Available online at www.sciencedirect.com
SCIENCE DIRECT
International Journal of Hydrogen Energy 31 (2006) 737–751
International Journal of HYDROGEN ENERGY
www.elsevier.com/locate/ijhydene

On the optimization of hydrogen storage in metal hydride beds

Eustathios S. Kikkinides[a, b, *], Michael C. Georgiadis[c, d], Athanassios K. Stubos[e]

[a] *Department of Engineering and Management of Energy Resources, University of Western Macedonia, Kastorias and Fleming Str., 50100 Kozani, Greece*
[b] *Chemical Process Engineering Research Institute, Centre for Research and Technology Hellas, P.O. Box 361, 57001 Thermi-Thessaloniki, Greece*
[c] *Process Systems Enterprise Limited, Academic Relations, Thermi Business Incubator, 9th km Thessaloniki-Thermi, P.O. Box 60053, Thermi-Thessaloniki 57001, Greece*
[d] *Centre for Process Systems Engineering, Department of Chemical Engineering, Imperial College London, London SW7 2AZ, UK*
[e] *National Center for Scientific Research "DEMOKRITOS", Institute of Nuclear Technology and Radiation Protection, 15310 Ag. Paraskevi Attikis, Athens, Greece*

Available online 9 August 2005

Abstract

This work presents a novel systematic approach for the optimal design and control of metal hydride beds used for hydrogen storage. A detailed 2-D mathematical model is developed and validated against experimental and theoretical literature results. Based on recent advances in dynamic optimization, the objective is then to find the optimal process design (e.g. cooling systems design) and operating strategy (e.g. cooling fluid profile over time, hydrogen charging profile, etc.) so as to minimize the storing time, while satisfying, a number of operating constraints. Such constraints account for pressure drop limitations, cooling fluid availability and maximum tank temperature. Optimization results indicate that almost 60% improvement of the storage time can be achieved, over the case where the system is not optimized, for a minimum storage capacity of 99% of the total bed capacity. Trade-offs between various objectives, alternative design options and optimal cooling control policies are systematically revealed illustrating the potential offered by modern optimization techniques.

1. Introduction

Environmental problems related to the emission of greenhouse-effect gases and to the depletion of fossil-fuel natural resources, have led to significant research effort on alternative and cleaner fuels. The simultaneous growth of the world population and of air-pollutant emissions produced by carbonaceous fuels, impose the replacement of gasoline by cleaner, renewable fuels such as hydrogen, which is especially attractive for electric vehicle use [1,2]. The need for a worldwide conversion from fossil fuels to hydrogen requires the elimination of several barriers imposed along the different steps involved in hydrogen technology. One of the main problems in large usage of hydrogen energy in automotive industry is the storage problem. Conventional storage methods such as gas compression and liquifying are impractical since the former requires very heavy gas tanks and the latter is too expensive to be employed in public vehicles. Storing hydrogen in metal hydrides beds as a chemical compound appears to be a promising, cost-effective and safe method of hydrogen storage in the near future [3]. This can be attributed to the reduced operating pressure compared to compress gas technology, thus ensuring less weight and better security.

* Corresponding author. Department of Engineering and Management of Energy Resources, University of Western Macedonia, Kastorias and Fleming Str., 50100 Kozani, Greece. Tel.: +30 2310 498116; fax: +30 2310 498131.
E-mail address: kikki@cperi.certh.gr (E.S. Kikkinides).

