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Analytical comparison and criteria for heat and mass transfer models in metal hydride packed beds

A. V. KUZNETSOV
Mechanical Engineering Research Institute of Russian Academy of Sciences, 101830 Moscow, Russia

and

K. VAFAI†
Department of Mechanical Engineering, The Ohio State University, Columbus, OH 43210, U.S.A.

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Abstract—This paper puts forward a mathematical model of transport processes in a metal hydride packed bed along with an analytical quantification of the frequently used assumptions. The complete model takes into account heat transfer by conduction, convection, mass transfer, as well as reaction kinetics and convective interactions between the fluid and solid phases. By analyzing several frequently used assumptions, a system of useful and practical analytical criteria is obtained. The criteria reveal when simplifications of the model are possible. The simplified models obtained by reduction of the complete model can be effectively used in describing transport processes in metal hydride beds within the limits determined by a set of analytical criteria. Specifically, criteria for the validity of local thermal equilibrium, steady-state approximation, the frontal model approximation, and the filtration approximation are established analytically.

1. INTRODUCTION

Application of metal hydrides ($LaNi_5H_x$, $LaNi_{5-y}Al_yH_x$, $FeTiH_x$, etc.) for different types of power installations is a subject of permanent interest [1–3]. This interest is due to high technological potentials, such as creating safe hydrogen storage systems (as an alternative to storage of liquid hydrogen), or devising highly effective thermochemical compressors and heat pumps and ecologically clean refrigeration systems (as an alternative to CFC refrigerators). These compounds have a unique property: under normal conditions they are able to reversibly absorb large amounts of hydrogen, and nothing but hydrogen, as they have a remarkably large hydrogen storage capacity [4].

For effective design of metal hydride installations, a mathematical model which effectively describes the physical and chemical processes in a metal hydride bed is needed. The main factors governing the dynamics of the metal hydride bed are: heat transfer from/to hydration zone, hydrogen filtration through the metal hydride matrix and the kinetics of hydrogen sorption-desorption processes. In recent publications a number of pertinent and effective mathematical models for the process have been suggested. For example, in Fateev *et al.* [5] a frontal model of chemical reaction in the metal hydride bed is proposed. In Sun and Deng [6] numerical results for the calculation of heat transfer processes in metal hydride containers of different configurations are presented. The influence of hydrogen filtration on the dynamics of the processes in metal hydride bed is considered in refs. [7–11]. The influence of the kinetics of the chemical reaction is considered in refs. [10–12].

Even though extensive studies of heat and mass transfer processes in metal hydride beds are presented in recent publications, most of the models which are used for describing the processes within the metal hydrides proceed from the assumption of local thermal equilibrium between hydrogen and hydride. Investigation of the local thermal equilibrium is of importance because temperature of hydrogen entering a metal hydride bed (in metal hydride heat pump or refrigerator) can substantially differ from temperature of the bed.

In ref. [13] an analytical investigation of a wave of temperature difference between solid and fluid phases for two-phase model of porous bed is presented. In refs. [14–16] Vafai and Sözen presented a very general set of volume-averaged governing equations for non-thermal equilibrium condensing forced fluid flow through a latent heat storage porous bed and they have carried out a comprehensive analysis of the phenomenon. Our present investigation is, to a considerable extent, based on this research. We introduce

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†Author to whom correspondence should be addressed.

NOMENCLATURE

a	thermal diffusivity, $\lambda/c_p\rho$ [$m^2\ s^{-1}$]
a_σ	effective thermal diffusivity of solid phase, $\lambda_{\sigma eff}/(c_p)_\sigma\rho_\sigma$ [$m^2\ s^{-1}$]
$a_{\sigma\gamma}$	specific surface area common to solid and gas phases [$m^2\ m^{-3}$]
c_p	specific heat at constant pressure [$J\ kg^{-1}\ K^{-1}$]
C_{eff}	effective specific heat per unit volume [$J\ m^{-3}\ K^{-1}$]
d	average diameter of metal hydride particle [m]
E_{hyd}	heat of formation of hydride per unit volume [$J\ m^{-3}$]
E_a	activation energy [$J\ kmol^{-1}$]
$h_{\sigma\gamma}$	hydrogen-to-particle heat transfer coefficient [$W\ m^{-2}\ K^{-1}$]
Δh_{hyd}	heat of hydration reaction [$J\ kmol^{-1}\ H_2$]
ΔH_{hyd}	heat of hydration reaction [$J\ kg^{-1}$]
K	hydration reaction rate constant [s^{-1}]
$\dot{m}$	sorption rate [$kg\ m^{-3}\ s^{-1}$]
M	molecular weight [$kg\ kmol^{-1}$]
p	pressure [Pa]
p_b	pressure which is maintained at hydrogen inlet—outlet boundary [Pa]
P_0	$= 10^5$ Pa—reference pressure at the hydration front
Pr	Prandtl number, $\mu(c_p)_\gamma/\lambda_\gamma$
q	heat flux [$W\ m^{-2}$]
$\tilde{R}$	universal gas constant [$J\ kmol^{-1}\ K^{-1}$]
Re_p	particle Reynolds number, jd/μ
Ste	Stefan number, $c_p\rho\Delta T/E_{hyd}$
t	time [s]
T	temperature [K]
T_b	temperature which is maintained at the heat removal boundary [K]
T^*	temperature calculated under local thermal equilibrium assumption [K]
$\tilde{T}$	temperature of upper discontinuity in isobar in piecewise-linear approximation [K]
ΔT_{eq}	equilibrium temperature range of hydration reaction [K]
ΔT	characteristic temperature interval [K]
$\mathbf{v}$	velocity vector [$m\ s^{-1}$].

Greek symbols

ε	porosity
κ	hydrogen concentration, that is hydrogen—metal atomic ratio [katom H/kmol intermetallide compound]
Θ	dimensionless temperature, $T/T^*\vert_{x=0}$
$\tilde{\Theta}$	dimensionless temperature, $T/T_\sigma\vert_{x=0}$
λ	thermal conductivity [$W\ m^{-1}\ K^{-1}$]
Λ	empirical filtration coefficient
μ	absolute viscosity for hydrogen [$kg\ m^{-1}\ s^{-1}$]
ρ	density [$kg\ m^{-3}$]
ω	hydrogen concentration ratio, $\langle\rho_H\rangle^\sigma/\langle\rho_H\rangle^\sigma_{max} = \kappa/\kappa_{max}$.

Subscripts

b	boundary
eff	effective property
eq	equilibrium state
f	hydration front
H	fixed hydrogen
in	inlet
il	inlet layer
max	maximum
0	initial
γ	gas phase (hydrogen)
σ	solid phase (metal hydride).

Superscripts

a	analytical solution
n	numerical solution.

and analyze a model which takes into account hydrogen diffusion, hydration reaction kinetics, and heat transfer by conduction, convection, and convective interaction between hydrogen and hydride. The possibility of considering locally non-thermal equilibrium processes makes this model more general than the models which have been suggested in refs. [5–12] for heat and mass transfer processes in metal hydride bed. By analyzing a more generalized model under different assumptions, a system of analytical criteria is obtained establishing conditions when simplifications of the model are possible. The simplified models obtained by reduction of the full model for particular values of the criteria will be demonstrated to coincide with different models suggested in refs. [5–12].

