLUENT 16.2.

IV. NUMERICAL SIMULATION OF CORRUGATED TUBEHYDROGEN STORAGE SYSTEM

4.1. Validation of numerical results

The numerical model is validated using a cylindrical reactor having 27mm internal diameter with 3mm wall thickness and 450mm length. Thermo-physical properties of the hydriding alloy and various constants used in the mathematical modelling are listed in Table 1.[13]

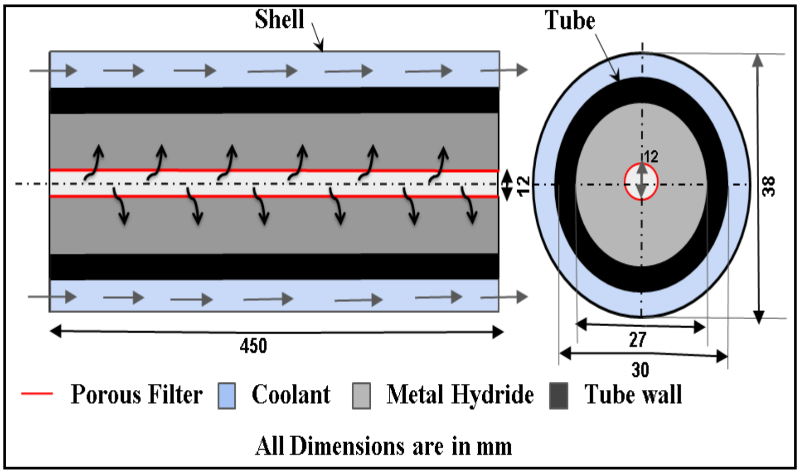

Figure 2.Schematic of Co-axial cylindrical reactor.

Table 1.Thermo-physical properties of the hydriding alloy and various constants

	Values
Properties of mNi$_{6.4}$Al$_{0.4}$	
Density of Metal	8400 kg/m^3
Specific heat of Metal	419 J/kgK
Effective thermal conductivity of MH bed	1.6 W/mK
Porosity	0.5
Effective Density of Saturation	4259 kg/ m^3
Effective Density of solid	4200 kg/m^3
Hydride activation energy	21,170 J/mol H_2
Entropy of Reaction	107.2 J/mol H_2 K
Enthalpy of Reaction	28,000 J/mol H_2
Properties of Hydrogen	
Thermal-conductivity Hydrogen	0.127 W/mK
Specific Heat of hydrogen	14,283 J/kgK
Density of Hydrogen	0.0838 kg/m^3
Molecularwt of Hydrogen	2.016 kg/mol
Constant used	
Universal Gas constant (R_u)	8.314 J/mol K
Reaction Constant (σ)	0.35
Slope Factor (ψ_s)	0.15
Constant (ψ_o)	0.2

Fig.2. Showed that the numerically predicted average bed temperature profile at 30 bar supply pressures are in good agreement with the experimental results reported. [12] The discrepancy in numerical prediction may be due to the

assumption of large void fraction (ε = 0.5) and local thermal equilibrium in the analysis. While, in the experimental studies thermal mass (copper matrix) was used in the void space for improving the effective thermal conductivity of the hydride bed.

4.2. Comparison of numerical results

The hydrogen storage system is modified by applying corrugated tubes instead of smooth tubes in shell and tube heat exchangers shown in Fig.4. Corrugated tubes create extra turbulence in the fluid that flows through the tubes. This extra turbulence translates itself in a large increase in heat transfer compared to smooth tube heat exchangers.