0360-3199/$30.00
doi:10.1016/j.ijhydene.2005.06.021

Nomenclature

Bi_z	dimensionless axial Biot number
Bi_r	dimensionless radial Biot number
HM*	dimensionless hydrogen to metal atomic ratio
k_z	dimensionless axial permeability
k_r	dimensionless radial permeability
L	total bed length
Le	dimensionless Lewis number
M_s	molecular weight of the metal hydride
M_t	total amount of hydrogen stored in the tank
P	pressure
P_0	pressure at the bed inlet
$Pe_{t,z}$	dimensionless axial thermal Peclet number
$Pe_{t,r}$	dimensionless radial thermal Peclet number
$Pe_{m,z}$	dimensionless axial Peclet number
$Pe_{m,r}$	dimensionless radial Peclet number
$\hat{P}_{eq}$	dimensionless equilibrium pressure
r	dimensionless radial distance
R	total bed radius
t_{res}	residence time
T_0	temperature at the bed inlet
$\hat{u}$	superficial gas velocity vector
u_0	reference velocity
u_f	cooling fluid velocity
W_0	dimensionless parameter
z	dimensionless axial distance

Greek letters

β	dimensionless parameter
δr	radial thickness
ε	void fraction
ε_a	dimensionless activation energy
$\hat{\rho}$	dimensionless hydrogen density in the gas phase
ρ_0	hydrogen density at the bend inlet
ρ_s	dimensionless solid density
$\hat{\rho}_{sat}$	"saturated" bed density
τ	dimensionless time
θ	dimensionless gas temperature
θ_c	dimensionless constant cooling temperature
θ_f	dimensionless temperature of the cooling fluid

The mathematical modeling of hydrogen storage in metal hydride beds has received considerable attention over the past ten years. Jemni and Nasrallah [4] presented a theoretical study of the mass and heat transfer dynamics in a metal hydride reactor. The effect of several parameters on the overall hydrogen storage efficiency was illustrated. Some of the assumptions used in this work were validated in a subsequent contribution [5]. Jemni et al. [6], presented an experimental approach to determine the reaction kinetics, equilibrium conditions and transport properties in $LaNi_5$–H_2 system. Heat and mass transfer effects were also determined experimentally. Experimental data were compared with model predictions from previous work. Nakagawa et al. [7], presented a 2-D model for the transient heat and mass transfer within a metal hydride bed. Several conclusions were made regarding the effect of the underlying operating condition on the storing process. Mat and Kaplan [8] developed a mathematical model to describe hydrogen absorption in a porous lanthanum metal bed. The model takes into account the complex mass and heat transfer and reaction kinetics. Model predictions were found to be in good agreement with experimental data. Aldas et al. [9] and Mat et al. [10], presented an integrated model of heat and mass transfer, reaction kinetics and fluid dynamics in a hydride bed. An important conclusion of this work is that the flow conditions do not significantly affect the amount of hydrogen absorbed. Askri et al. [11] extended the previous studies [4–6], by investigating the effect of radiate heat transfer in $LaNi_5$–H_2 and Mg–H_2 hydride beds. Simulation results showed that radiation effects are negligible on the sorption process in the $LaNi_5$–H_2 bed but they play an important role in the case of Mg–H_2 hydride bed. Recently, Kaplan and Veziroglu [3] extended the work of Mat et al. [10] and investigated numerically the hydrogen storage process in a 2-D metal hydride bed including the full momentum balance equation, which is considered to be important when large pressure gradients exist in the system. The effect of the underlying operating conditions on the hydride formation rate was investigated and several conclusions were drawn up.

The operation of hydrogen storage tank using metal hydrides presents distinct challenges such as the possible appearance of a maximum in the temperature profile (hot spot) and the possibility of temperature runaway. The occurrence of excessive temperatures (often due to parametric sensitivity) can obviously have detrimental consequences on the storage management of the tank such as potential explosions, limited storage capacity, etc. These considerations motivate the need for effective heat management strategies for such systems and novel design options (e.g. design of cooling systems, tank diameter, etc.). In this vein, control strategies that regulate the magnitude of the hot spot temperature, while ensuring a maximum storage capacity at minimum total costs are of paramount important. Research indicates that attempts to address this problem directly and systematically are rather rare.

This work first presents a detailed 2-D mathematical model describing the adsorption of hydrogen in a metal hydride bed. Heat, mass and momentum transfer effects are modeled in detail. The model is validated and found to be in excellent agreement with theoretical and experimental data from the literature [4–6]. Then, a cooling medium