



Niche applications of metal hydrides and related thermal management issues



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ABSTRACT

This short review highlights and discusses the recent developments and thermal management issues related to metal hydride (MH) systems for hydrogen storage, hydrogen compression and heat management (refrigeration, pump and upgrade, etc.). Special attention is paid to aligning the system features with the requirements of the specific application. The considered system features include the MH material, the MH bed on the basis of its corresponding MH container, as well as the layout of the integrated system.

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1. Introduction

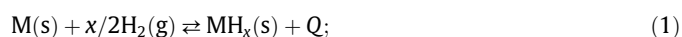
Metal hydride (MH) materials are able to reversibly absorb and desorb large amounts of hydrogen in a wide range of temperatures and pressures. They are characterised by some unique properties, including extremely high volume density of H atoms incorporated in the metal matrix [1–5], wide “tuneability” of thermodynamic performances [3–7] and fast kinetics of hydrogenation/dehydrogenation [3–9]. MH technologies utilising such features are typical application-driven niche areas able to provide very efficient solutions for hydrogen handling, particularly for end-use applications, by the tuning of component and phase composition of the parent MH material, system layout and operation parameters. MHs offer several advantages over traditional hydrogen storage (e.g. compressed hydrogen, liquefied hydrogen, etc.) and processing systems, such as compactness, safety (due to low pressures <10 bar), possibilities of hydrogen supply at pre-defined pressures, absence of moving parts, long operation lifetime, efficient and technologically flexible energy conversion between low-grade heat and the energy of pressurised H₂ [7,10]. These systems have found a number of promising gas phase applications including hydrogen storage and supply, hydrogen compression, heat management (refrigeration, pump, and upgrade), etc. [1–10].

The present work herein briefly highlights features of the most important gas-phase niche applications of MH. Methods for

improving heat transfer performances of the corresponding MH reactors are also described and discussed.

2. Thermodynamic properties of MH and main applications

The chemical bonding of gaseous hydrogen in MH can be written as:



where M is the metal, alloy or intermetallic compound forming hydride, MH_x, (s) and (g) denote solid and gas phase, respectively.

The direct process of reaction (1), hydride formation/hydrogen absorption is an exothermic reaction, and the generated heat, Q, is approximately equal to the absolute value of the enthalpy of reaction (1). The heat has to be effectively removed in order to achieve the desired H₂ charge rate. Accordingly, the endothermic hydride decomposition/H₂ desorption (reverse process of reaction (1)) needs efficient supply of the approximately same amount of the heat to provide the necessary rates of H₂ discharge.

It has to be noted that the driving force of both direct and reverse processes of reaction (1) strongly depends upon the deviation of the actual hydrogen pressure, P, from the value, P_{eq}, of equilibrium dissociation pressure of the MH which is an important property of hydride forming materials. Temperature dependence of P_{eq} in the plateau region of a Pressure–Composition Isotherm (PCI) is expressed by the van't Hoff equation:

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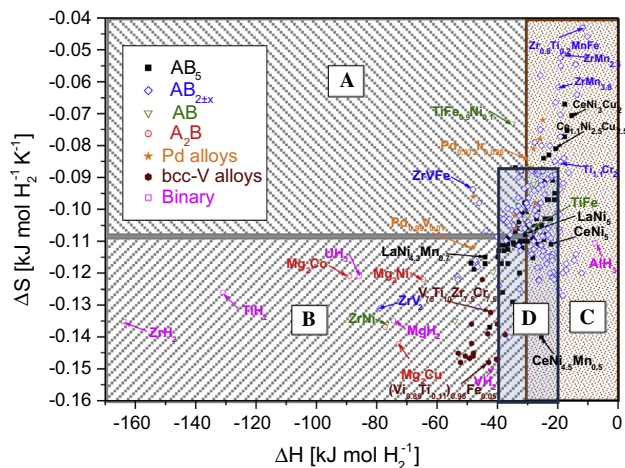


Fig. 1. Hydrogenation enthalpies and entropies of various hydride materials and their suitable applications: (A) heat pumps, (B) heat storage, (C) hydrogen storage, (D) hydrogen compression.

$$\ln \frac{P_{eq}}{P_0} = -\frac{\Delta S}{R_u} + \frac{\Delta H}{R_u T}; \quad (2)$$

where T [K] is the temperature, R_u [$\text{J mol}^{-1} \text{K}^{-1}$] is the universal gas constant, ΔH [J mol H_2^{-1}] and ΔS [$\text{J mol H}_2^{-1} \text{K}^{-1}$] represent the associated changes in enthalpy and entropy, and P_0 is the reference pressure (1 atm).¹

Fig. 1 shows ΔS and ΔH values for 278 metal hydride materials [7,10]. For the most of the MH materials, ΔS does not vary significantly around $-111 \text{ J mol}^{-1} \text{H}_2 \text{K}^{-1}$ which is close to the change of entropy of gaseous H_2 for the reaction (1), of $-130 \text{ J mol}^{-1} \text{H}_2 \text{K}^{-1}$. The plateau pressure, P_{eq} , is mainly determined by the reaction enthalpy, ΔH , which varies from -166 to $-6.6 \text{ kJ mol}^{-1} \text{H}_2$ for ZrH_2 [11,12] and AlH_3 [13], respectively. At the same time, in some MH materials the contribution of the entropy of the solid into the total entropy change for reaction (1) can be significant thus resulting in significant deviations of ΔS from the average value specified above. Binary metal hydrides and solid solution alloys exhibit lower ΔS , down to $-150 \text{ J mol}^{-1} \text{H}_2 \text{K}^{-1}$, while multicomponent $\text{AB}_{2\pm x}$ metal hydrides can have hydrogenation entropy as high as $-45 \text{ J mol}^{-1} \text{H}_2 \text{K}^{-1}$.

