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Thermodynamic and electric study of the $LaNi_{3,6}Al_{0,4}Co_{0,7}Mn_{0,3}$ alloy

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ABSTRACT

Improving hydrogen storage properties, as a very interesting energy vector, is the objective of several recent researches. AB_5 compounds, whose prototype compound is LaNi5 and its derivatives, have caught the attention of many researchers especially because they have a storage reversibility of hydrogen at ambient temperature and standard pressure. This paper presents an experimental study of hydrogen storage properties and an electric study of $LaNi_{3.6}Al_{0.4}Co_{0.7}Mn_{0.3}$ alloy. For this reason, the equilibrium isotherms are traced at different temperatures. The effect of temperature, pressure and coolant flow on the kinetics of the hydrogen absorption was also, studies. X-ray structural analysis indicated the formation of a single-phase orthorhombic structure with lattice parameters: a = 0.7517 nm, b = 0.7459 nm and c = 0.8565 nm.

Microstructural study showed a densely packed uniform distribution of grains over the surface of the sample. The Alternative current (a.c.) impedance plots were used as tools to analyze the electrical response of the sample as a function of frequency at room temperature. These plots revealed the presence of a relaxation process, which may be attributed to the same type of charge carrier. The frequency dependent conductivity was found to display the universal dynamic response behavior widely observed in disordered solid electrolytes.

Introduction

The depletion of fossil fuels and the increasing energy demands have led researchers to develop new energy storage techniques. Among these, the storage of hydrogen in the form of metal hydrides in Metal-Hydrogen Reactors (MHR) is recommended. Although hydrogen can be stored as compressed gas or cryogenic liquid, the storage of hydrogen in the form of metal hydride is considered more promising because it

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provides a large storage capacity of hydrogen under conditions of pressure and temperature close to the atmosphere (more security).

The hydrogen absorption properties of compounds AB_5compounds were discovered on the $LaNi_5$ in Philips Eindhoven Laboratories.

$LaNi_5$ has a storage reversibility of hydrogen with low weight percentage compared to Mg [1]. The addition of other metals to improve its storage performance is frequently used by researchers. Several studies have been conducted on compounds of $LaNi_{5-x}M_x$ type. Replacing a part of Ni atoms by M atoms is to increases or decreases the lattice parameter. Different elements have been used to replace Ni to increase the lattice parameter and consequently to lower the plateau pressure (Al, Mn, Cu). The substitution Aluminum for Nickel in different stoichiometric coefficients is studied by [2–6]. H. Dhaou et al. [7] substituted Ni aluminum ($LaNi_{4.85}Al_{0,15}$) and iron ($LaNi_{4.75}Fe_{0.25}$) for Ni. Their study shows that the hydruration rate is higher when they used the substituted samples than with $LaNi_5$ and that the level of equilibrium plateau is higher when they used the parent sample. F. Laurencelle et al. [8] studied $LaNi_{4.8}Sn_{0.2.}$ and They found that the compound stores a quantity of 1.6% of hydrogen at 80 °C.The effect of Sn on the stability of $LaNi_{5-x}Sn_x$ ($0 < x < 0.5$) was studied by E.M. Borzone et al. [9]. They observe a high stability for the sample which contains Sn, On the other hand, the absorption time is not strongly influenced by the addition of the Sn, however the desorption is delayed when Sn content is higher than 0.4 at [10]. studied the effect of the adding of Co on the hydriding kinetics of $LaNi_5$ and $LaNi_{4.73}Sn_{0.27}$. They found that the absorption/desorption reactions are strongly delayed in the presence of Co and that the sample which contains the Sn presents an equilibrium state after 10 cycles with no further degradation occurring in the past points. J.M.Joubert et al. [11] indicate that the addition of the Co makes the intermetallics resistant to the decrepitation, while the addition of Al reduces the dislocation rate. The $LaNi_{3.95}Co_{0.75}Al_{0.25}$compound combines the two effects. The interest in boron-based intermetallics is justified by their potential to store large quantities of hydrogen. This property was studied in detail by Buschowet al. [12]. Recently, many mathematical and experimental works have concentrated on metal hydride beds to be used for hydrogen storage medium and studied the effect of measurement parameters on hydrogen storage and thermodynamic properties [13,14].

The aim of this paper is to study experimentally the hydrogen sorption properties and the electric behavior of $LaNi_{3,6}Al_{0,4}Co_{0,7}Mn_{0,3}$.

Experimental procedure

For the thermodynamic study, the method used is the Sievert method [15]. After verifying of the etanchity in the tester bed, the primary vacuum is created using the vacuum pump (0.01 bar). For the absorption test, hydrogen is introduced in the hydrogen tank at a pressure P_0. The experimental procedure for the absorption begins by starting the vacuum pump to maintain a low pressure in the reactor (0.01 bar) at high temperature (50 °C) for a long period of time (4 h) to ensure that the sample is emptied of hydrogen (or other gases). Then, thermostated water flow through the heat exchanger to bring the container to a needed value of average temperature. The reference volume is fixed then loaded with hydrogen at a desired pressure (for example P = 11 bars. The container and the reference volume are then interconnected and the data. Hydrogen pressure P applied to the container and the equilibrium pressure Peq dependent on the hydride bed temperature [15].

Results and discussion

Thermodynamic study

Fig. 1, represent the absorption and desorption equilibrium isotherms at 20 °C, 30 °C and 40 °C, respectively. The equilibrium isotherms have the same appearance. In fact each isotherm is represented in three phases (α, α + β and β). The hysteresis phenomenon between absorption and desorption isotherms is presented in three temperatures. These isotherms show that the equilibrium pressure is reduced considerably compared to the equilibrium pressure of the parent compound ($LaNi_5$). It is noted that the increase in temperature leads to an increase of the equilibrium plateau pressure (Fig. 2).

In addition, the influence of the initial conditions on the absorption kinetics have been studied. That's why, the effect of the flow of the cooling fluid, of the initial temperature and that of the pressure have been tested.

Fig. 3 represents the effect of the flow of the cooling fluid (6 g/s, 9 g/s and 12 g/s) at the same conditions of temperature and pressure (P = 11 bars, T = 15 °C). It clearly seen observe that hydrogen concentration increases gradually to reach the same saturation value for the three coolant flows (H/M = 4).

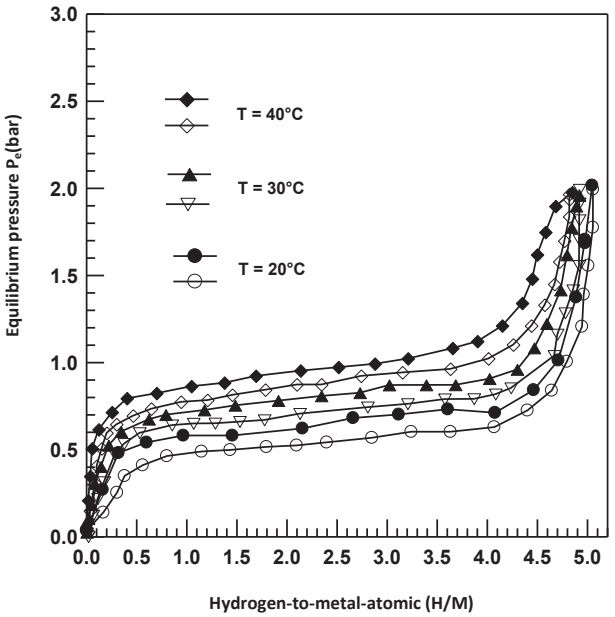

Fig. 1 – Equilibrium isotherm at T = 20 °C, T = 30 °C and T = 40 °C.

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