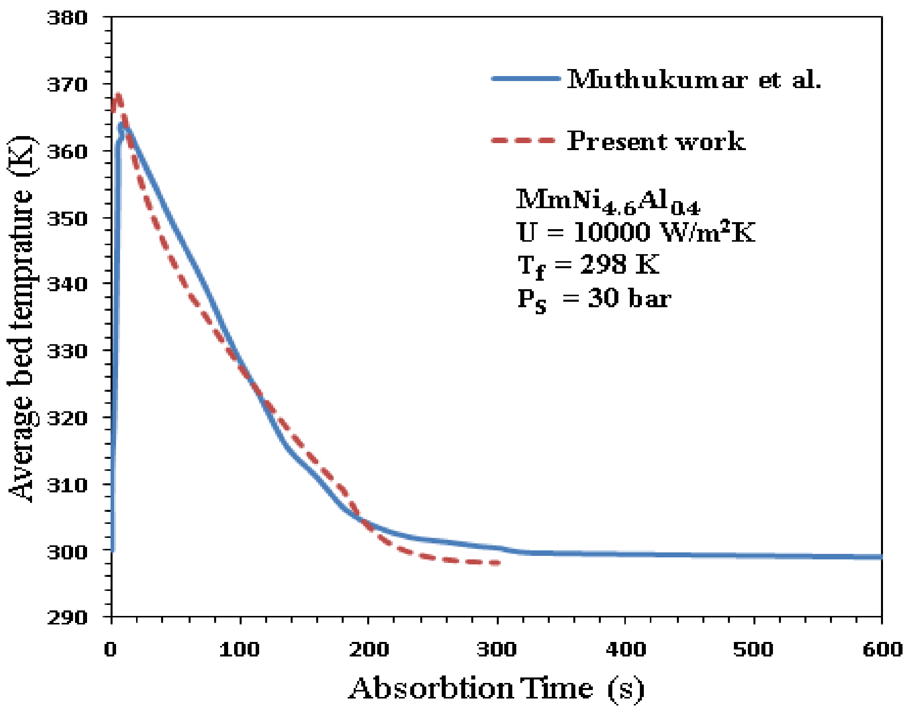

Figure 3.Validation of predicted average bed temperature profiles.

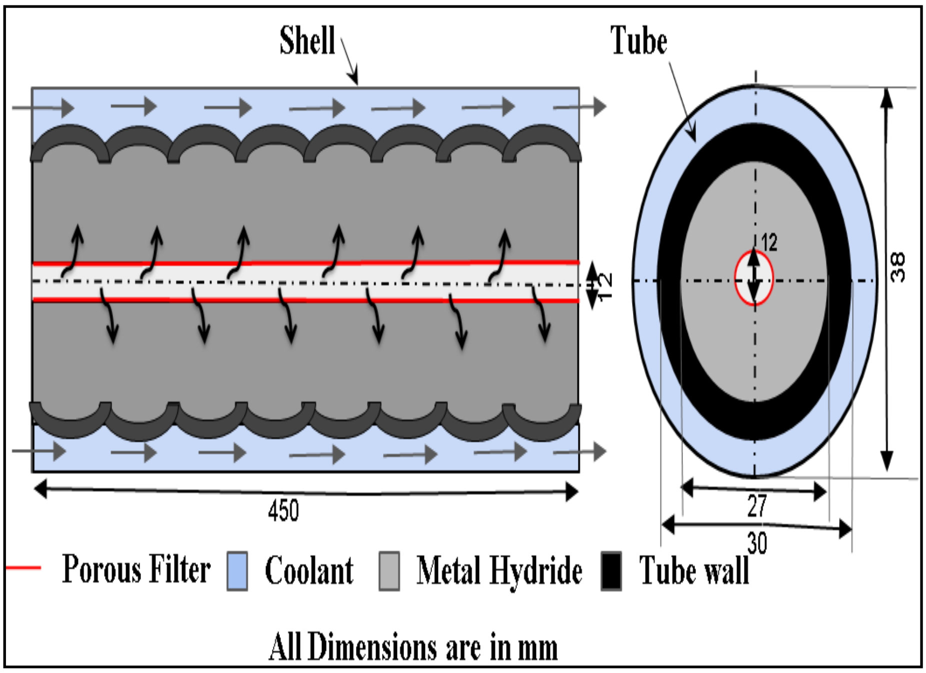

Figure 4.Schematic of corrugated tube hydrogen storage system.

Fig. 5 shows that the three-dimension numerically predicted average bed temperature profile at supply pressure of 30 bar in corrugated tube system are better than plain tube system. Because of the geometry modelling surface area of the corrugated tube increases by 5.68 %.

