

PERFORMANCE OF AIR COOLED HYDROGEN STORAGE DEVICE WITH EXTERNAL FINS

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ABSTRACT: A parametric study of an air-cooled annular cylindrical hydrogen storage device with external fins highlighting the influence of different geometric and operational parameters is reported in this paper. The simulation of the storage device is carried out with LaNi₅ as the hydriding alloy.

Keywords: Hydrogen Storage, Metal hydride, Heat and mass transfer, Simulation, LaNi₅

NOMENCLATURE

b	bed thickness [m]
C_p	specific heat [$\text{J kg}^{-1} \text{K}^{-1}$]
D	diffusivity [$\text{m}^2 \text{s}^{-1}$]
d	diameter [m]
E_a	activation energy [J mol^{-1}]
k	thermal conductivity [$\text{W m}^{-1} \text{K}^{-1}$]
k_B	boltzmann constant [eV/K]
$\dot{m}$	hydrogen mass absorbed [$\text{kg m}^{-3} \text{s}^{-1}$]
p	pressure [Pa]
r	radial coordinate [m]
R	universal gas constant [$\text{J mol}^{-1} \text{K}^{-1}$]
s	pitch distance [m]
T	temperature [K]
u	velocity [m s^{-1}]

Greek Letters

ΔH^0	heat of formation [J kg^{-1}]
ε	porosity
θ	polar coordinate [radian]
ρ	density [kg m^{-3}]
τ	time [s]

Subscripts

b	base
c	coolant
e	effective
eq	equilibrium
f	fin
g	gas
i	inside
in	inlet
o	outside
s	solid
sat	saturated
0	initial

1. INTRODUCTION

Solid state hydrogen storage devices contain a bed or matrix of storage material into which hydrogen is absorbed during the charging process. This exothermic process demands effective cooling of the adsorbent in order to achieve the desired storage capacity and charging rate. Depending on the application, the heat transfer medium can be either a liquid or a gas.

A large number of simulation studies reported in literature consider liquid cooled/heated hydrogen storage devices while those dealing with air-cooled/heated systems are scarce. MacDonald and Rowe [1, 2] studied the effects of external convection on the thermodynamic behaviour in a metal hydride tank. Their preliminary resistive analysis suggested that the addition of external fins on the tank could have a significant impact on the overall heat transfer. This was verified by a numerical model, which revealed that when the hydrogen is discharged by a pulsed mass flow rate demand, a finned tank is able to maintain a higher supply pressure. Another interesting observation made in this study was that the hydride in the centre region contributes little to fuel delivery, and removing it would reduce the weight and cost of the system. The paper presented here takes this observation into account by the introduction of a filter in the center of the tube. A parametric study of an air-cooled annular cylindrical hydrogen storage device with external fins highlighting the influence of different geometric and operational parameters is reported here.

2. PHYSICAL MODEL

The annular cylindrical storage device shown in Fig. 1 consists of a filter located at the centre for uniform distribution of hydrogen. The stainless steel container tube has external aluminum fins which transfer the exothermic heat generated to the surrounding air stream. The diameter of the base tube is r_2 and its thickness t_b . The outer diameter of the fin is r_3 and the fin thickness is t_f . The filter has an outer radius r_1 . The pitch distance between consecutive fins is 's'.

Hydrogen gas introduced into the device through filter gets absorbed in the alloy releasing heat. The absorption process will be faster close to the periphery of tubes

and those regions will be saturated earlier than the rest. One can visualize a smooth interface, known as a reaction front which demarcates the fully hydrided portion from the rest of the unsaturated regions. As time proceeds, the reaction front moves inwards from the periphery of tubular device to the unsaturated areas of the alloy near the filter.

