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# Numerical study on a two-stage Metal Hydride Hydrogen Compression system

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ABSTRACT

A multistage Metal Hydride Hydrogen Compression (MHHC) system uses a combination of hydride materials in order to increase the total compression ratio, whilst maximizing the hydrogenation rate from the supply pressure at each stage. By solving the coupled heat, mass and momentum conservation equations simultaneously the performance of a MHHC system can be predicted. In the current work a numerical model is proposed to describe the operation of a complete compression cycle. Four different MHHC systems are examined in terms of maximum compression ratio, cycle time and energy consumption and it was found that the maximum compression ratio achieved was 22:1 when operating $LaNi_5$ ($AB_5$-type) and a Zr–V–Mn–Nb ($AB_2$-type intermetallic) as the first and second stage alloys respectively in the temperature range of 20 °C (hydrogenation) to 130 °C (dehydrogenation).

## 1. Introduction

Hydrogen compression based on the reversible hydrogenation/dehydrogenation ability of metal hydrides has been investigated as a reliable process to compress hydrogen to high pressure without contamination and with low energy costs [1,2]. A multistage Metal Hydride Hydrogen Compression (MHHC) system uses a combination of different metal hydrides to increase the final compression ratio while maximizing the hydrogenation process from both the supply pressure of each stage. Over the last decade a large number of scientists have made efforts in the subject of MHHC systems and some promising results regarding the material selection and the compression conditions were found both experimentally [3–9] and numerically [10–13]. In most cases, three combinations of materials are selected for a two-stage compression system. The first combination is based on two $AB_5$-type, usually $LaNi_5$ as the first stage and a $MmNi_{5-y}$–$X_y$ alloy as the second stage yielding a pressure ratio 12:1 when the compressor operates between 20 and 90 °C [14]. The second case uses an $AB_5$ material ($LaNi_5$, Ce [15 at.%] + La [Balance]) for the stage 1 and an $AB_2$ (Ti–Zr–Mn) for the stage 2 [15]. The third typical combination involves two $AB_2$ intermetallics for stage 1 and stage 2, respectively [16].

The multistage operation approach introduces strict requirements to the tune-ability of the Pressure–Composition-Isotherm (PCI) characteristics, because the coupling of the first stage (dehydrogenation) and the second stage (hydrogenation) requires the plateau pressure ($P_{eq}$) for the stage 1 metal hydride to be higher than that for stage 2 as shown in Fig.1. Other requirements include: fast kinetics, reduction of compression cycle time; reversibility, high storage capacity to reduce the amount of hydride needed; low plateau slope for the isotherms and low hysteresis. Finally, the cost of the compression process should be affordable [17].

In the current work, a numerical study of a two-stage MHHC is presented. The proposed model was validated with experimental results extracted from a lab scale Sievert-type apparatus and the comparison showed good agreement between the experimental and numerical results. Four different MHHC systems were examined, by using different combinations of materials for the first and second stages, in terms of maximum compression ratio, cycle time and system energy consumption.

## 2. Model formulation and problem definition

### 2.1. Introduction of a two-stage MHHC cycle

Fig. 1 illustrates the two-stage compression cycle on a van't Hoff plot, where it is assumed that the temperature range for the stage 1 and stage 2 hydride beds is the same and moves from a low

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**Nomenclature**

| | |
|---|---|
| $C_a$ | hydrogenation constant ($s^{-1}$) |
| $C_d$ | dehydrogenation constant ($s^{-1}$) |
| $C_p$ | specific heat (J/kg K) |
| $E$ | activation energy (J/mol$H_2$) |
| $h$ | heat transfer coefficient (W/$m^2$ K) |
| $k$ | thermal conductivity (W/m K) |
| $K$ | permeability ($m^2$) |
| $M$ | molecular weight (kg/mol) |
| $m$ | kinetic expression |
| $t$ | time (s) |
| $T$ | temperature (K) |
| $v$ | gas velocity (m/s) |
| $P$ | pressure (bar) |
| $R$ | gas global constant (J/mol K) |

*Subscripts*

| | |
|---|---|
| $a$ | hydrogenation |
| $d$ | dehydrogenation |
| $e$ | effective |
| $eq$ | equilibrium |
| $g$ | gas |
| $s$ | solid |
| $ss$ | saturation |

*Greek letters*

| | |
|---|---|
| $\varepsilon$ | porosity |
| $\mu$ | dynamic viscosity (kg/ms) |
| $\rho$ | density (kg/$m^3$) |
| $\Delta H$ | reaction enthalpy (J/mol) |
| $\Delta S$ | reaction entropy (J/mol K) |

temperature, $T_L$, up to a high temperature $T_H$. The compression cycle process is summarized as follows:

Step A: A low pressure hydrogen supply (e.g. an electrolyser) is attached to the first stage, at pressure $P_s$. The temperature of stage 1 is maintained at $T_L$, during hydrogenation.
Step B–C: A sensible heating process for the stage 1 metal hydride bed occurs heating the bed to $T_H$ increasing the pressure of the stage 1 vessel.
Step D–E: A coupling process between stage 1 (dehydrogenation at $T_H$) and stage 2 (hydrogenation at $T_L$) occurs.
Step F–G: Stage 2 hydride bed undergoes sensible heating in order to achieve the delivery pressure of $P_d$.
Step H: During dehydrogenation of stage 2 high pressure hydrogen is released from the compressor at $P_d$.

### 2.2. Mathematical model

The following assumptions are made for simplifying the hydrogen storage and compression analysis.

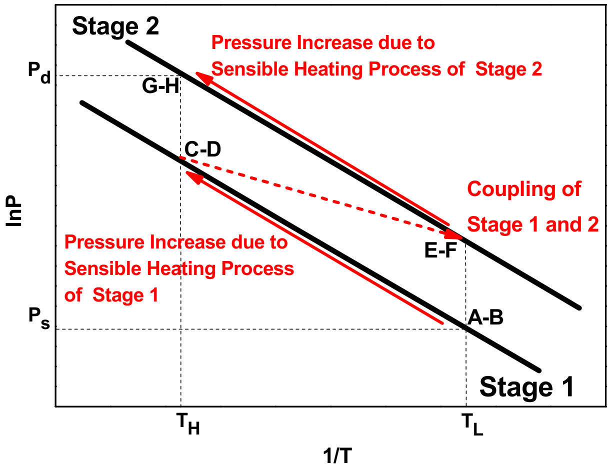

**Fig. 1.** A van't Hoff plot illustrating the operation of a two-stage Metal Hydride Hydrogen Compression system from the low temperature $T_L$ to the high temperature $T_H$. A sensible heating process is performed after each hydrogenation process to increase $P_{eq}$ inside each compression stage. For clarity, the black lines represent the van't Hoff plot for the hydrogenation process for stage 1 (lower black line) and for stage 2 (upper black line). The coupling process between stage 1 and stage 2 is represented by the dashed line.

(a) Initially the temperature and pressure profiles are uniform.
(b) Thermal conductivity and specific heat of the hydrides are assumed to be constant during the compression cycle.
(c) The medium is in local thermal equilibrium which implies that there is no heat transfer between solid and gas phases
(d) Hydrogen is treated as an ideal gas from a thermodynamic point of view.

### 2.3. Heat equation

Assuming thermal equilibrium between the hydride powder and hydrogen gas, a single heat equation is solved instead of separate equations for both solid and gas phases:

$$(\rho \cdot Cp)_e \cdot \frac{\partial T}{\partial t} + (\rho_g \cdot Cp_g) \cdot \vec{v}_g \cdot \nabla T = \nabla \cdot (k_e \cdot \nabla T) + m \cdot \left( \left( \frac{\Delta H}{M_{H_2}} \right) - T \cdot (Cp_g - Cp_s) \right) \quad (1)$$

where, the effective heat capacity is given by;

$$(\rho \cdot Cp)_e = \varepsilon \cdot \rho_g \cdot C_{pg} + (1 - \varepsilon) \cdot \rho_s \cdot C_{ps} \quad (2)$$

and the effective thermal conductivity is given by;

$$k_e = \varepsilon \cdot k_g + (1 - \varepsilon) \cdot k_s \quad (3)$$

The terms $\rho_g$, $C_{pg}$, $C_{ps}$ and $m$ refers to the density of the gas phase, the heat capacity of the gas phase, the heat capacity of the solid phase and the kinetic term for the reaction respectively.

### 2.4. Hydrogen mass balance

The equation that describes the diffusion of hydrogen mass inside the metal matrix is given by:

$$\varepsilon \cdot \frac{\partial(\rho_g)}{\partial t} + \mathrm{div}(\rho_g \cdot \vec{v}_g) = \pm Q \quad (4)$$

where, (−) is for the hydrogenation process and (+) is for the dehydrogenation process, $v_g$ is the velocity of gas during diffusion within the metal matrix (see Section 2.5) and $Q$ is the so-called Mass Source term describing the mass of hydrogen diffused per unit time and unit volume in the metal lattice.

### 2.5. Momentum equation

The velocity of a gas passing through a porous medium can be expressed by Darcy's law. By neglecting the gravitational effect,