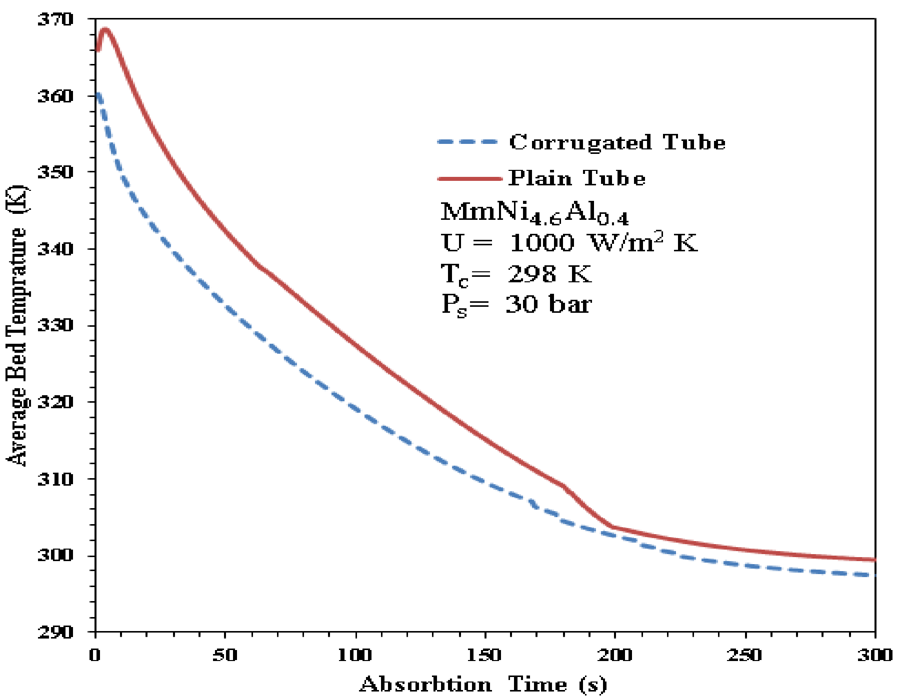

Figure 5.Predicted average bed temperature profiles of Corrugated tube HSS

The cooling rate of corrugated tube hydrogen storage system is faster than the plain tube hydrogen storage system due to increasing surface area of convection boundary.

4.3. Simulation of multi corrugated tube metal hydride based hydrogen storage system

In the multi-corrugated tubes metal hydride based hydrogen storage system design, employs a cylindrical shell with a number of corrugated parallel tubes inserted inside the shell shown in Fig. 6. Metal hydride is present in the tubes and the coolant flows through the shell. In order to increase the heat and mass transfer from the fluid, baffles are mounting in the shell side. These types of designs presented in reference. The number of tubes chosen is based on the consideration of weight of metal hydride in the bed. The optimization in this study involves only for the most important geometric parameters for corrugated tube. However, further optimization of the design is possible.

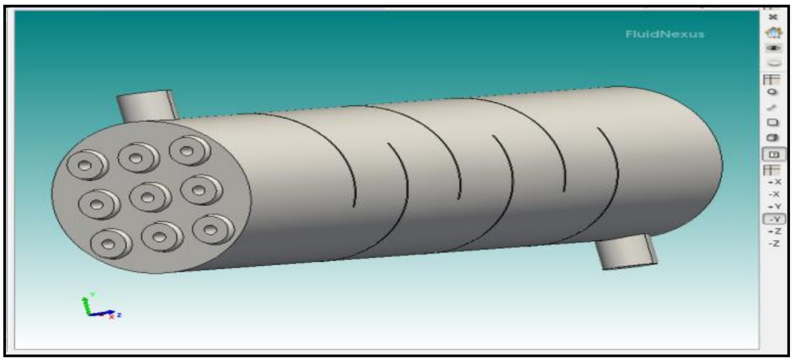

Figure 6.Simulation Model of Multi-Corrugated tube hydrogen storage system

Table 2.Design parameters of multi corrugated tube metal hydride based hydrogen storage system

Parameter	Value
Outer diameter of tube, d_o	0.02676 m
Inner diameter of tube, d_i	0.02576 m
Outer diameter of shell, D_S	0.013 m
Length of shell, L	0.45333 m
Number of tube counts, N_t	9

The present analysis is carried out for a multi-corrugated tube metal hydride based hydrogen storage system with 9 corrugated tubes having maximum thermal performance factor of 2.33 is taken for the enhanced tube with pitch ratio, P/D_H=0.27 and rib-height ratio, e/D_H=0.06 are arranged in square pitch layout (i.e. 90 degree) having diameter 25.76 mm with 1 mm wall thickness and 450.33 mm length. Thermo-physical properties of the hydriding alloy and various constant used in mathematical modelling are listed in Table 3. The mesh independent test is carried out for analysing the effect of mesh size on the average bed temperature with absorption time. As illustrated in Fig. 7 good agreement is observed between mesh size 1.5 and 3.0 million. Hence, the present analysisis carried out using a minimum mesh size 1.5 million.