2. PROBLEM STATEMENT

2.1. *Assumptions*

In establishing a model for analyzing this problem, the following customary assumptions and simplifications are employed:

(I) hydrogen in the hydride pores behaves as an ideal gas;

(II) natural convection effects are neglected;

(III) the hydride matrix has uniform porosity and is isotropic;

(IV) inter-particle radiation heat transfer is neglected;

(V) kinetics of chemical reaction is described by the shrinking core model [10].

2.2. *Governing equations*

The governing conservation equations as well as the thermodynamic relations are developed by utilizing the 'local volume averaging' technique [14–18]. Following the analysis in refs. [14–18], the volume-averaged governing equations can be written as:

gas phase (hydrogen) continuity equation

$$\frac{\partial}{\partial t}(\varepsilon\langle\rho_\gamma\rangle^\gamma)+\nabla\cdot(\langle\rho_\gamma\rangle^\gamma\langle\mathbf{v}_\gamma\rangle) = -\langle\dot{m}\rangle \qquad (1)$$

gas phase equation of motion

$$\nabla\langle p_\gamma\rangle^\gamma = -\frac{\mu}{\Omega_\gamma}\langle\mathbf{v}_\gamma\rangle^\gamma \qquad (2)$$

where $\Omega_\gamma = \frac{\Lambda\varepsilon^3\,\mathrm{d}^2}{(1-\varepsilon)^2}$ is obtained from experimental results [9];

gas phase energy equation

$$\varepsilon\langle\rho_\gamma\rangle^\gamma(c_\mathrm{p})_\gamma\frac{\partial\langle T_\gamma\rangle^\gamma}{\partial t}+\langle\rho_\gamma\rangle^\gamma(c_\mathrm{p})_\gamma\langle\mathbf{v}_\gamma\rangle^\gamma\nabla\langle T_\gamma\rangle^\gamma$$
$$= \nabla\cdot[\lambda_{\gamma\mathrm{eff}}\nabla\langle T_\gamma\rangle^\gamma]+h_{\sigma\gamma}a_{\sigma\gamma}[\langle T_\sigma\rangle^\sigma-\langle T_\gamma\rangle^\gamma] \qquad (3)$$

solid phase (metal hydride) energy equation

$$(1-\varepsilon)\rho_\sigma(c_\mathrm{p})_\sigma\frac{\partial\langle T_\sigma\rangle^\sigma}{\partial t} = \nabla\cdot[\lambda_{\sigma\mathrm{eff}}\nabla\langle T_\sigma\rangle^\sigma]$$
$$-h_{\sigma\gamma}a_{\sigma\gamma}[\langle T_\sigma\rangle^\sigma-\langle T_\gamma\rangle^\gamma] \qquad (4)$$

equation of state for the hydrogen gas

$$\langle p_\gamma\rangle^\gamma = \langle\rho_\gamma\rangle^\gamma\tilde{R}\langle T_\gamma\rangle^\gamma/M_\gamma \qquad (5)$$

continuity equation for the adsorbed hydrogen

$$\frac{\partial}{\partial t}((1-\varepsilon)\langle\rho_\mathrm{H}\rangle^\sigma) = \nabla\cdot(D_\mathrm{eff}\nabla\langle\rho_\mathrm{H}\rangle^\sigma)+\langle\dot{m}\rangle \qquad (6)$$

kinetic equation of hydrogen sorption-desorption is obtained from the results presented in ref. [10]

$$\langle\dot{m}\rangle = K\exp\left(-\frac{E_\mathrm{a}}{\tilde{R}\langle T_\sigma\rangle^\sigma}\right)$$
$$\times\frac{(\langle p_\gamma\rangle^\gamma)^{1/2}-p_\mathrm{eq}^{1/2}}{[(1-\langle\rho_\mathrm{H}\rangle^\sigma/\langle\rho_\mathrm{H}\rangle^\sigma_\mathrm{max})]^{-1/3}-1}. \qquad (7)$$

In all analyses in metal hydride beds it is customary to use the following additional assumption:

(VII) kinetics of the chemical reaction are not the limiting factor, so instead of equation (7) it is possible to use equations which determine the local chemical equilibrium at each point between the free and fixed hydrogen $\langle p_\gamma\rangle^\gamma = p_\mathrm{eq}(\langle T_\gamma\rangle^\gamma, \langle\rho_\mathrm{H}\rangle^\sigma)$.

Applicability of assumption VII depends on the type of metal hydride used. According to refs. [5, 9] this assumption is valid, for example, for $\mathrm{LaNi_5H}_x$ metal hydride. If Darcy's law in the form of equation (2) is used as an equation of motion, it is possible to combine equations (1) and (2). Substitution of equation (2) into equation (1) leads to:

$$\frac{\partial}{\partial t}(\varepsilon\langle\rho_\gamma\rangle^\gamma) = \nabla\cdot\left(\frac{\Omega_\gamma}{\mu_\gamma}\langle\rho_\gamma\rangle^\gamma\nabla\langle p_\gamma\rangle^\gamma\right)-\langle\dot{m}\rangle. \qquad (8)$$

If more general models are employed for the flow (such as those discussed in Amiri and Vafai [19]) this simplification is not possible. The set of equations (3)–(8) determines the behavior of the system under consideration. The unknowns for this set of equations are $\langle T_\gamma\rangle^\gamma$, $\langle T_\sigma\rangle^\sigma$, $\langle p_\gamma\rangle^\gamma$, $\langle\rho_\gamma\rangle^\gamma$ and $\langle\rho_\mathrm{H}\rangle^\sigma$.

3. ANALYSIS

The set of equations (3)–(8) describes a general model for heat and mass transfer accompanied by chemical reaction in a metal hydride bed. The models suggested in refs. [5–12] can be obtained from this general model once additional assumptions are made.

It is of both theoretical and practical importance to obtain analytical criteria determining the conditions when pertinent assumptions can be applied to simplify the general model described by equations (3)–(8). This will establish the area of applicability and will allow the determination of simple engineering models for particular technical applications. Analytical criteria for the validity of the following assumptions/approximations will be considered in this work:

(VIII) local thermal equilibrium assumption;

(IX) steady state approximation;

