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PARAMETRIC INVESTIGATION ON THE PURIFICATION CHARACTERISTICS OF $La_{0.9}Ce_{0.1}Ni_5$ UNDER VARIOUS ABSORPTION/DESORPTION CONDITIONS

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ABSTRACT

Metal hydride-hydrogen system can provide efficient solution for hydrogen separation from the impure gas mixtures. The present study reports the effect of various absorption/desorption conditions on the hydrogen separation and alloy poisoning characteristics of $La_{0.9}Ce_{0.1}Ni_5$. The experiments is being conducted by varying absorption pressure in the range of 5-20 bar, absorption temperature between 20-30 °C and desorption temperature between 60-90 °C. According to the experimental outcomes, absorption kinetics improves with increase in the absorption pressure and decrease in the absorption temperature. For 20 bar and 20 °C, 12.8232 g of hydrogen get absorbed for 10% impure gas mixture. The desorption temperature played a significant effect on the bed poisoning characteristics of the reactor bed. Desorption kinetics improves with increase in the bed temperature, and is significant for 80-90 °C. For lower range of desorption temperature (60-70 °C), the bed poisoning was significant. Even for 10% impurity, the reaction kinetics of alloy decreases significantly after 4-5 purification cycle. However, for higher desorption temperature (90 °C), the alloy regenerated itself and the bed poisoning is delayed. The system is capable of delivering 99.9995% pure hydrogen for an impurity range from 5-15% for gaseous impurities like CO_2, CH_4, N_2, Ar etc.

Keywords: Metal hydride, Hydrogen storage, Hydrogen purification, Alloy poisoning, Alloy regeneration

INTRODUCTION

In order to restrict the global rise in the temperature within 1.5 °C, the world is shifting their energy dependency to renewables like, wind, solar, hydro, hydrogen etc. Hydrogen is available in plenty in the form of hydrocarbons and chemical compounds; however, it has to be extracted through certain processes for the end user application. However, in most of the extraction processes, the hydrogen delivery is not 100% pure [1]. Hence, it becomes essential to purify hydrogen for end user application. There are several well established hydrogen purification techniques such as membrane diffusion, pressure swing adsorption, cryogenic separation, catalytic purification, metal hydride based purification, etc. available for hydrogen purification from gas mixture (hydrogen & other gases) of different impurities level, [1-4]. In comparison to different purification techniques, metal hydride (MH) based hydrogen purification technique is the simplest one to purify/separate hydrogen through thermally driven procedure, wherein the purification process is accomplished in three simple steps, i.e. absorption, flushing and desorption [5].

Schweppe et al. [6] reported the poisoning effect of O_2, CO_2, H_2S, CO or N_2 gases on $LaNi_5$ alloy used for hydrogen storage application. They used Sieverts apparatus, which was attached with a diffusion pump, to perform experimental studies related to reactor activation, absorption and alloy poisoning. It was observed that the presence of N_2 in the void space of the reactor, did not affect the rate of hydrogen absorption at all, while for the other prefilled gases, the initial rate of reaction got reduced by a range of 2 times to 10 times. However, O_2 critically damaged the absorption characteristics of the alloy by forming an oxide layer on the surface of $LaNi_5$ alloy. Similarly, Oztek [7] tested Mg_2Ni, VTiNi and $LaNi_5$, as a separation platform to purify hydrogen from a mixture of hydrogen and helium. Considering the reversible characteristics of $LaNi_5$ on interaction with the mixture, it was considered as suitable alloy for hydrogen separation. Further, Lin et al. [8] explored $LmNi_{4.8}Al_{0.2}$ alloy from AB_5 group of alloys, and studied its anti-poisoning characteristics for hydrogen storage application. The alloy was activated in the presence of pure hydrogen, and then they were exposed to CO and H_2S, each 1000 ppm as impurity with pure hydrogen. The poisoning effect of CO was more dominating as compared to H_2S.