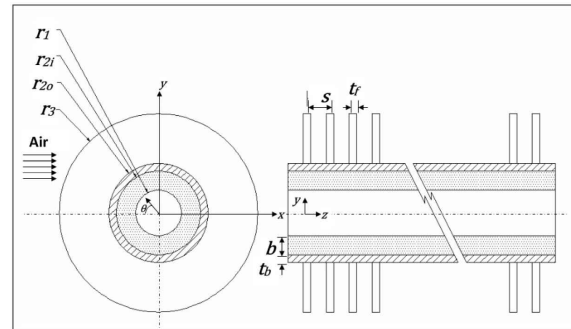


Fig. 1 Schematic diagram of tubular metal hydride storage device with external fins

Various assumptions made for the simplified treatment of the problem are given below:

- i. Hydrogen behaves as an ideal gas at the pressure levels existing in the bed.
- ii. Effects of hysteresis and plateau slope are neglected [3].
- iii. Natural convection and radiation effects within the bed are negligible.
- iv. The hydride bed is assumed to have uniform porosity and is isotropic.
- v. Pressure drop within the bed is negligible.
- vi. Local thermal equilibrium is assumed between the gas and solid within the bed [4].
- vii. Thermo-physical properties are independent of bed temperature, pressure and concentration.

3. PROBLEM FORMULATION

3.1 Mass balance of metal

Hydrogen entering the alloy bed through filters is absorbed forming the

metal hydride. Transient changes in hydride density are attributed to both absorption reaction and diffusive transport due to spatial variation in concentration. As listed under assumptions, constant values of porosity and diffusivity are taken. Conservation of mass for the solid phase of the reactor in cylindrical coordinates is expressed in the following form:

$$(1-\varepsilon)\frac{\partial \rho_s}{\partial \tau} = \dot{m} + (1-\varepsilon)D\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \rho_s}{\partial r}\right) + (1-\varepsilon)D\frac{1}{r^2}\frac{\partial}{\partial \theta}\left(\frac{\partial \rho_s}{\partial \theta}\right) + (1-\varepsilon)D\frac{\partial}{\partial z}\left(\frac{\partial \rho_s}{\partial z}\right) \dots (1)$$

ρ_s denotes the density of the hydride, D denotes the diffusivity and $\dot{m}$ represents the reaction term. D may be expressed by the Arrhenius expression [5].

$$D = D_0 \exp\left(\frac{-H_a}{k_B T}\right) \dots (2)$$

Where H_a represent activation enthalpy and k_B is the Boltzmann constant.

Transient variation in density on the left hand side of eqn. (1) is expressed as the sum of three terms on the right hand side. The first term represents the mass of hydrogen absorbed due to reaction. The second and third terms of the equation signify the effect of diffusive transport due to the concentration gradients existing within the bed. Convection term is neglected.

3.2 Reaction kinetics

The amount of hydrogen absorbed is directly related with the reaction rate, and could be given as follows:

$$\dot{m} = -C_a \exp\left(-\frac{E_a}{RT}\right) \ln\left(\frac{P}{P_{eq}}\right)(\rho_{sat} - \rho_s) \dots (3)$$

where C_a is a material dependent constant, E_a is the activation energy of the material,

ρ_{sat} is the density of hydride at saturation. The change in volume if any, due to hydrogen absorption is not considered in the computation of density of the bed. The equilibrium pressure is determined by the van't Hoff relationship as given below:

$$\ln P_{eq} = A - \frac{B}{T} \dots (4)$$

Where A and B are van't Hoff constants.

3.3 Energy balance

Heat generated due to exothermic reaction causes associated spatial temperature imbalances within the bed. Energy conservation equation of the bed could be expressed as given below:

$$(\rho C_p)_e \frac{\partial T}{\partial \tau} = k_e \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) + k_e \frac{1}{r^2} \frac{\partial}{\partial \theta} \left(\frac{\partial T}{\partial \theta} \right) + k_e \frac{\partial}{\partial z} \left(\frac{\partial T}{\partial z} \right) - \dot{m} \Delta H^0 \dots (5)$$

Equation (5) assumes the existence of local thermal equilibrium between the solid and gas within the bed. Hence the heat transfer between the two phases is omitted. Radiation heat transfer to the ambient is also neglected. Heat of formation of hydride represents the source term in the equation.