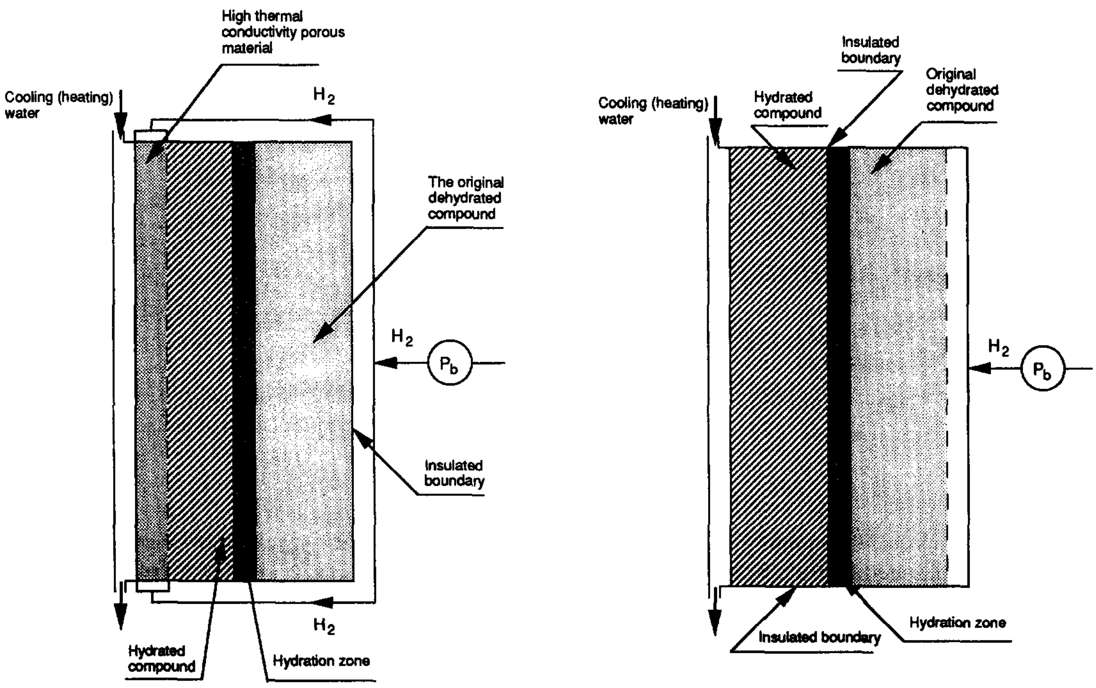
(X) a frontal model approximation for the hydration reaction;

(XI) uniform hydrogen pressure approximation.

In order to introduce strict mathematical criteria for the applicability of assumptions VIII–XI a systematic analysis will be considered based on a number of simplified problems which are derived from the general model. Each of these problems will be solved with and without additional assumptions leading to an analytical criterion. These analytical criteria and formulations of simplified problems employed for their derivation are summarized in Table 1. We will consider two bed configurations. For the first configuration [Fig. (1a)] hydrogen inlet boundary and heat removal (or heating) boundary coincide: hydrogen enters at the same boundary from which heat is extracted. The high thermal conductivity porous medium used in this configuration is used just for providing a uniform inlet for the hydrogen. In the second configuration [Fig. (1b)] hydrogen inlet and heat removal (or heating) boundaries do not coincide. For all simplified problems a one-dimensional approach is utilized: heat and mass transfer processes are assumed to be in the x-direction only. It is important to note that for either one of these configurations

Table 1. Analytical criteria for the validity of the assumptions

Assumption	Analytical criteria for the validity of the assumptions	Formulation of the simplified problem employs the following assumptions	Corresponding equation number
VIII—local thermal equilibrium	$\frac{5p_{\mathrm{in}}v_\gamma}{T_{\mathrm{in}}L}\frac{M_\gamma(c_{\mathrm{p}})_\gamma}{\tilde{R}_{\sigma\gamma}a_{\sigma\gamma}} \ll 1$	I–V, VII, IX–XIII	(16)
IX—steady state approximation	For heat transfer: $\frac{(c_{\mathrm{p}})_\sigma\rho_\sigma[T_{\mathrm{eq}}(p_{\mathrm{b}},\kappa_0)-T_{\mathrm{b}}]}{6E_{\mathrm{hyd}}} \ll 1$ For filtration: $\frac{2M_\sigma}{1-\varepsilon}\frac{1}{\rho_\sigma(\kappa_{\max}-\kappa_0)}\frac{\varepsilon\Delta p}{\tilde{R}T} \ll 1$	I–V, VII, VIII, X, XI, XIV	(23) (24)
X—a frontal model. This criterion also characterizes assumption X′	$\frac{1}{2}\frac{\Delta T_{\mathrm{eq}}}{\tilde{T}(p_{\mathrm{b}})-T_{\mathrm{b}}}+\frac{1}{2}\frac{(c_{\mathrm{p}})_\sigma\rho_\sigma\Delta T_{\mathrm{eq}}}{E_{\mathrm{hyd}}} \ll 1$ Different inlet boundaries additionally require: $\frac{T_{\mathrm{f}}^2}{2B(T_{\mathrm{f}}-T_{\mathrm{b}})}\frac{p_{\mathrm{b}}^2-p_{\mathrm{f}}^2}{p_{\mathrm{f}}^2} \ll 1$	I–V, VII, VIII, X′, XI, XIV	(27) (36)
XI—hydrogen pressure is assumed to be the same throughout the bed	$\frac{\frac{B}{A-\ln(p_{\mathrm{b}}/P_0)}\left(\frac{B}{A-\ln(p_{\mathrm{b}}/P_0)}-T_{\mathrm{b}}\right)}{p_{\mathrm{b}}^2}\frac{\gamma_\sigma}{E_{\mathrm{hyd}}} \times \frac{(\kappa_{\max}-\kappa_0)\rho_\sigma}{\Lambda M_\sigma}\mu\tilde{R}\frac{(1-\varepsilon)^2}{\varepsilon^3}\left(\frac{1}{d}\right)^2 \ll 1$	I–V, VII–X, XIV	(34)
XIV—convective heat transfer is negligibly small compared to conductive heat transfer	$\frac{(c_{\mathrm{p}})_\gamma M_\gamma\Delta T_\gamma}{\Delta h_{\mathrm{hyd}}} \ll 1$	I–V, VII, IX–XIII	(17)



(a) (b)

Fig. 1. (a) A schematic of metal hydride bed for which hydrogen inlet boundary and heat removal boundary coincide. This scheme is utilized in analyzing assumptions VIII and XIV. (b) A schematic of metal hydride bed for which hydrogen inlet boundary and heat removal boundary coincide. This schematic is utilized in analyzing assumptions IX, X and XI.

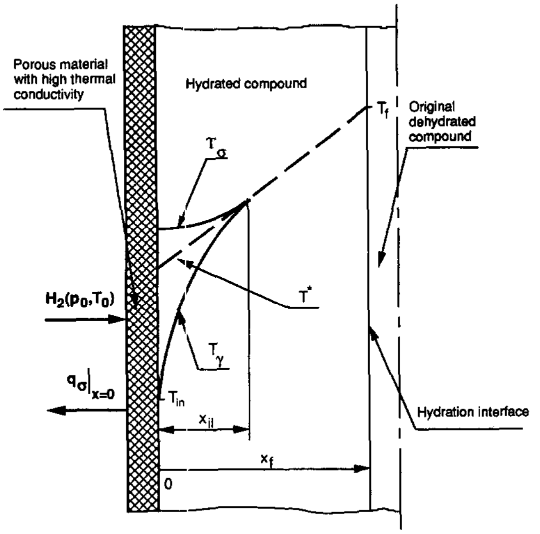


Fig. 2. Schematic of the problem used for calculation of hydrogen and hydride temperatures in a one-dimensional metal hydride layer.

hydration occurs only as a result of the temperature distribution. That is hydration is dominated by the heat transfer process.

3.1. *Criterion for the validity of local thermal equilibrium assumption* (*assumption* VIII)

To estimate the influence of convective heat exchange between the gas and the solid matrix in a metal hydride packed bed, the scheme shown in Fig. 1(a) is utilized. The problem shown in Fig. 2 which corresponds to the scheme shown in Fig. 1(a) is analyzed. The heat generated at the hydration front is removed at the left boundary $x = 0$. The temperature of hydrogen at the inlet is T_{in} and the inlet pressure is p_{in}. To simplify the notations the averaging operator is omitted. To solve the problem analytically, in addition to assumptions I–V, VII, IX and X the following assumptions are also invoked:

(XII) heat transfer in the gas phase is due to forced convection only—i.e. conduction in the gas phase is neglected;

(XIII) $h_{\sigma\gamma}$ = constant—this is true for small values of Reynolds number.