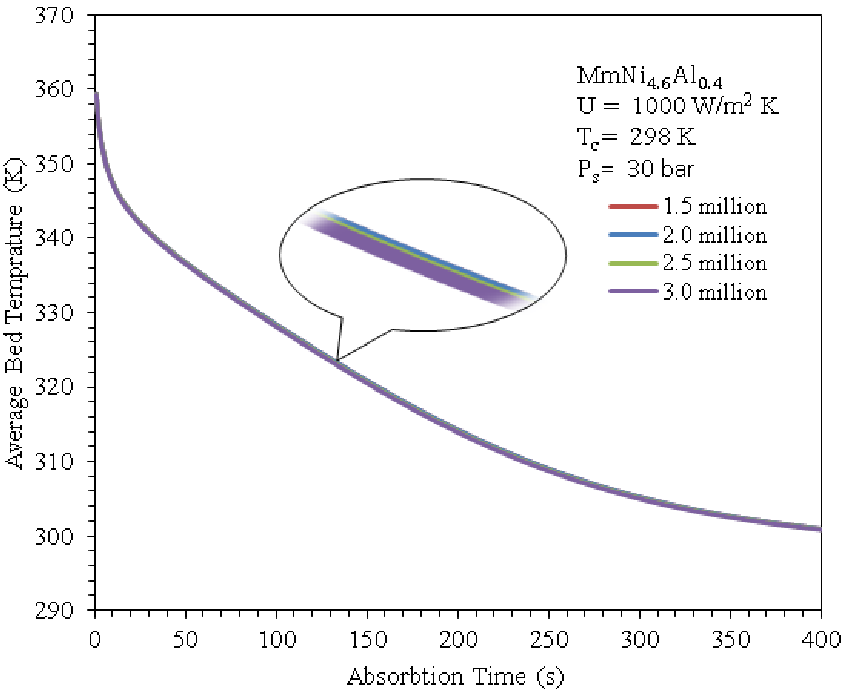

Figure 7.Mesh independent test

Table 2.Design parameters of multi corrugated tube metal hydride based hydrogen storage system

Bed specifications	**Values**	**Values**	**Values**	**Units**
Tube Pitch layout	45°	60°	90°	
Number of beds	9	9	9	
Deliverable hydrogen	0.093	0.093	0.093	kg
Length of tubes	0.45333	0.4533	0.45333	m
Diameter of tube (inner)	0.02576	0.0257	0.02576	m
Diameter of tube (outer)	0.02676	0.0267	0.02676	m
Diameter of bed (inner)	0.008	0.008	0.008	m
Storage capacity of the metal hydride	1.3	1.3	1.3	wt%
Metal hydride per tube	0.7931	0.7931	0.7931	kg
Number of tubes	9	9	9	
Total mass of metal hydride	7.14	7.14	7.14	kg
Total tube volume	1.785428	1.785	1.785428	L
Total tube mass	1.26441	1.2644	1.26441	kg
Mass of end plates, baffles and caps	0.57699	0.8255	0.57699	kg
Shell mass	0.53722	0.6411	0.53722	kg
Cooling fluid mass	3.77814	6.2303	3.77814	Kg
Total system volume	5.973396	8.5552	5.973396	L
Total system mass	9.38	9.8712	9.38	Kg

Fig. 8 shows that the 3D numerically predicted average bed temperature profile at supply pressure of 30 bar for different pitch layout arrangement of multi-corrugated tubes metal hydride based hydrogen storage system. The square and rotated square pitch layout shows better performance than the triangular pitch layout in the multi-corrugatedtube metal hydride based hydrogen storage system. The hydrogen absorption rate of multi-corrugated tubes metal hydride based hydrogen storage system is near about 400s.

