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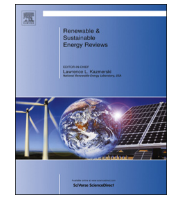

Solid state storage of hydrogen and its isotopes: An engineering overview

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A R T I C L E I N F O

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A B S T R A C T

Solid state storage of hydrogen in the form of a reversible metal or alloy hydride has been proven to be a very effective and compact way of storing hydrogen and its isotopes for both stationary and mobile applications. Other than metal based systems, a wide variety of materials have been studied for this purpose and their thermodynamic properties, storage capacity, etc. have been determined. Heat transfer issues form an important consideration for the engineering design of a metal hydride based hydrogen storage system, hence several kinds of storage beds have been fabricated and their performance analyzed. The kinetics and mechanism of these hydriding processes for various types of storage materials have also attracted a great deal of interest. This work summarizes some of the information available on solid state storage of hydrogen isotopes which is essential for the engineering design of a storage system. The focus is on the engineering and technical issues and the practical considerations pertinent to the design and operation of such storage systems for various applications.
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1. Introduction

Hydrogen is often described as being the fuel of the future [1]. For widespread use of hydrogen in place of the more traditional fossil fuels, it is imperative to have very efficient storage and transportation systems for it. Use of metal getter beds is one of

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Nomenclature

A,B	Van't Hoff constants for equilibrium temperature–pressure relation, dimensionless and K^{-1} respectively
A_h	convective heat transfer area available in the bed, m^2
c	density of solid at any time, $kg\ m^{-3}$
C_{Pg}	specific heat capacity of the gas, $J\ kg^{-1}\ K^{-1}$
C_{Ps}	specific heat capacity of the solid, $J\ kg^{-1}\ K^{-1}$
c_{SS}	density of fully hydrided or saturated solid, $kg\ m^{-3}$
d_p	diameter of catalyst, m
E_a	activation energy of the hydriding reaction, kJ/mol
E_d	activation energy of the dehydriding reaction, kJ/mol
h_{sg}	convective heat transfer coefficient from hydrogen to solid phase, $W\ m^{-2}\ K^{-1}$
H	enthalpy of hydriding reaction, $J\ mol^{-1}\ K^{-1}$
k_a	frequency factor for hydriding reaction, s^{-1}
k_d	frequency factor for dehydriding reaction, s^{-1}
k_e	effective thermal conductivity of the porous solid, $W\ m^{-1}\ K^{-1}$
k_g	thermal conductivity of hydrogen, $W\ m^{-1}\ K^{-1}$
k_s	thermal conductivity of the metal/metal hydride, $W\ m^{-1}\ K^{-1}$
K	permeability, m^2
$\dot{m}$	reaction rate, $kg\ m^{-3}\ s^{-1}$
M_g	molecular weight of hydrogen (or its isotope), $gm\ mol^{-1}$
m_s	mass of solid, kg
P	hydrogen pressure, Pa
P_{eq}	equilibrium pressure of hydrogen over metal hydride, Pa
R	universal gas constant, 8.314 $J\ mol^{-1}\ K^{-1}$
t	time, s
T_g	absolute gas temperature, K
T_S	absolute solid temperature, K
v_g	superficial velocity of gas in the porous bed, $m\ s^{-1}$
V_S	volume of solid in the getter bed, m^3
$\in$	void fraction in the hydride bed, dimensionless
μ	gas viscosity, Pa s
$(\rho C_p)_e$	effective specific heat capacity of the hydrogen–metal hydride system, $J\ kg^{-1}\ K^{-1}$
ρ_s	solid density, $kg\ m^{-3}$
ρ_g	gas density, $kg\ m^{-3}$

the most viable options for both short and long term storages and handling of hydrogen and its isotopes. This applies to stationary systems like laboratories, fusion energy research centers as well as mobile systems like automobiles, mining vehicles, and submarines and so on. The adsorbed hydrogen (or its isotopes D, T) on a metal or alloy M forms MH_3 (or MD_3 or MT_3) by chemisorption. MH_3 is dissociated by heating and hydrogen is liberated from the solid phase. On cooling, the metal or alloy quickly reabsorbs the hydrogen gas [2]. This reversible liberation and uptake of hydrogen or deuterium or tritium from getter beds can be performed many times under appropriate conditions depending on the type of material chosen without loss of efficiency. These properties of the metal hydrides or metal tritides make them excellent hydrogen/tritium storage and pumping materials. The major advantage of this route of hydrogen storage is that a large amount of gas can be stored in a very small volume when compared to traditional hydrogen storage methods involving compressed gas cylinders or cryogenic storage of liquid hydrogen, and once hydrogen is recovered from the adsorbent bed, it is of a very high purity as well [3].

Despite all the advantages mentioned above the actual design of a solid state hydrogen storage system presents several engineering challenges mainly in the form of heat transfer issues, poor chemical kinetics and the formation of possible explosive reaction mixtures with oxygen and moisture, low gravimetric storage capacity of many materials, powder formation and volume expansion of the storage material during hydriding, fatigue based deformation of the bed under cyclic pressure changes and thermal loads, as well as safety issues on account of the toxic and pyrophoric nature of many storage materials. No one material offers the best set of thermodynamic and kinetic properties, but it is often possible to engineer the storage system in such a way that an optimal design is obtained for a chosen storage material. It must be mentioned that the choice of materials as well as the ultimate system design is mainly governed by the intended application, whether it is onboard storage for vehicular applications which must necessarily be more compact or land based tritium storage and delivery system where the space constraint often arises from the need to handle all tritium based systems inside fume hoods or glove boxes. Thus different system designs have been proposed and they have their own merits and demerits.

This paper presents a brief overview about various kinds of hydrogen storage materials developed and studied, with greater focus on the design considerations for the storage vessels, the heat and mass transfer aspects of storage and the computational models developed to study the behavior of these beds. Most of the practical applications have been based on metal based systems and this review places emphasis on the various issues pertinent to such systems.

2. Classes of materials for hydrogen storage

The desirable properties of a storage material for hydrogen are generally stated to be high gravimetric and volumetric capacity, reversibility of hydriding and dehydriding steps, favorable equilibrium temperature–pressure characteristics, adequate stability of the hydride formed and low sensitivity to impurities present in feed gas. All the desired properties are yet to be found in one single material even after decades of research in this field. Researchers have examined several classes of materials for solid state hydrogen storage. The two major routes by which hydrogen is immobilized in a solid matrix are (i) the physical adsorption of hydrogen on the storage material or (ii) the dissociative chemisorption of hydrogen gas and diffusion of atomic hydrogen in the solid matrix, under appropriate conditions of temperature and pressure. The major groups of materials are briefly reviewed and compared in the following sections. More detailed and comprehensive reviews of the materials aspects have already been published, some of the more recent ones being the one by Dalebrook et al. [37], by Durbin et al. [3] and Lototskyy et al. [151].

2.1. Heavy metals and their alloys

Storage of hydrogen isotopes as a metal hydride, deuteride or tritide is one of the most common methods adopted in laboratories and especially in tritium handling facilities the world over [4,5]. Several heavy metals especially transition metals and rare earths and more commonly their binary, ternary and more complex alloys have been studied and their thermodynamic and kinetic behavior evaluated [6–9]. These alloys are generally represented as AB, AB_5

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