Effective volumetric heat capacity is expressed as follows:

$$(\rho C_p)_e = (\varepsilon \rho_g C_{pg} + (1-\varepsilon) \rho_s C_{ps}) \dots (6)$$

Effective thermal conductivity is expressed as given below:

$$k_e = \varepsilon k_g + (1-\varepsilon) k_s \dots (7)$$

3.4 Initial and boundary conditions

Initially the pressure and temperature of reactor bed are assumed to be uniform.

$$p = p_0; \quad T = T_0; \quad \rho = \rho_0 \quad \text{at} \quad \tau = 0 \dots (8)$$

The boundary walls of the reactor are assumed to be impermeable and adiabatic. The reaction heat is removed from the porous bed by passing the air stream over the tubular device. “No slip condition” is valid at the reactor walls. Hydrogen is supplied using porous filter tubes. Convective flux conditions prevail at the filter walls which are assumed to be adiabatic. Based on the above assumptions, the following boundary conditions are written:

Reactor wall at $r = r_{2i}$:

$$\frac{\partial p}{\partial r} = 0 \quad ; \quad \text{at} \quad \tau > 0 \quad \dots\dots (9)$$

Reactor wall at $r = r_{2o}$:

$$-k \frac{\partial T}{\partial r} = h_f (T_f - T) \quad \text{at} \quad \tau > 0 \quad \dots(10)$$

Filter wall at $r = r_1$:

$$p = p_{in} \quad ; \quad \frac{\partial T}{\partial r} = 0 \quad \text{at} \quad \tau > 0 \dots (11)$$

4. SIMULATION METHODOLOGY

The simulation of the metal hydride bed employing the above mentioned partial differential equations were carried out using the commercial code COMSOL MULTIPHYSICS [6]. An unstructured mesh of element size 0.001 m is generated by defining suitable mesh parameters. Most of the cases involve degrees of freedom in the range 50000 – 100000. The mass (convection-diffusion) and energy equations (transient conduction) in differential form are solved with specified initial and boundary conditions.

5. RESULTS AND DISCUSSION

Results are computed for LaNi₅ as the hydriding material. Fast reaction kinetics

and acceptable operating temperature makes it a material of choice in the present study. Moreover, reaction kinetics and thermo-physical property data for LaNi₅ are readily available in literature as given in Table 1. The simulation is carried out for various operating parameters, the ranges of which are given in Table 2.

Parameter	LaNi ₅	Hydrogen
Density, kg m ⁻³	8200	0.0838 at NTP
Specific heat, J kg ⁻¹ K ⁻¹	419	14,890
Effective thermal conductivity, W m ⁻¹ K ⁻¹	1.2	0.12
Activation energy, J mol ⁻¹	21179.6	-
Constants in eqns. (2), (3), (4): D ₀ (m ² /s)	3.4 x 10 ⁻⁷	-
H _a (eV)	0.29	-
C _a	59.187	-
A	12.99	-
B	3704.59	-

Table 1 Thermo-physical properties of LaNi₅ and hydrogen [3, 5, 7]

Parameter	Range of values
Initial temperature of hydride bed, T ₀ (K)	293 (Constant)
Supply pressure of hydrogen, p (bar)	7.0 – 15.0
Coolant (air) temperature, T _c (K)	290 – 320
Velocity of air stream, u (m/s)	3.5
Porosity, ε	0.5 (Constant)
Bed thickness, b (mm)	2 – 6
Thickness of the base tube, t _b (mm)	2
Thickness of fin, t _f (mm)	2
Pitch distance, s (mm)	6
Fin height, L (mm)	3 (low-fin), 13 (high-fin)