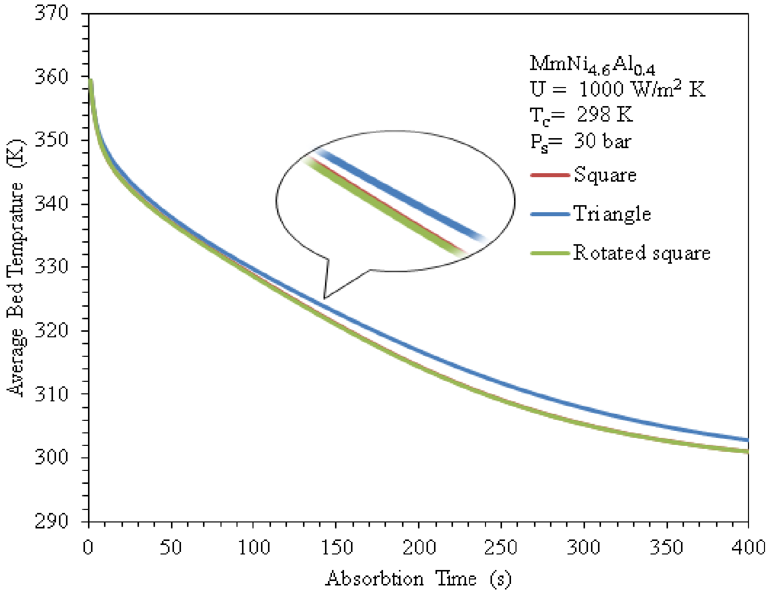

Figure 8.Predicted average bed temperature profiles of multi-corrugated tubes metal hydride based hydrogen storage.

4.4. Effect of supply pressure on average bed temperature and hydrogen concentration during absorption

Fig.5.21 and 5.22 shows the effect of hydrogen supply pressure on average bed temperature and hydrogen concentration of multi-corrugated tubes metal hydride based hydrogen storage respectively. It is clear from the reaction kinetic equation Eq. (3.6) that the difference between hydrogen supply and hydride equilibrium pressure $(P_s - P_{eq})$ is worked as driving force. Initially, the pressure difference is very large resulting in rapid exothermic reaction and hence, high amount of heat is released to the metal hydride bed. Due to poor thermal conductivity of the metal hydride bed, heat is not transferred completely to the cooling fluid hence itself resulting in sudden rise in metal hydride bed temperature. As a result the increasing equilibrium pressure, resulting in decreasing driving force and corresponding to fall in hydrogen absorption rate. As the time progresses, the average bed temperature of metal hydride bed decreases gradually due to decreasing reaction rate and heat transfer from the metal hydride bed to cooling fluid, causing to promote further hydrogen absorption. Hydrogen absorption continues till the hydride equilibrium pressure equals to hydrogen supply pressure. It is clear from Fig. 5.21 that increasing metal hydride bed temperature is higher for higher supply pressures due to faster reaction rate.

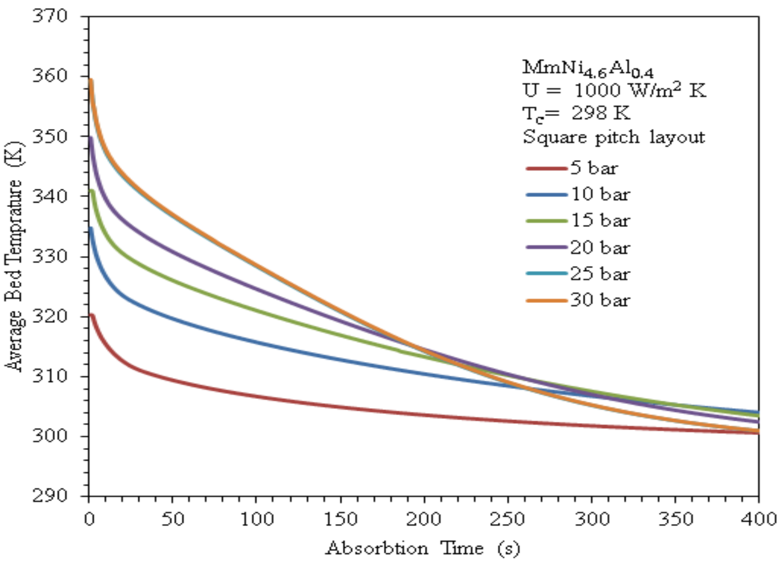

Figure 5.21.Effect of supply pressure on average bed temperature during absorption