It should be noted that the criteria for non-customary assumptions, IX and X used in here will be established later on. Under the above-cited assumptions the problem is described by the following set of equations:

$$\frac{d}{dx}\left(\frac{p_\gamma}{T_\gamma}\frac{dp_\gamma}{dx}\right) = 0 \tag{9}$$

$$\frac{p_\gamma}{T_\gamma}\frac{dT_\gamma}{dx} = \beta_1[T_\sigma - T_\gamma] \tag{10}$$

$$\frac{d^2T_\sigma}{dx^2} = \beta_2[T_\sigma - T_\gamma] \tag{11}$$

subject to the following boundary conditions

$$\lambda_\sigma \frac{\partial T_\sigma}{\partial x}\bigg|_{x=0} = \text{const.} \quad T_\sigma|_{x=x_f} = T_f$$

$$T_\gamma|_{x=0} = T_{in} \quad p|_{x=0} = p_{in} \quad p|_{x=x_f} = p_f = f(T_f)$$

where

$$\beta_1 = \frac{h_{\sigma\gamma}a_{\sigma\gamma}}{(c_p)_\gamma v_\gamma}\frac{\tilde{R}}{M_\gamma}$$

and

$$\beta_2 = \frac{h_{\sigma\gamma}a_{\sigma\gamma}}{(1-\varepsilon)\lambda_\sigma}.$$

Equation (9) is obtained from using equations (2) and (5) in equation (1) and equations (10) and (11) are obtained from equations (3) and (4), respectively. The system of equations (9)–(11) is still complicated for an analytical solution, so assumption XI is also used, i.e. $p_\gamma \approx p_{in}$ = const. throughout the metal hydride. The criterion for this assumption will be listed later. Now the set of equations (9)–(11) can be reduced to the following set

$$\frac{d^2T_\sigma}{dx^2} = \beta_2\left[T_\sigma - T_{in}\exp\left(\frac{\beta_1}{p_0\beta_2}\left(\frac{dT_\sigma}{dx} - \frac{dT_\sigma}{dx}\bigg|_{x=0}\right)\right)\right] \tag{12}$$

$$T_\gamma = T_{in}\exp\left(\frac{\beta_1}{p_0\beta_2}\left(\frac{dT_\sigma}{dx} - \frac{dT_\sigma}{dx}\bigg|_{x=0}\right)\right). \tag{13}$$

Equation (12) is then solved analytically utilizing a Taylor series expansion resulting in

$$T_\sigma = C_1\exp\left(-\frac{T_{in}\beta_1}{p_{in}}x\right) + C_2\exp\left(\frac{p_{in}\beta_2}{T_{in}\beta_1}x\right) - \left[\frac{T_{in}\beta_1}{p_{in}\beta_2}\frac{dT_\sigma}{dx}\bigg|_{x=0} - T_{in}\right] \tag{14}$$

where

$$C_2 = \frac{T_f + \left[\frac{T_{in}\beta_1}{p_{in}\beta_2}\frac{dT_\sigma}{dx}\bigg|_{x=0} - T_{in}\right]}{\exp\left(\frac{p_{in}\beta_2}{T_{in}\beta_1}x_f\right)}$$

$$C_1 = \frac{\frac{dT_\sigma}{dx}\bigg|_{x=0} - C_2\frac{p_{in}\beta_2}{T_{in}\beta_1}}{-\frac{T_{in}\beta_1}{p_{in}}}.$$

The x-dependency of T_σ is described by the first two terms in equation (14). The term $C_1\exp(-(T_{in}\beta_1/p_{in})x)$ essentially differs from zero only in a small neighborhood next to $x = 0$, where temperatures of solid and gas differ significantly. The term $C_2\exp(p_{in}\beta_2/T_{in}\beta_1)x)$

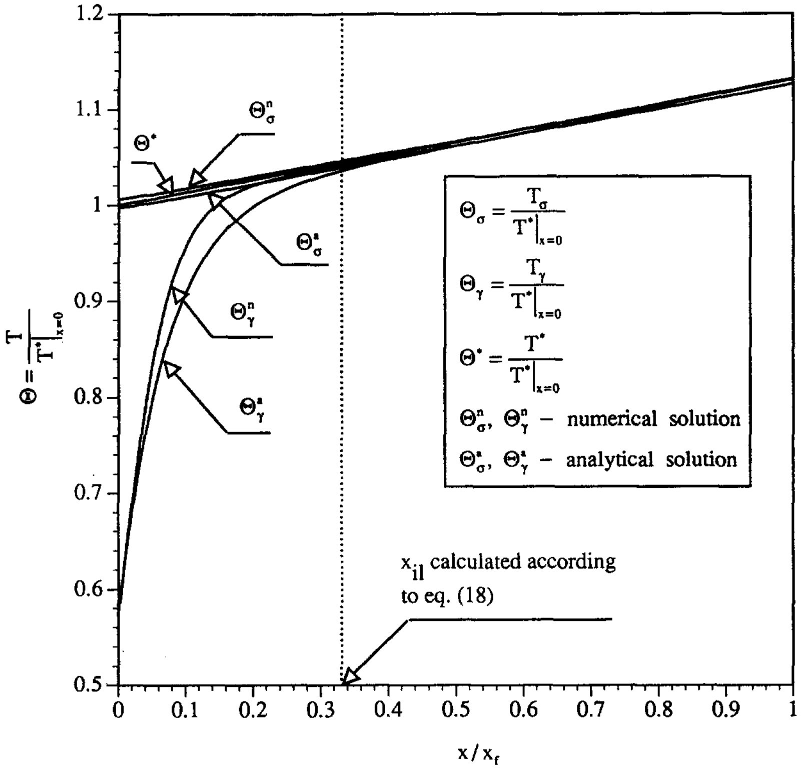


Fig. 3. Comparisons between numerical and analytical results. Temperature distributions in a metal hydride bed calculated numerically (Θ_γ^n and Θ_σ^n) and analytically (Θ_γ^a and Θ_σ^a), and also the temperature distribution Θ^* calculated under local thermal equilibrium assumption.

differs from zero over the whole region $0 < x < x_f$. Thus as soon as local thermal equilibrium is established, the solution is determined by the second term while the first term is responsible for non-thermal equilibrium effects.