Table 2 Parameter values used in the present study

The main parameters which influence the mass transfer process in the hydride bed include the hydrogen gas pressure, hydride bed temperature and the hydrogen concentration. Metal hydrides generate large amount of heat due to the exothermic hydride forming reaction. The reaction within the bed is proportional to the pressure gradient between the gas supply pressure and equilibrium pressure corresponding to the bed temperature. While the supply pressure remains constant, the reactions become more vigorous when

the bed temperature and the corresponding equilibrium pressure are lower. The bed temperature increases rapidly during the initial phase of the reaction. Heat transfer from the bed to the cooling media dissipates the heat generated in the process. Since the thermal diffusion process is comparatively slower than the chemical reaction, the hydriding rate drops as the time progresses.

Exothermic heat generated within the bed during absorption is being carried away by the air stream flowing over the tubular device. Heat transfer to the surrounding air stream is enhanced by means of fins. Convective heat transport by means of air at different regions is attributed to the nature of flow field. Figure 2 shows the slice plots of temperature profile of metal hydride bed at different time intervals. At the initial phase of reaction the exothermic heat generation will be excessive which causes a rapid rise in temperature within the bed. Later, the rate of reaction is controlled by the heat transfer to the surrounding media, which causes a gradually decreasing temperature within the bed.

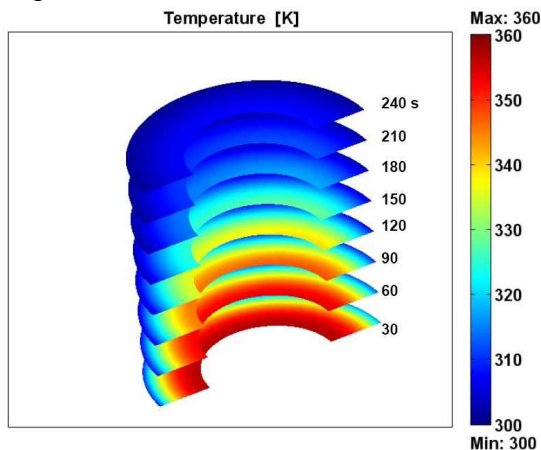


Fig. 2 Temperature profiles of metal hydride bed at selected time intervals

Figure 3 depicts the concentration profiles within the bed. The reaction front progresses relatively faster during the early stages and becomes more and more sluggish as time proceeds, mainly attributed to the increasing conduction path length

and poor heat transfer. The above figure also shows the temperature profile inside the surrounding flow field. The rate of removal of heat is more during the early stages as depicted in figure, further indicative of the higher rate of reaction.

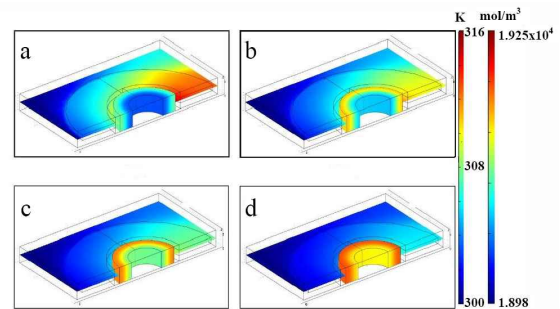


Fig. 3 Formation of hydride inside tubular storage device with fins kept within the air stream during absorption at different time intervals a) 30 s b) 60 s c) 90 s d) 120 s (b=5 mm, p=15 bar, $T_c=300$ K)