Several studies have been reported on bed poisoning and hydrogen separation through metal hydride systems. However, the studies related to optimizing the operating parameters for a particular metal hydride and alloy regeneration are lagging in the literature. This motivates for the present study, wherein the hydrogen purification parameters and alloy regeneration has been discussed for $La_{0.9}Ce_{0.1}Ni_5$.

EXPERIMENTAL SETUP

The experimental setup was developed to carry out detailed purification study on the $La_{0.9}Ce_{0.1}Ni_5$ based MHHPS, wherein the impure sample preparation, absorption of impure mixture, flushing, desorption and sample collection for gas chromatography were carried out using a single gas circuit of specific configuration, as depicted in Fig. 1. The experimental setup mainly comprised of a MH reactor of embedded cooling tube (ECT) design with 6 ECT, Coriolis mass flowmeter to record amount of gas transferred, high & low temperature recirculating baths, vacuum pump, valves & fittings for gas and HTF circuit, gas cylinders, gas regulators, gas chromatograph and data acquisition system.

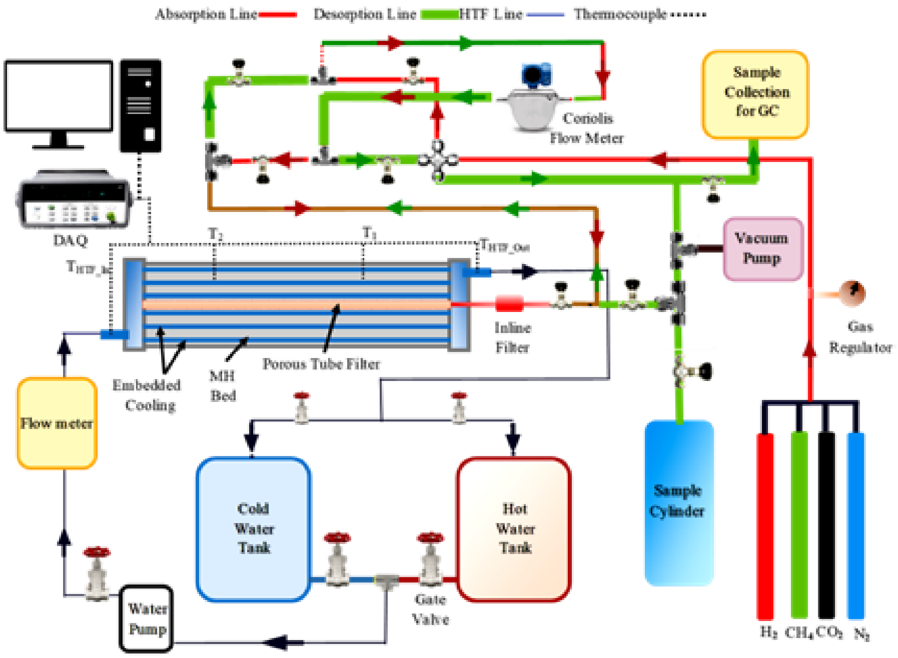

Fig. 1. Schematic of experimental test setup

REACTOR ACTIVATION AND EXPERIMENTAL PARAMETERS

Table 1. Experimental Parameters

Type of Study	Half Cycle	Operating parameters	Value
Parametric Study with Impure Hydrogen	Absorption	Supply pressure (**P_s**)	5, 10, 15 and 20 bar
		Absorption temperature (**T_a**)	20, 25 and 30 °C
		HTF flow rate	4 lpm
		Impurity level	5%, 10%, 15%
	Desorption	Desorption temperature (**T_d**)	60, 70, 80 and 90 °C
		HTF flow rate	4 lpm
	Purification Cycle	No. of Cycle	25 Cycle