Fig. 1 can guide in selecting metal hydrides for different gas phase applications including:

A – Heat pumps: for efficient operation, the MH material should have high heat effects ($|\Delta H| > 30 \text{ kJ mol}^{-1} \text{H}_2$) while providing moderate H_2 pressures during the operation. The absolute value of the reaction entropy should be as low as possible ($|\Delta S| < 110 \text{ J mol}^{-1} \text{H}_2 \text{K}^{-1}$).

B – Heat storage: similarly, the MH material has to have high heat effects ($|\Delta H| > 30 \text{ kJ mol}^{-1} \text{H}_2$), but highly exothermic MH should have not too low H_2 pressures at operating temperatures ($|\Delta S| > 110 \text{ J mol}^{-1} \text{H}_2 \text{K}^{-1}$).

C – H_2 storage: for this type of application, low energy consumption for hydrogen release is important ($|\Delta H| < 30 \text{ kJ mol}^{-1} \text{H}_2$).

D – H_2 compression: this application requires compromise between energy consumption for hydrogen desorption and high compression ratio; accordingly the MH materials should have medium heat effects ($|\Delta H| = 20\text{--}30 \text{ kJ mol}^{-1} \text{H}_2$) and high absolute values of the hydrogenation entropy ($|\Delta S| > 100 \text{ J mol}^{-1} \text{H}_2 \text{K}^{-1}$).

3. Thermal management issues

3.1. General features

When the rates of heat dissipation (exothermic H_2 absorption) or heat supply (endothermic H_2 desorption) from/to the MH material are lower than the rates of its heating/cooling due to heat effect, ΔH , of reaction (1), the material reaches an equilibrium temperature, T_{eq} , when $P = P_{eq}$, and no further absorption or desorption occurs. The value of T_{eq} can be estimated by a solution of Eq. (2) with respect to the temperature for the each actual pressure (“dynamic van’t Hoff equation”) as it was suggested by Goodell [14]. Thus, further rates of hydrogen absorption/desorption in any part of the MH material in a reactor will be limited by the rate of heat exchange between the material (elementary volume in MH bed) at $T = T_{eq}$ and the cooling/heating means at $T = T_0$. Accordingly, the integral H_2 absorption/desorption rates in a MH reactor can be determined only in the course of the solving of, in 3D statement, the Fourier heat equation with a source term [3]:

$$\frac{\partial T}{\partial t} = \nabla \cdot \left(\frac{k_e}{\rho c_p} \nabla T \right) \pm \frac{\Delta H}{\rho c_p}; \quad (3)$$

where T [K] is the MH temperature, t [s] is time, k_e [$\text{W m}^{-1} \text{K}^{-1}$] is the effective thermal conductivity, ρ [kg m^{-3}] is the apparent specific weight and c_p [$\text{J kg}^{-1} \text{K}^{-1}$] is the specific heat capacity of the packed material, “+” and “−” correspond to H_2 desorption and absorption, respectively.

This approach has been taken in numerous investigations for the modelling of heat-and-mass transfer in MH beds [15–17]. At the same time, the details of the task statement strongly depend upon a number of factors, first of all, effective heat conductivity of the MH bed, its geometry, ways of heat supply and its removal. These parameters related to heat management issues may significantly differ in numerous applications. However, the common strategy in the improvement of heat transfer performances of MH beds should include the “maximizing” of the term $\frac{k_e}{\rho c_p}$, first of all, by increasing the effective thermal conductivity, k_e . The shortening of the characteristic heat transfer distance is also very important.

3.2. Augmentation of hydride bed heat transfer

Enhancing the effective thermal conductivity forms the central objective of all techniques applied to porous MH beds. Impregnations of heat conduction matrices have been found to be very useful. The approach includes 2D and 3D structures made of heat conductive metals, e.g. copper or aluminium. Common impediments to the above solution include the parasitic weight of the heat conductive material, disintegration and deterioration upon cycling, and high cost. Representative values of effective thermal conductivity and void fraction achieved by different augmentation techniques are listed in Table 1.

Suda et al. [24] carried out experimental investigations aimed at improving the effective thermal conductivity of activated MH. A 3D structure of porous aluminium foam increases k_e in 9–10 times ($4 \text{ W m}^{-1} \text{K}^{-1}$ at 0.5 MPa H_2) as compared to the unmodified hydride bed. Laurencelle and Goyette [19] numerically studied the impact of aluminium foam to enhance hydrogenation reactions. The effective thermal conductivity of the hydride bed was improved to $10 \text{ W m}^{-1} \text{K}^{-1}$. The reaction rate in the reactor with foam depends upon the resistance inside the foam cells, and also along the path to the reactor wall. With foam, the MH bed thickness could be increased without hampering the charging time. The foams with pore density less than 20 PPI (Pores Per Inch) were not recommended due to poor heat transfer. Fleming et al. [25] showed that

¹ Since left-hand side of Eq. (2) contains the ratio P_{eq}/P_0 , their values can be taken in any but the same units.

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