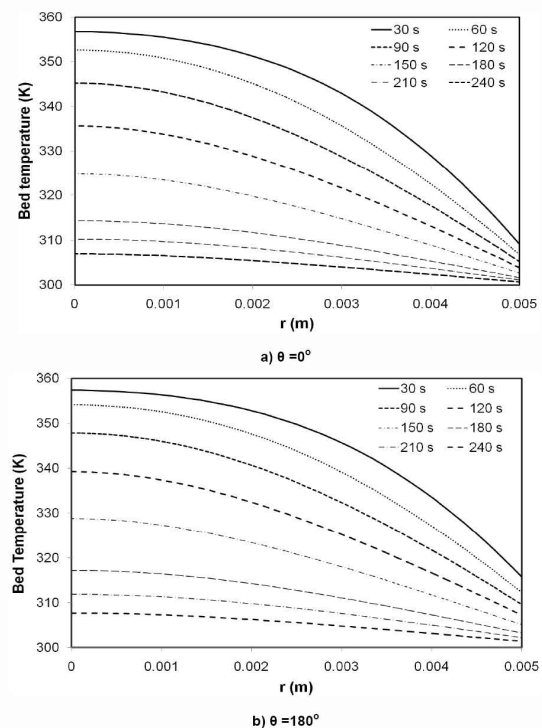


Fig. 4 Profiles of temperature variation across the bed at different radial distances from filter and two different angles a) $\theta = 0^\circ$ b) $\theta = 180^\circ$ (b=5 mm, p=15 bar, $T_c=300$ K)

Heat transfer from the device shows some disparity between the leading and trailing regions, owing to heat pickup of the air stream as it flows along. Thereby development of reaction fronts shows a non uniform distribution i.e., higher rate of absorption in the leading half compared to the trailing half of the bed. Figure 4 shows the temperature variation across two different cross sections at angles 0 and 180 degrees during different time intervals. Even though the two plots exhibit similar trends, the temperature profile at $\theta=0^\circ$ show lower bed temperature compared to $\theta=180^\circ$. The differences are more evident during the initial phase of the reaction and near the finned wall where the reaction rates are more predominant.

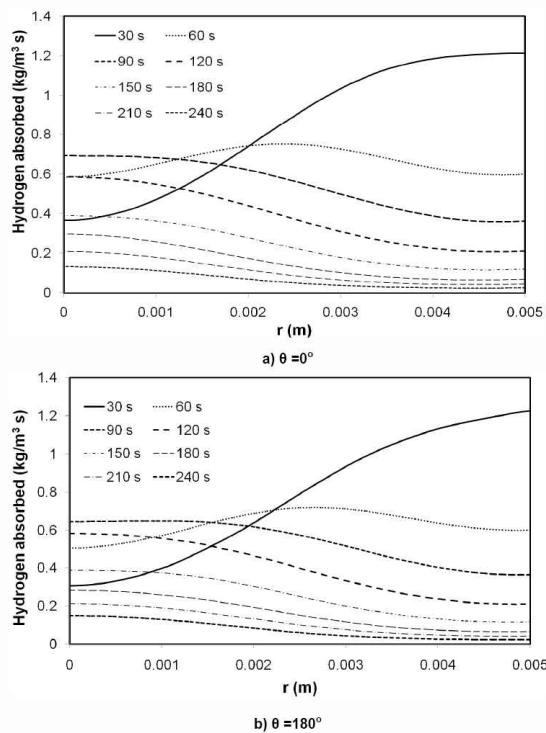


Fig. 5 Profiles of hydrogen absorption across the bed at different radial distances from filter and two different angles a) $\theta = 0^\circ$ b) $\theta = 180^\circ$ ($b=5$ mm, $p=15$ bar, $T_c=300$ K)

Lower bed temperature essentially lowers the corresponding equilibrium pressure within the device, which enhances the rate of hydrogen sorption within the bed. Figure 5 shows rate of hydrogen sorption at

two different cross sections at angles 0 and 180 respectively. At the initial stages of reaction there exist a disparity in absorption between the finned wall and filter wall which is clearly evident from the above figure. At later stages the above disparity smoothens out, may be an indication of the reaction becomes more heat transfer controlling. Still the disparity between the leading and trailing halves may be observed at locations where the reaction is more vigorous. The plots depict a progressive wave front behaviour from tube wall to the filter wall indicative of the progressing reaction front moving across the cross section.