Parametric studies with impure hydrogen was performed to obtain optimum absorption, desorption and flushing temperatures. The experimental parameter is summarised in Table 1. Before activation, the reactor was tested with argon gas at 80 bar to ensure leak proof system. Further, it was heated up to 90 °C, and evacuated up to 0.02 mbar to ensure no moisture was present in the alloy bed. The reactor activation was performed at hydrogen supply pressure (**P_s**) of 30 bar and 25 °C absorption temperature (**T_a**). In the first activation cycle, 14.4501 g hydrogen was absorbed in 527 s, while the peak bed temperature recorded was around 44 °C. Considering slow reaction kinetics, reactor was again evacuated keeping the bed at 90 °C. Further, the activation cycle was repeated 3 times to ensure the complete activation of the MH bed, where in amount of hydrogen absorbed was recorded as 15.4484 g, 15.8293 g & 15.5207 g, respectively in 2nd, 3rd and 4th activation cycle, with absorption time of 190 to 200 s. The repeatability in the amount of hydrogen absorbed and bed temperature variation during 2nd, 3rd and 4th activation cycle presented in Fig. 2, depicts the successful activation of the MH reactor with 1.2 kg of $La_{0.9}Ce_{0.1}Ni_5$, having storage capacity of 1.32 wt.%. The uncertainties in the experimental measurements were estimated using the method adopted by Moffat J Robert [8]. The uncertainties in measuring HTF flow rate and H_2 supply pressure were ± 1.67% and ± 3.0%, respectively, while the uncertainty in assessment of H_2 storage capacity was ± 2.5%. The error in the thermocouple reading was recorded ± 0.5 °C.

The detailed study and experimental outcomes will be reported in the full manuscript.

(a)

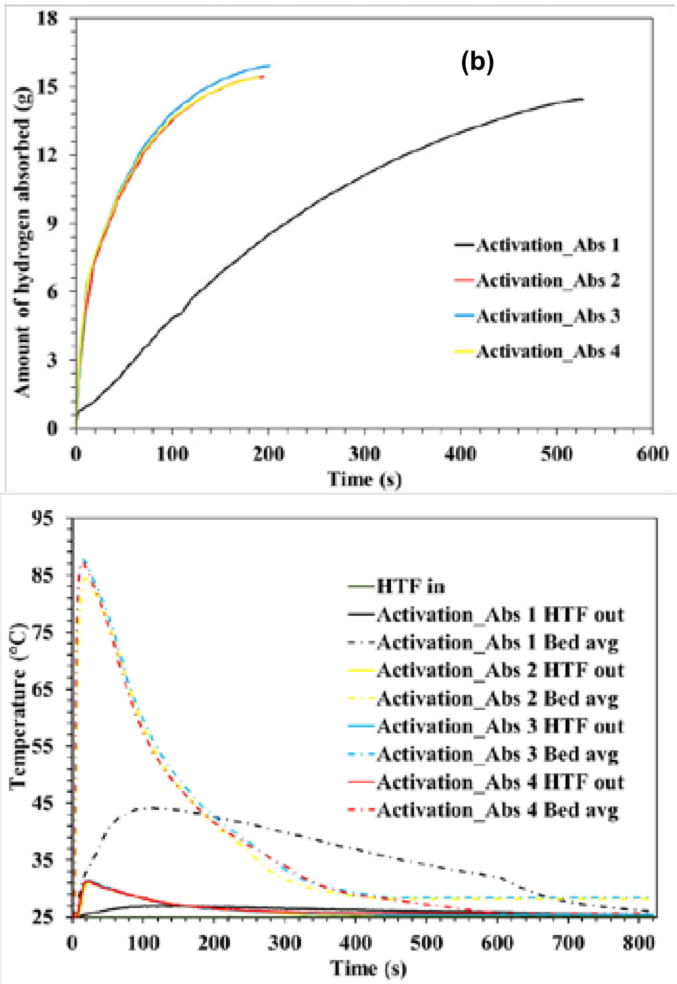

Fig 2. (a) Amount of hydrogen absorbed during each activation cycle, and (b) the average bed and HTF temperature variation for respective activation cycle.

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