The term 'inlet layer' is defined for this problem as a region next to the inlet boundary where non-thermal equilibrium effects are considerable. The length of the inlet layer x_{il} is estimated by determining the region over which the first term in equation (14) is significantly different than zero. This will result specification of x_{il} as

$$x_{il} = 5\frac{p_{in}}{T_{in}\beta_1}. \tag{15}$$

The local thermal equilibrium assumption can be used if the length of the inlet layer is considerably less than the length of the metal hydride bed, that is

$$\delta_1 = \frac{x_{il}}{L} = 5\frac{p_{in}\mathbf{v}_\gamma}{T_{in}L}\frac{M_\gamma(c_p)_\gamma}{\tilde{R}h_{\sigma\gamma}a_{\sigma\gamma}} \ll 1. \tag{16}$$

Expression (16) gives the criterion for the validity of the assumption VIII. To check the validity of equation (14) used in obtaining this criterion, the solution is compared against the numerical solution of equations (10) and (11). Figure 3 shows the dimensionless temperature distribution $\Theta = T/T^*|_{x=0}$ in a metal hydride bed calculated numerically using equations (10) and (11) (Θ_σ^n and Θ_γ^n), and using the approximate analytical solution (14) [Θ_σ^a has been calculated according to equation (14) and Θ_γ^a—according to equation (13) utilizing the Taylor series expansion which was mentioned earlier]. The temperature distribution Θ^* for the local thermal equilibrium case is also shown in that figure. Calculations have been carried out for the following values of parameters: $d = 3 \times 10^{-3}$ m (granules), $\varepsilon = 0.4$, $p_{in} = 5 \times 10^5$ Pa, $T_{in} = 200$ K, $x_f = 0.01$ m and $\lambda_\sigma \mathrm{d}T_\sigma/\mathrm{d}x|_{x=0} = 3.4 \times 10^3$ W m^{-2}. It can be seen that the approximate analytical solution (14) is close to the numerical solution, and that expression (15) gives a very good estimation for the width of the inlet layer. It is interesting to note that the local thermal equilibrium assumption introduces large errors only in calculating the gas phase temperature Θ_γ. But this approximation does not introduce any significant error when calculating the solid phase temperature Θ_σ. The reason for this behavior is that in a metal hydride bed conductive heat transfer in the solid phase far exceeds convective heat transfer in the gas phase. This result allows introduction of an additional assumption for hydrogen–metal hydride systems:

(XIV) convective heat transfer is negligibly small compared to conductive heat transfer in the metal hydride bed.

The heat transferred by hydrogen flow through a metal hydride bed can be estimated by

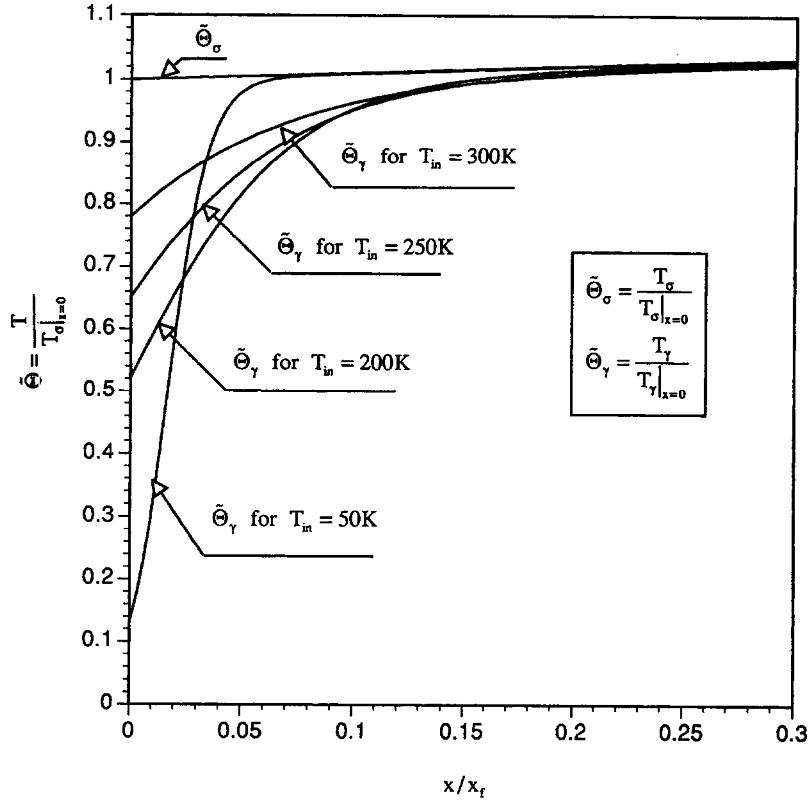


Fig. 4. Dependence of gas phase temperature distribution on inlet temperature of hydrogen T_{in}—numerical calculations.

$q_{conv} = \rho_\gamma (c_p)_\gamma \mathbf{v}_\gamma \Delta T_\gamma$, where ΔT_γ specifies the temperature difference between the hydration front and the inlet boundary. Consequently, assumption XIV can be estimated by the criterion

$$\delta_2 = \frac{q_{conv}}{q_{cond}} = \frac{\rho_\gamma (c_p)_\gamma \mathbf{v}_\gamma \Delta T_\gamma}{\lambda_\sigma \frac{\partial T_\sigma}{\partial x}} = \frac{(c_p)_\gamma M_\gamma \Delta T_\gamma}{\Delta h_{hyd}} \ll 1. \tag{17}$$

The last part of equation (17) is obtained using equation (14).

Figure 4 depicts the dependence of the dimensionless gas phase temperature

$$\tilde{\Theta}_\gamma = \frac{T_\gamma}{T_\sigma|_{x=0}}$$

on the inlet temperature of the gas, T_{in}. Values of the parameters used in these calculations are the same as the values used for Fig. 3. It can be seen that for relatively small deviations of T_{in} from $T_\sigma|_{x=0}$ (the curves for $T_{in} = 300$ K and $T_{in} = 250$ K) the width of the inlet layer increases as T_{in} decreases in accordance with equation (15). But further decrease of T_{in} (the curves for $T_{in} = 200$ K and $T_{in} = 50$ K) results in a decrease in the length of inlet layer. That is, equation (15) is no longer valid. This is expected since equations (14) and (15) are obtained based on a linear expansion. Therefore, equation (15) becomes inaccurate when T_γ/T_{in} is significantly different from 1.

3.2. *Criterion for the validity of the steady state approximation* (*assumption* IX)

To obtain a criterion for the steady state approximation, the scheme shown in Fig. 1(b) is utilized. The analytical solutions for the transient and steady state processes for the problem depicted in Fig. 5(a) are compared. The boundary $x = L$ is the hydrogen inlet boundary, at this boundary a constant hydrogen pressure $p(L, t) = p_b$ is maintained. Heat is removed from the boundary $x = 0$. At this boundary a constant temperature $T^*(0, t) = T_b$ is maintained. Due to assumption XIV the boundary $x = L$ can be considered to be insulated. Under the assumptions I–V, VII, VIII, X, XI, XIV mathematical formulation of the problem is

$$\frac{\partial T^*}{\partial t} = a_\sigma \frac{\partial^2 T^*}{\partial x^2} \quad 0 < x < x_f(t)$$

$$T^* = T_{eq}(p_b, t) \quad x_f(t) < x < L \tag{18}$$

where parameter a_σ is described in the nomenclature.