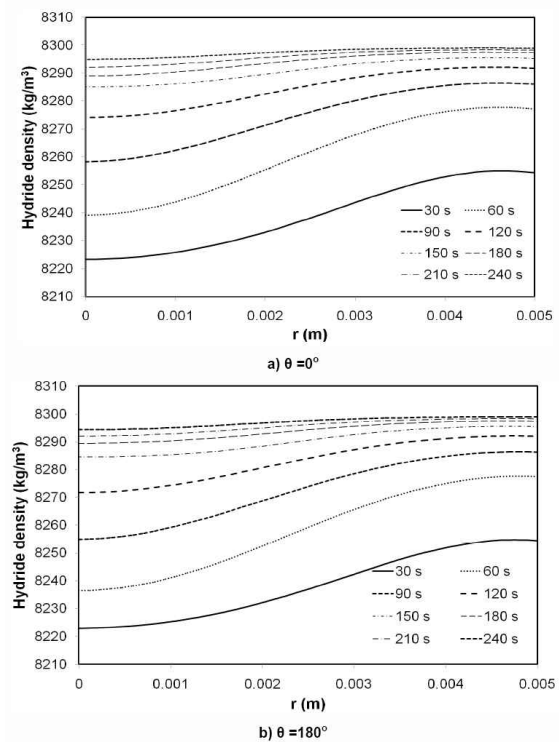


Fig. 6 Profiles of hydride density across the bed from filter at different radial distances and two different angles a) $\theta = 0^\circ$ b) $\theta = 180^\circ$ ($b=5$ mm, $p=15$ bar, $T_c=300$ K)

Figure 6 depicts time varying profiles of hydride density across the bed at two different cross sections at angles 0 and 180 degrees respectively. Hydride density at the finned wall is obviously high at all different cases. The rate of increase in density is higher at the initial stages of reaction and

becomes more and more sluggish at the ending stage. Both plots exhibit similar behavior barring the usual disparity between the leading and trailing halves.

Bed thickness has been recognized as an important geometric parameter that influences the performance of any hydrogen storage device. For thinner beds, the conduction path is relatively short. If the thickness exceeds an optimum value, the gain due to higher quantity of hydride is negated by the increased conduction resistance to heat transfer. At specified conditions of cooling media temperature and pressure, rate of absorption depends on bed thickness. Figure 7 shows the influence of bed thickness on absorption of hydrogen.

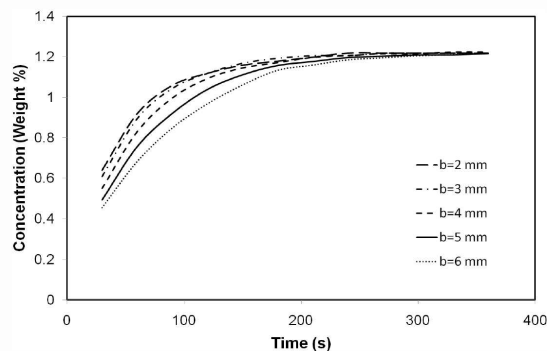


Fig. 7 Effect of bed thickness on hydride formation ($p=15$ bar, $T_c=300$ K)

In all cases, the same saturated state of approx. 1.2 weight % H_2 is achieved after different time intervals. At a lower bed thickness of $b=2$ mm, the hydride bed reaches saturation earlier than others leading to the least cycle time. Higher bed thicknesses result in flatter profiles and take longer time to reach the saturation state. Higher bed temperature, lower reaction rate, larger conduction path length and low thermal conductivity material are collectively important for the above described phenomena. For a given bed thickness, heat transfer as well as reaction kinetics are jointly important in determining the absorption time. As reaction kinetics is more material dependent, for a specified effective thermal conductivity value, there exists an optimum

bed thickness satisfying the prescribed performance and system weight.

