



Thermodynamic simulation of hydrogen based solid sorption heat transformer

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ABSTRACT

The hydrogen absorption and desorption pressure-concentration isotherms (PCI) of $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ and $\text{LaNi}_{4.6}\text{Al}_{0.4}$ alloys are measured by static and dynamic method. The hydrogen absorption and desorption kinetics of the alloys also measured. The measured properties are used to study the performance of hydrogen based solid sorption heat transformer (MHHT). The thermodynamic simulation of MHHT is conducted using statically and dynamically measured PCI and thermodynamic properties. Due to variation in statically and dynamically measured metal hydride properties, significant variations in coefficient of performance (COP) and heat transformation capacity of MHHT are observed. In case of thermodynamic analysis using dynamically measured properties the heat transformation capacity and COP are decreased by 42.6% and 22.2% respectively, compared to statically measured data. This is due to the decrease in amount of hydrogen transmission and pressure differential between paired metal hydride reactors, and increase in reaction enthalpies. Later, the actual thermodynamic cycle for MHHT is constructed by considering the variation in pressure during hydrogen transfer processes between the metal hydride reactors.

1. Introduction

Hydrogen can be hoarded safely in metal hydrides by chemical absorption and retract again by supplying heat. During the storage and extraction of hydrogen to and from metal hydrides, a large amount of heat is released and supplied, respectively. These heat interactions provide possibility to the development of metal hydride based thermodynamic devices like heat pumps, air-conditioners, heat transformers, energy storage systems, etc. which can be operated using low-grade heat. For the development and thermodynamic simulation of any metal hydride based thermodynamic system, the fundamental requisition is to measure the PCI, reaction kinetics and thermodynamic properties of metal hydride.

Several studies [1–18] have been conducted to fine tune different composition of metal hydrides for the development of metal hydrides thermodynamic systems. The thermodynamic performance of one of the non-hydrogen storage applications of metal hydrides i.e. metal hydride based heat transformer, has been experimentally and theoretically studied by several researchers [19–32]. The methodology for the selection of suitable metal hydride pairs for heat transformers is presented by Balakumar et al. [20]. The suitability of low-pressure side and high-

pressure side for different thermodynamic applications is reported by Sun [27]. In 1984, Tuscher et al. [32] have employed a pair of $\text{LaNi}_{4.7}\text{Al}_{0.3}$ – $\text{MmNi}_{4.5}\text{Al}_{0.5}$ for heat transformer. The $\text{LaNi}_{4.7}\text{Al}_{0.3}$ – LaNi_5 pair is employed for heat pumps [19]. Other group [22–24,31] have reported the performance of metal hydride heat pumps for pair of different AB_5 alloys. On the other hand, metal hydride based system for simultaneous heating and cooling application was proposed by Ahmed and Murthy in 2004 [30]. Abraham et al. [14–16] have reported the influence of heat and mass recovery on the performance of heat pumps. These studies have been conducted using statically measured metal hydride properties. But, the hydrogen transfer processes involved in metal hydride based thermodynamic cycles are dynamic. Therefore, some researchers [32–39] have attempted to study the metal hydride thermal systems performance using dynamic property data. The consequences of static and dynamic working conditions on system performance were studied by Nagel et al. [34,35]. Vinod et al. [33,39] have reported that the statically measured PCIs are not suitable to study the actual variation in pressure differential among paired metal hydride. The processes involved in thermal system are dynamic in nature.

While several researchers have addressed the performance evaluation of MHHT considering static and dynamic operating conditions, it is

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observed that the studies on the effect of variation in actual pressure difference between paired metal hydride reactors and hydrogen flow rate on thermodynamic performance are scarce. Therefore, recently, authors have reported the effects of variation in amount of hydrogen transmission and pressure differential between the paired metal hydride reactors in the actual thermodynamic cycles on the performance of solid sorption cooling system and multi stage simultaneous heating and cooling system [32,39]. As an extension to that, in the present study, authors have measured pressure-concentration isotherms, reaction enthalpies and reaction rates of $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ and $\text{LaNi}_{4.6}\text{Al}_{0.4}$ hydride to determine their suitability toward the thermodynamic performance of MHHT. Finally, the actual thermodynamic cycle for MHHT is discussed.

2. Selection of metal hydrides

The desirable properties of metal hydrides required for the development of MHHT are (a) high hydrogen storage capacity that increases the amount of hydrogen transmission, (b) fast reaction kinetics to reduce overall cycle time, (c) flatter $\alpha + \beta$ phase and low hysteresis, which facilitates higher mass flow rates between metal hydride beds due to high pressure difference at fixed operating temperatures, (d) and favourable thermodynamic properties, generally, high absorption enthalpy to increase heat transformation and low desorption enthalpy to reduce regeneration input. In the present work, for better performance of MHHT, the low-pressure (LP) hydride should have high absorption enthalpy and high-pressure (HP) hydride should have low desorption enthalpy.