Equation (18) is obtained from equation (4). The following boundary and initial conditions are utilized

$$T^*(0,t) = T_b \quad T^*(x_f,t) = T_{eq}(p_b,\kappa_0) \tag{19a}$$

$$\lambda_\sigma \frac{\partial T^*}{\partial x}(x_f,t) = E_{hyd}\dot{x}_f(t) \quad \frac{\partial T^*}{\partial x}(L,t) = 0 \tag{19b}$$

$$\kappa(x,0) = \kappa_0 \quad T^*(x,0) = T_{eq}(p_b,\kappa_0). \tag{20}$$

Exact analytical solution of the problem posed by equations (18)–(20) is [20]:

$$x_f = \beta t^{1/2} \tag{21}$$

where β can be found from the following transcendental equation

$$\lambda_\sigma[T_{eq}(p_b,\kappa_0)-T_b]\exp\left(-\frac{\beta^2}{4a_\sigma}\right) = E_{hyd}\frac{\pi^{1/2}\beta}{2}a_\sigma^{1/2}\operatorname{erf}\left(\frac{\beta}{2a_\sigma^{1/2}}\right). \tag{22}$$

The steady state approximation leads to the same expression for the position of hydration front as the transient solution given by equation (21), however the constant β is different and is determined by the following expression:

$$\beta_{st.st.} = \left[\frac{2\lambda_\sigma}{E_{hyd}}(T_{eq}(p_b,\kappa_0)-T_b)\right]^{1/2} = \tilde{\beta}.$$

The steady state approximation is applicable if $\beta/\tilde{\beta} \approx 1$. After approximating the error function in equation (22) by an expansion the steady state approximation is found to be applicable if

$$\delta_3 = \frac{Ste}{6} = \frac{(c_p)_\sigma\rho_\sigma[T_{eq}(p_b,\kappa_0)-T_b]}{6E_{hyd}} \ll 1 \tag{23}$$

where $Ste = (c_p)_\sigma\rho_\sigma\Delta T/E_{hyd}$. Validity of the steady state approximation for the heat transfer process is then determined by the difference between temperature at the hydration front $T_f = T_{eq}(p_b,\kappa_0)$ and the temperature at the boundary of the metal hydride container T_b.

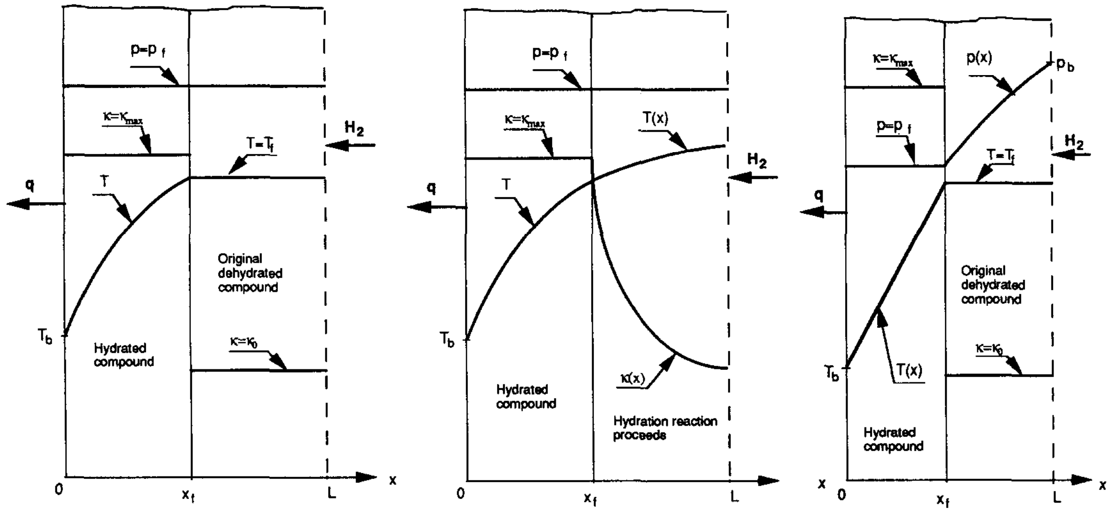
Physically, the Stefan number is the ratio of the specific heat capacity of metal hydride to the specific heat of the hydration reaction. Therefore, if the heat capacity is negligible compared to the heat of reaction, i.e. a small Stefan number, it is possible to assume that the temperature profiles at every instant are close to the steady state profiles. The analog of the Stefan number for the filtration process is the ratio of the mass of hydrogen which can be accumulated in the pores of the hydride matrix to the mass of hydrogen which can be released during dehydration. Therefore, the criterion for applying the steady state approximation for a filtration process in a metal hydride bed is given by

$$\delta_4 = \frac{2M_\sigma}{(1-\varepsilon)}\frac{1}{\rho_\sigma(\kappa_{max}-\kappa_0)}\frac{\varepsilon\Delta p}{\tilde{R}T} \ll 1. \tag{24}$$

The maximum pressure variation in metal hydride bed $(p_b-p_f)_{max}$ can be employed as the characteristic pressure difference, Δp, in equation (24).

3.3. *Criterion for the validity of the frontal model of hydration reaction* (*assumption* X)

Assumption X allows the localization of the chemical reaction at the hydration front. To check its applicability, the scheme shown in Fig. 1(b) is utilized. To



(a) (b) (c)

Fig. 5. (a) A schematic of temperature and hydrogen concentration distributions in a one-dimensional metal hydride layer under the assumptions I–VIII, X, XI, XIV. (b) A schematic of temperature and hydrogen concentration distributions in a one-dimensional metal hydride layer under the assumptions I–VIII, X′, XI, XIV. (c) A schematic of temperature and pressure distributions in the one-dimensional metal hydride layer under the assumptions I–X, XIV.

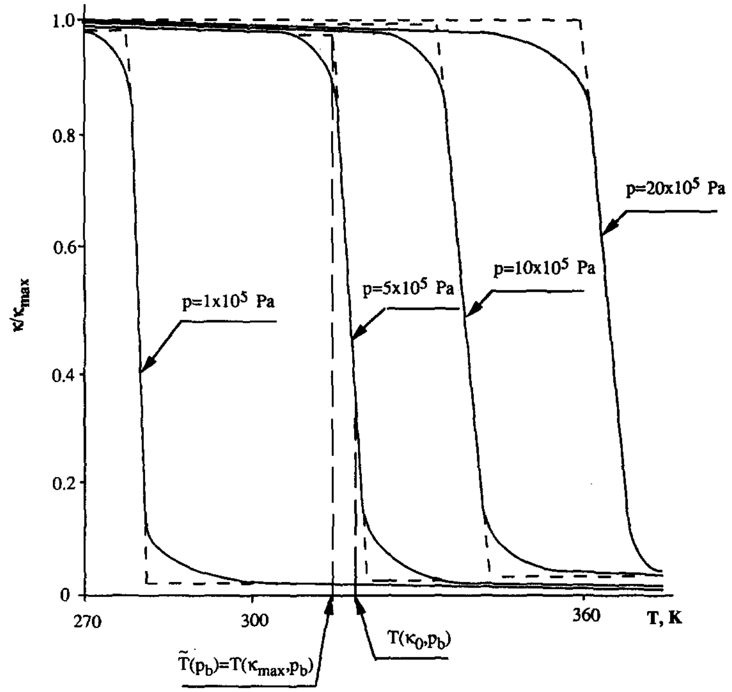


Fig. 6. Equilibrium isobars and their piecewise-linear approximation for $LaNi_5$–H_2 system. Solid line—real isobars, dotted line—piecewise-linear approximation of the equilibrium isobars.

analyze the validity of the frontal model, assumptions for the steady state criterion, i.e. I–V, VII, VIII, X, XI and XIV are employed. However, this time instead of assumption X, assumption X′ described below is utilized:

(X′) the equilibrium isobars are approximated not by a constant function, as in the frontal model but by a piecewise-linear model.