Heat transfer augmentation is always important in the design of metal hydride devices. Figure 8 shows the absorption rates with and without fins. Device with fins shows visible improvement in heat transfer over the one with no fin leading to higher rates of absorption. It may be noted further that the performance improvement of high-fin (13 mm) tube over the low-fin (3 mm) tube is small.

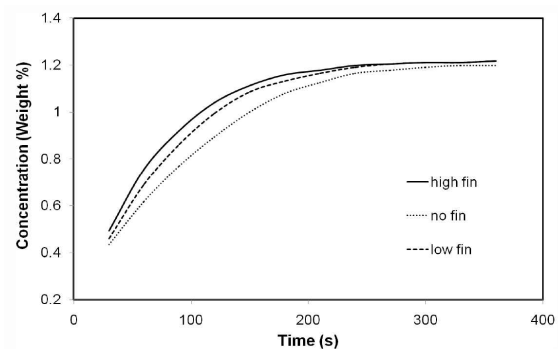


Fig. 8 Effect of external fins on rate of hydriding ($b=5$ mm, $p=15$ bar, $T_c=300$ K)

There are two major operational parameters which control the overall performance of the device, namely, system supply pressure and surrounding coolant media (air) temperature. Supply pressure is a very important operating parameter controlling the hydriding rates. The pressure differential between equilibrium pressure and imposed pressure within the reactor controls the rate of absorption process. A higher pressure differential reduces the cycle time as illustrated in Fig. 9. Cooling media temperature is a major parameter in controlling the temperature of the bed. Effect of cooling media temperature on hydriding is shown in Fig. 10. At any given supply pressure, lower coolant temperature causes a lower bed temperature and thereby lower equilibrium pressure. It consequently increases the driving potential for mass transfer, resulting in a higher rate of reaction.

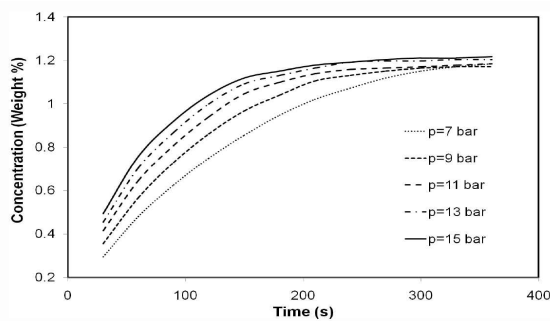


Fig. 9 Effect of supply pressure on hydrogen sorption ($b=5\text{mm}$, $T_c=300\text{ K}$)

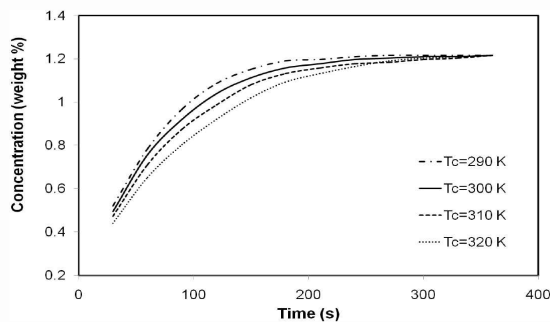


Fig. 10 Effect of coolant (air) temperature on hydride formation ($b=5\text{mm}$, $p=15\text{ bar}$)

6. CONCLUSIONS

Heat and mass transfer simulation of air cooled hydrogen storage device with external fins is carried out with LaNi_5 as the hydriding material for different geometric parametric ranges and operational conditions. Results highlight disparity in hydriding rates between the leading and trailing halves of the tubular hydrogen storage device. This is more pronounced during the early stages of reaction when the reaction rate is high. The results confirm the importance of bed thickness as a major geometric parameter controlling the rate of hydrogen absorption. Providing external fins enhances the hydrogen sorption of metal hydride bed, however effect of fin height on the overall performance is minimal. Hydrogen pressure within the bed is another important operational parameter influencing the hydriding rate. Temperature of cooling air

stream passing over the tubular device influences the overall hydriding performance of the device to a reasonable extent.

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