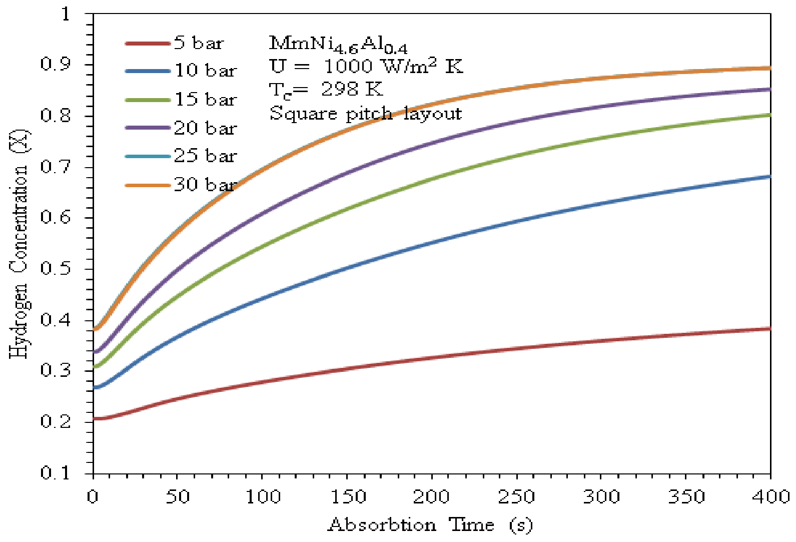

Figure 5.22.Effect of supply pressure on hydrogen concentration during absorption

4.5. Effect of cooling fluid temperature on average bed temperature and hydrogen concentration during absorption

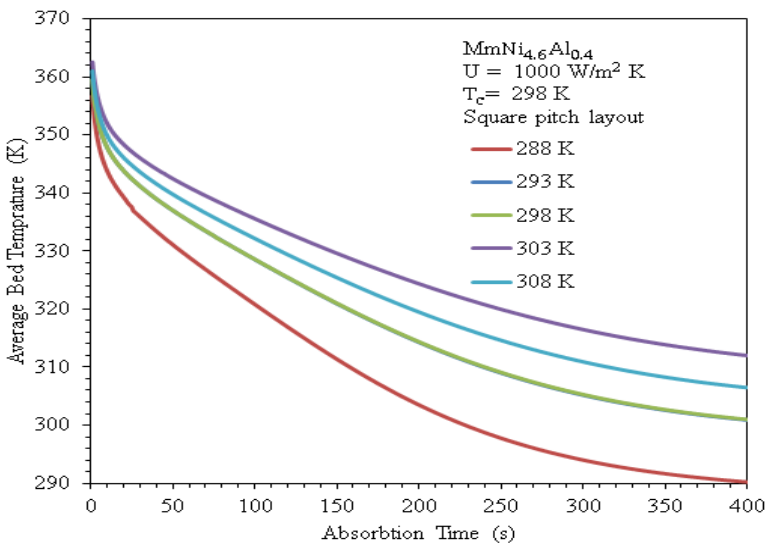

Figure 5.23.Effect of cooling fluid temperature on average bed temperature during absorption

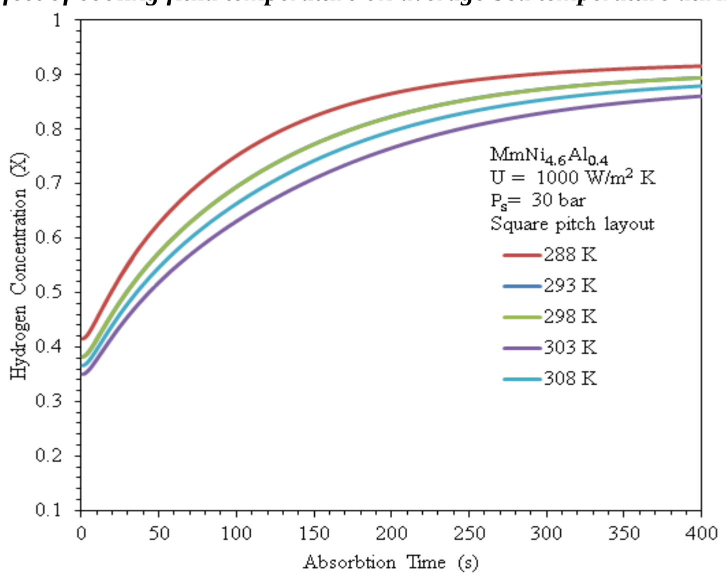

Figure 5.24. Effect of cooling fluid temperature on hydrogen concentration during absorption

Fig. 5.22 and 5.23 shows the effect of cooling fluid temperature on average bed temperature and hydrogen concentration in the of multi-corrugated tube metal hydride based hydrogen storage system respectively. Figure show that at cooling fluid temperature 288 K absorption time is lowest.