For the present work, $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ and $\text{LaNi}_{4.6}\text{Al}_{0.4}$ hydrides are selected based on the thermodynamic properties, reaction kinetics, and static and dynamic PCI properties measured in authors' previous studies [3,5,9,18]. The construction of experimental setups and procedure of PCIs and kinetics measurements are available in author's previous articles [3,5,9,18] in detail. The experimental setups for reaction kinetics and, static and dynamic PCI measurements are fabricated at Indian Institute of Technology Indore. The experimental setups are fabricated using 1/4-inch stainless steel (SS-316L) tubes for pipelines for gas flow, high-pressure bellow sealed valves (Swegelok) to regulate gas flow, calibrated cylinders (SS-316L) for hydrogen storage (supply volume) and piezoresistive pressure transducers (Omicron, Accuracy: $\pm 0.1\%$ of full scale) with working range of 0–200 bar are used to measure supply-volume pressure and metal hydride equilibrium pressure. A thermal mass flow controller (AALBORG, Accuracy 1%) with flow range of 0–200 ml/min is mounted for constant flow rate hydrogen supply. A thermostatic bath (Julabo, stability: ± 0.01 °C) is used to maintain isothermal condition. The operating temperature ranges from -50 to 200 °C. Water is used as working fluid for 0–80 °C and thermal bath fluid (Polydimethyl Siloxan, boiling point above 315 °C) is used as working fluid for 80–200 °C. Metal sheathed K type thermocouples (range: -200 to 1360 °C, accuracy: ± 0.1 °C, time constant: 0.2 s) are fitted at different locations to measure the temperature of hydrogen. Hydrogen gas of 99.99% purity is used for PCI and kinetics measurements, and argon of 99.99% purity is used for volume measurements, purging and in leak tests of the entire experimental setup. A vacuum pump (Hind Hivac) is used to evacuate the system to 5×10^{-3} mbar

with a digital Pirani gauge (accuracy of $\pm 0.5\%$). All the pressures and temperatures are logged using data logger (Agilent, 600 readings/sec). The dynamic PCIs are measured at 20 and 80 ml per min of hydrogen flow rate. In author's previous work, PCIs and reaction kinetics of these metal hydrides are measured in specific temperature range e.g. 20–80 °C. But in the present study, PCIs and reaction rates of chosen metal hydrides are measured at MHHT working temperature range. The maximum hydrogen storage capacity, plateau slope and hysteresis at mid-point are observed as 1.52 wt%, 0.55 and 0.54 respectively, for $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ hydride while those are observed as 1.41 wt%, 1.2 and 0.46 respectively, for $\text{LaNi}_{4.6}\text{Al}_{0.4}$ hydride. It is also observed that for same measurement temperatures $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ hydride having fast reaction kinetics than $\text{LaNi}_{4.6}\text{Al}_{0.4}$ hydride.

The estimated static and dynamic absorption and desorption enthalpies of chosen metal hydrides are as follows. For $\text{LaNi}_{4.6}\text{Al}_{0.4}$ – static absorption and desorption enthalpies are -34 kJ/mol and 36 kJ/mol whereas dynamic properties are -34.1 and 36.1 kJ/mol at 20 ml/min, and 36.1 and 38.3 kJ/mol at 80 ml/min, respectively. For $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ – static absorption and desorption enthalpies are -26 kJ/mol and 28 kJ/mol whereas dynamic properties are -26.1 and 28.1 kJ/mol at 20 ml/min, and 27.2 and 29.3 kJ/mol at 80 ml/min, respectively.

The measurement uncertainties for the hydrogen storage capacity and thermodynamic properties are estimated using the uncertainty measurement method available in authors' previous article [3]. The maximum cumulative uncertainty in storage capacity for $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ is calculated as 2.11%, whereas it is 1.31% for $\text{LaNi}_{4.6}\text{Al}_{0.4}$. The percentage errors in the reaction enthalpy measurements for $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ during absorption and desorption are calculated as 4.31% and 4.91%, respectively, whereas for $\text{LaNi}_{4.6}\text{Al}_{0.4}$, the percentage errors are 4.58% and 4.87%, respectively. Similarly, the percentage errors in reaction entropy measurements during absorption and desorption for $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ are 3.31% and 3.99%, respectively, whereas for $\text{LaNi}_{4.6}\text{Al}_{0.4}$, they are 4.43% and 4.96%, respectively. On the other hand, the maximum uncertainties in the estimation of storage capacity during reaction kinetics measurements are calculated using the method available in authors' other article [9]. They are calculated as 1.98% for $\text{La}_{0.9}\text{Ce}_{0.1}\text{Ni}_5$ and 2.41% for $\text{LaNi}_{4.6}\text{Al}_{0.4}$. The properties like storage capacity, reaction kinetics, and reaction enthalpy and entropy, of metal hydrides are calculated by measuring temperature, pressure and volume.

The maximum possible uncertainty is estimated from the minimum value of measured quantity and accuracy of the measurement system. The maximum uncertainties of the measured quantities are listed Table 1, and the calibration details of thermocouples and pressure transducers are presented in Appendix A.

3. Results and discussion

3.1. MHHT thermodynamic cycles

The basic principle and ideal thermodynamic cycle of metal hydride based heat transformer is given in authors' recent review article [17]. The MHHT system consists of two hydrogen transfer processes; heat transformation and regeneration. During heat transformation process, high-pressure (H-P) hydride desorbs hydrogen by taking low-grade

Table 1
Maximum uncertainty of the measured quantities.

S. No.	Measurement	Device	Accuracy	Min quantity	Max. Uncertainty
1	Hydrogen gas temp. in piping	K type Thermocouple	± 0.1 K	298 K	$\pm 0.033\%$
2	Metal Hydride bed temp.	K type Thermocouple	± 0.1 K	273 K	$\pm 0.036\%$
3	Pressure of Hydrogen gas	Piezoresistive type transducer	± 0.01 bar	1 bar	$\pm 0.01\%$
4	Mass flow rate of hydrogen	Thermal mass flow controller	± 0.001 g	0.35 g	$\pm 0.29\%$
5	Mass of alloy for PCI measurement	Digital weighing balance	± 0.001 g	20 g	$\pm 0.02\%$
6	Length	Vernier Caliper	± 0.1 mm	5 mm	$\pm 2\%$

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