The equilibrium isobars and their piecewise-linear approximation for $LaNi_5-H_2$ system based on data reported in ref. [21] are depicted in Fig. 6. In our analysis the hysteresis (difference between absorption and desorption isobars) is neglected. In this case, the sorption reaction occurs not over the front but within a hydration zone as shown in Fig. 1(b). There is no sorption in the region $0 < x < x_f$, because $\kappa = \kappa_{max}$, however, in the region $x_f < x < L$ sorption proceeds [(Fig. (5b)]. The mathematical formulation of the modified problem is:

$$\frac{\partial T^*}{\partial t} = a_\sigma \frac{\partial^2 T^*}{\partial x^2} \quad 0 < x < x_f(t)$$

$$(c_p)_\sigma \rho_\sigma \frac{\partial T^*}{\partial t} = \lambda_\sigma \frac{\partial^2 T^*}{\partial x^2} + \frac{\Delta h_{hyd}}{2M_\sigma} \rho_\sigma \frac{d\kappa}{dT^*} \frac{\partial T^*}{\partial t}$$

$$x_f(t) < x < L.$$

The boundary and initial conditions for this problem are

$$T^*(0,t) = T_b \quad T^*(x_f,t) = \tilde{T}(p_b)$$

$$\frac{\partial T^*}{\partial x}(x_f^-,t) = \frac{\partial T^*}{\partial x}(x_f^+,t) \quad \frac{\partial T^*}{\partial x}(L,t) = 0$$

$$\kappa(x,0) = \kappa_0 \quad T(x,0) = T_{eq}(p_b,\kappa_0).$$

Due to a piecewise-linear approximation of the equilibrium isobars $d\kappa/dT^* = \text{const.}(< 0)$ for $x_f(t) < x < L$. This makes it possible to take into account the term describing heat of hydration reaction. Thus introducing the effective specific heat per unit volume,

$$C_{eff} = (c_p)_\sigma \rho_\sigma - \frac{\Delta h_{hyd}}{2M_\sigma} \rho_\sigma \frac{d\kappa}{dT^*} = (c_p)_\sigma \rho_\sigma + \frac{E_{hyd}}{\Delta T_{eq}}$$

allows for a finite zone of phase change (in this particular case, change from dehydrated to hydrated state). Therefore, using C_{eff} the governing equation for the region $x_f < x < L$ is recasted in the same form as that for the $0 < x < x_f$ region. Approximate analytical solution for this problem follows from the work described in ref. [22]:

$$x_f = \chi t^{1/2} \tag{25}$$

where χ is found from the transcendental equation

$$\frac{\exp\left(-\frac{\chi^2}{4a_\sigma}\right)}{\operatorname{erf}\left(\frac{\chi}{2a_\sigma^{1/2}}\right)} \left(\frac{(c_p)_\sigma \rho_\sigma}{C_{eff}}\right)^{1/2} \frac{\tilde{T}(p_b) - T_b}{T_{eq}(p_b,\kappa_0) - \tilde{T}(p_b)}$$

$$= \frac{\exp\left(-\frac{\chi^2}{4a_\sigma}\frac{C_{eff}}{(c_p)_\sigma \rho_\sigma}\right)}{\operatorname{erfc}\left(\frac{\chi}{2a_\sigma^{1/2}}\frac{C_{eff}^{1/2}}{[(c_p)_\sigma \rho_\sigma]^{1/2}}\right)}. \tag{26}$$

Equation (25) is of the same form as equation (21), but χ and β are found from different transcendental equations. The ratio β/χ characterizes the influence of nonzero width of the sorption zone on the dynamics of heat transfer processes. If it is close to unity, the sorption zone [the hydration zone shown in Fig. 1(b)] may be neglected and the frontal model can be applied. If β/χ is not close to unity, then it is necessary to take into account the real shape of the equilibrium isotherms by approximating them by a piecewise-linear approximation (as in assumption X′) or by some other non-constant function.

Expanding the error and complementary error functions in equations (22) and (26), the following expression for β/χ is obtained

$$\frac{\beta}{\chi} \approx 1+\frac{1}{2}\frac{\Delta T_{\mathrm{eq}}}{\tilde{T}(p_{\mathrm{b}})-T_{\mathrm{b}}}+\frac{1}{2}\frac{(c_{\mathrm{p}})_{\sigma}\rho_{\sigma}\Delta T_{\mathrm{eq}}}{E_{\mathrm{hyd}}}.$$

Thus $\beta/\chi \approx 1$ if

$$\delta_5 = \frac{1}{2}\frac{\Delta T_{\mathrm{eq}}}{\tilde{T}(p_{\mathrm{b}})-T_{\mathrm{b}}}+\frac{1}{2}\frac{(c_{\mathrm{p}})_{\sigma}\rho_{\sigma}\Delta T_{\mathrm{eq}}}{E_{\mathrm{hyd}}} \ll 1. \quad (27)$$

3.4. *Criterion for the validity of the filtration approximation* (*assumption* XI)

To obtain a criterion for determining the limits of the above approximation, the scheme shown in Fig. 1(b) is utilized. A one-dimensional hydration problem under assumptions I—V, VII—X, XIV is considered. The schematic diagram for the problem is depicted in Fig. 5(c). As was the case for previous models the boundary $x = L$ is the hydrogen inlet boundary. At this boundary a constant pressure of hydrogen $p(L, t) = p_{\mathrm{b}}$ is maintained. Heat is removed from the left-hand boundary, $x = 0$, where a constant temperature boundary condition $T^*(0, t) = T_{\mathrm{b}}$ is maintained. Based on assumption XIV, the right-hand boundary, $x = L$, can be considered to be insulated. Here, in contrast to the problems which were depicted in Fig. 5(a) and Fig. 5(b) hydrogen pressure variation between the inlet boundary and the hydration front is not neglected.

To derive the desired criterion, the problem is solved accounting for the pressure variation within the bed. To solve the problem the procedure suggested in refs. [7, 8] for metal hydride elements of cylindrical form is employed. For the problem under consideration the steady state filtration equation is

$$\frac{\mathrm{d}}{\mathrm{d}x}\left(p\frac{\mathrm{d}p}{\mathrm{d}x}\right) = 0$$

with boundary conditions $p(L) = p_{\mathrm{b}}$, $p(x_{\mathrm{f}}) = p_{\mathrm{f}}$. The above equation was obtained from equation (9) assuming constant temperature in the region $x_{\mathrm{f}} < x < L$. The pressure distribution in the layer is then simply found to be

$$p = \left[\frac{p_{\mathrm{b}}^2-p_{\mathrm{f}}^2}{L-x_{\mathrm{f}}}x+\left(p_{\mathrm{b}}^2-\frac{p_{\mathrm{b}}^2-p_{\mathrm{f}}^2}{L-x_{\mathrm{f}}}L\right)\right]^{1/2} \quad x_{\mathrm{f}} < x < L$$

$$p = p_{\mathrm{f}} \quad 0 < x < x_{\mathrm{f}}. \quad (28)$$

The hydrogen flux at the hydration front is obtained using equations (2) and (5):

$$j_{\mathrm{f}} = -\vartheta\left(\frac{p}{T^*}\frac{\partial p}{\partial x}\right)\bigg|_{x=x_{\mathrm{f}}} = -\frac{\vartheta}{2T_{\mathrm{f}}}\frac{p_{\mathrm{b}}^2-p_{\mathrm{f}}^2}{L-x_{\mathrm{f}}} \quad (29)$$

where

$$\vartheta = \Lambda\frac{\varepsilon^3}{\mu}\left(\frac{d}{1-\varepsilon}\right)^2\frac{M_{\gamma}}{\tilde{R}}.$$

On the other hand, hydrogen flux can also be determined by the hydration front velocity

$$j_{\mathrm{f}} = -\varsigma\dot{x}_{\mathrm{f}} \quad (30)$$

where $\varsigma = [(\kappa_{\max}-\kappa_0)/2M_{\sigma}]\rho_{\sigma}M_{\gamma}$.