4.6. Contours of average bed temperature

Fig.5.22a to 5.22h shows contours of average bed temperature during absorption process at different time for square pitch layout in multi-corrugated tubes metal hydride based hydrogen storage system at 30 bar supply pressure and 298 K cooling fluid temperature upto 300 sec. Initially, the pressure difference is very large resulting in rapid exothermic reaction (absorption) and hence, high amount of heat is released to the metal hydride bed. This heat is taken out from the system through cooling fluid gradually.

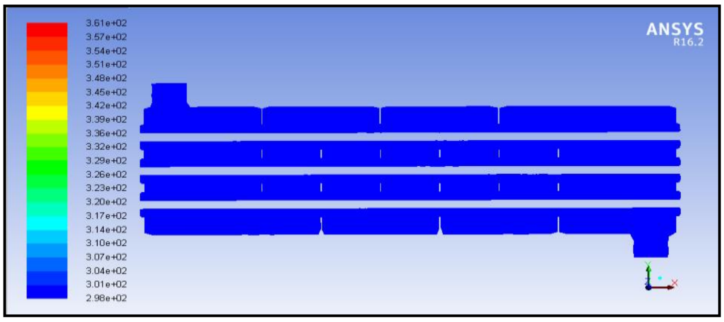

Figure 5.25a. Contours of average bed temperature during absorption at 0 sec

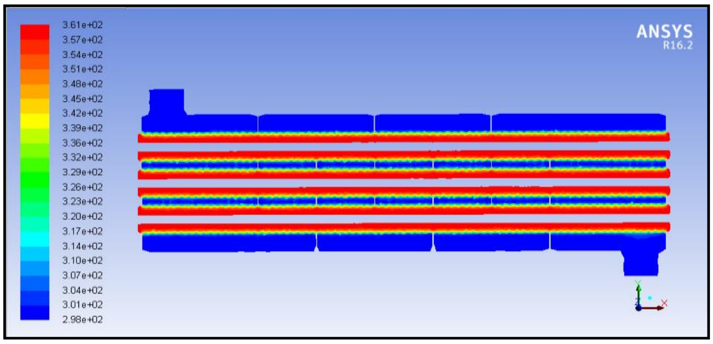

Figure 5.25b. Contours of average bed temperature during absorption at 2 sec

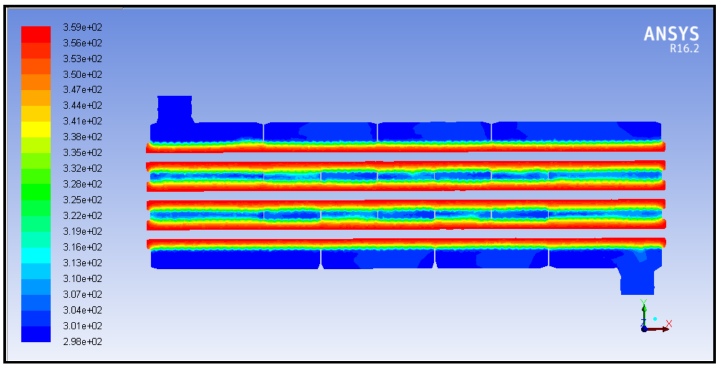

Figure 5.25c. Contours of average bed temperature during absorption at 12 sec

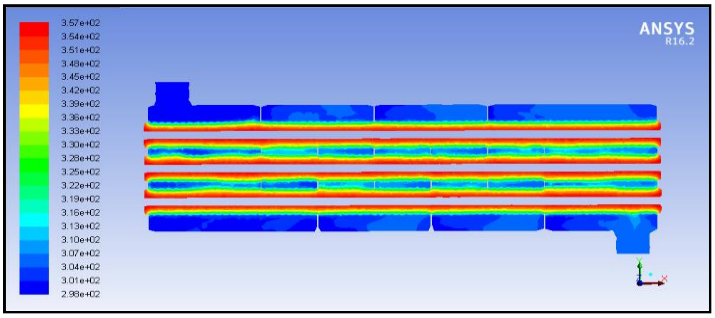

Figure 5.25d. Contours of average bed temperature during absorption at 25 sec

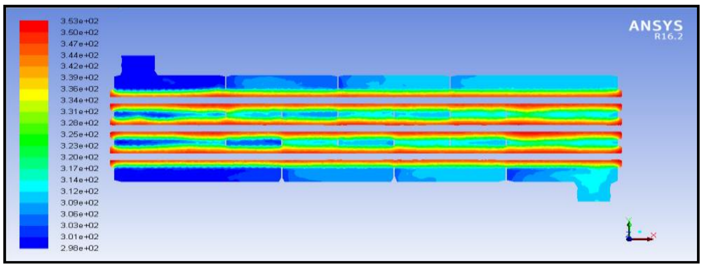

Figure 5.25e. Contours of average bed temperature during absorption at 50 sec

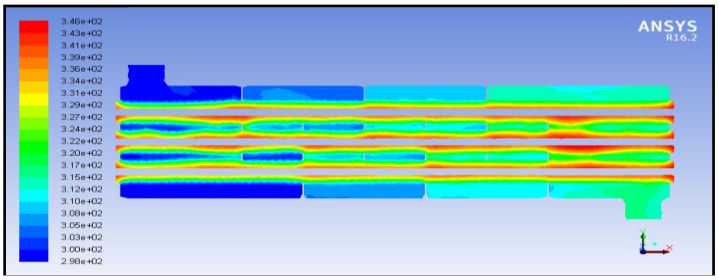

Figure 5.25f.Contours of average bed temperature during absorption at 100 sec

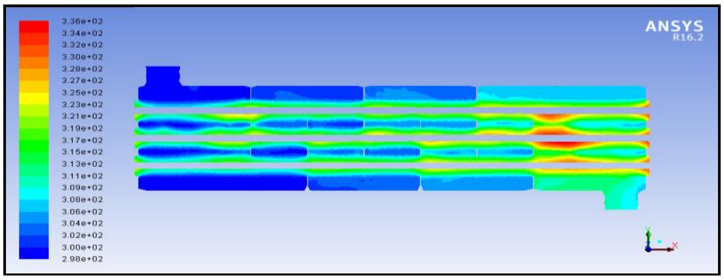

Figure 5.25g. Contours of average bed temperature during absorption at 200 sec

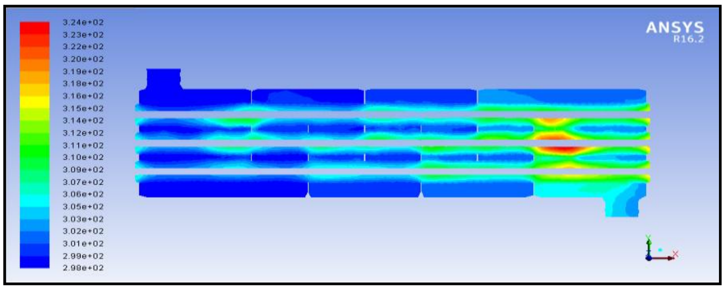

Figure 5.25h. Contours of average bed temperature during absorption at 300 sec

V. CONCLUSIONS

In this paper simulation of 3D coupled heat and mass transfer analysis of $MmNi_{4.6}Al_{0.4}$ based multi corrugated tube hydrogen storage system are investigated for different supply pressure of hydrogen gas with the help of FLUENT 16.2. The predicted average bed temperature and hydrogen concentration profiles at different supply pressure of hydrogen gas are reported. In order to increasing performance of the system the heat and mass transfer rate of the system have to be improved. It has been shown that the Corrugated Tube is better than Plain Tube hydrogen storage System because of the Surface Area of the corrugated tube increases by 5.68% due to geometry modelling resulting increases heat and mass transfer of the system. However, it should be noted that only the most important geometric parameters are considered for the corrugated tube hydrogen storage system and further optimisation of the design is possible.

The three different pitch layout of multi-corrugated tubes metal hydride based hydrogen storage systems are designed for the on-board applications. The performance of these systems are numerically investigated and square pitch layout arrangement of tubes in the hydrogen storage system shows better performance. The effect of supply pressure and cooling fluid temperature on absorption process is carried on square pitch layout of tubes in the hydrogen storage system. It is observed that for 30 bar supply pressure and 298 K cooling fluid temperature shows better performance.

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