Equating the expressions (29) and (30) leads to

$$\dot{x}_{\mathrm{f}} = \frac{\vartheta}{2\varsigma T_{\mathrm{f}}}\frac{p_{\mathrm{b}}^2-p_{\mathrm{f}}^2}{L-x_{\mathrm{f}}}. \quad (31)$$

A similar analysis for heat transfer process leads to the following temperature distribution in the bed

$$T^* = \frac{T_{\mathrm{f}}-T_{\mathrm{b}}}{x_{\mathrm{f}}}x+T_{\mathrm{b}} \quad 0 < x < x_{\mathrm{f}} \quad (32a)$$

$$T^* = T_{\mathrm{f}} \quad x_{\mathrm{f}} < x < L. \quad (32b)$$

The last expression, equation (32b), which determines the temperature distribution between the hydration front and hydrogen inlet boundary is required in order to justify the frontal model. This assumption will be checked after the condition for negligible pressure variation within the metal hydride layer is established. Repeating a similar procedure for the heat transfer process as that which has just been carried out for the filtration process results in the following equation for $\dot{x}_{\mathrm{f}}$:

$$\dot{x}_{\mathrm{f}} = \frac{\lambda_{\sigma}}{E_{\mathrm{hyd}}}\frac{T_{\mathrm{f}}-T_{\mathrm{b}}}{x_{\mathrm{f}}}. \quad (33)$$

The above expression was obtained by first using equation (32a) to obtain the heat flux at the hydration front and then equating the resulting expression with

$$\lambda_{\sigma}\frac{\partial T^*}{\partial x}(x_{\mathrm{f}}) = E_{\mathrm{hyd}}\dot{x}_{\mathrm{f}}$$

i.e. equation (19b). Equating expressions (31) and (33) results in the following relationship between temperature and pressure at the hydration front

$$p_{\mathrm{f}}^2 = p_{\mathrm{b}}^2-2T_{\mathrm{f}}(T_{\mathrm{f}}-T_{\mathrm{b}})\frac{\lambda_{\sigma}\varsigma}{E_{\mathrm{hyd}}\vartheta}\frac{L-x_{\mathrm{f}}}{x_{\mathrm{f}}}.$$

The desired criterion for assumption XI is then expressed in terms of the ratio $(p_{\mathrm{b}}^2-p_{\mathrm{f}}^2)/p_{\mathrm{b}}^2$ using the

Table 2. Pertinent criteria which are to be met for each of the referenced models for transport processes in a metal hydride bed

Model	Governing equations that were used	Analytical criteria and assumptions which have to be met
Fateev *et al.* [5]	Solid phase energy equation	(16), (17), (23), (24), (27), (34) + assumption VII
Sun and Deng [6]	Solid phase energy equation	(16), (17), (34) + assumption VII
Kuznetsov and Liventsov [7, 8]	Gas phase continuity equation and solid phase energy equation	(16), (17), (23), (24), (27), 36 + assumption VII
Artemenko *et al.* [9]	Gas phase continuity equation and solid phase energy equation	(16) + assumption VII
Choi and Mills [10, 11]	Gas phase continuity equation, solid phase energy equation and kinetic equation	(16)

equilibrium relationship between temperature and pressure in hydration zone

$$\delta_6 = \frac{\dfrac{B}{A-\ln(p_b/P_0)}\left(\dfrac{B}{A-\ln(p_b/P_0)} - T_b\right)}{p_b^2}\frac{\lambda_\sigma}{E_{\text{hyd}}} \times \frac{(\kappa_{\max}-\kappa_0)\rho'_\sigma}{\Lambda M_\sigma}\mu\tilde{R}\frac{(1-\varepsilon)^2}{\varepsilon^3}\left(\frac{1}{d}\right)^2 \ll 1. \quad (34)$$

The above equation was obtained by omitting the term $(L-x_f)/x_f$ since it is of the order of 1 for most of the spatial location. The criterion given by equation (34) shows that the filtration approximation gives the best results for higher porosities and larger particle diameter of the metal hydride bed.

Now the condition given by equation (32b) can be checked. This condition would be valid if the temperature gradient to the right of hydration front is considerably less than the temperature gradient to the left of hydration front, that is if

$$\frac{\left.\dfrac{\partial T^*}{\partial x}\right|_{x_f^+}}{\left.\dfrac{\partial T^*}{\partial x}\right|_{x_f^-}} = \frac{\left.\dfrac{\partial T^*}{\partial x}\right|_{x_f^+}}{\dfrac{T_f-T_b}{x_f}} \ll 1 \quad (35)$$

where equation (32a) has been used to determine $\partial T^*/\partial x|_{x_f^-}$ in equation (35). To calculate $\partial T^*/\partial x|_{x_f^+}$ the pressure distribution given by equation (28) along with the equilibrium relationship between temperature and pressure for the hydration region namely, $p = P_0 \exp[A-(B/T)]$, are used. This leads to

$$\left.\frac{\partial T^*}{\partial x}\right|_{x_f^+} = \frac{B}{[A-\ln(p_f/P_0)]^2}\frac{p_b^2-p_f^2}{2p_f^2}\frac{1}{L-x_f}$$

consequently

$$\frac{\left.\dfrac{\partial T^*}{\partial x}\right|_{x_f^+}}{\left.\dfrac{\partial T^*}{\partial x}\right|_{x_f^-}} = \frac{x_f}{L-x_f}\frac{T_f^2}{2B(T_f-T_b)}\frac{p_b^2-p_f^2}{p_f^2}.$$

Thus the condition given by equation (32b) is satisfied when

$$\delta_7 = \frac{T_f^2}{2B(T_f-T_b)}\frac{p_b^2-p_f^2}{p_f^2} \ll 1. \quad (36)$$

Thus the criterion given by equation (36) is satisfied if variation of hydrogen pressure within the metal hydride layer is small. In other words, the criterion given by equation (36) follows from the criterion (34). But the criterion given by equation (36) also has its own merits for problems for which hydrogen inlet boundary does not coincide with the heat removal boundary [as for the problem depicted in Fig. 1(b)]. For such problems the criterion given by equation (36) determines the possibility of calculating filtration processes using the frontal model. From equation (36) it is easy to see that the frontal model is applicable to problems of this type if temperature variation within the bed is relatively large (intensive processes) and pressure variation is relatively small (high porosity and relatively large metal hydride particles).

4. CONCLUSIONS

Analysis carried out in this work establishes useful and practical criteria for several frequently used assumptions. Specifically, criterion for the validity of local thermal equilibrium, criterion for the validity of steady state approximation, criterion for the validity of the frontal model and criterion for the validity of the filtration approximation are established analytically. The above-referenced set of analytical criteria provides a vehicle for systematic determination of conditions under which previous models are better suited for. Table 1 summarizes all the pertinent assumptions and the analytical criteria for the validity of these assumptions and Table 2 summarizes various pertinent models and criteria under which they are best suited for.

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