Journal of Solid State Chemistry 260 (2018) 73–79

Contents lists available at ScienceDirect

Journal of Solid State Chemistry

journal homepage: www.elsevier.com/locate/jssc

Preparation and characterization of mechanically alloyed AB_3-type based material $LaMg_2Ni_5Al_4$ and its solid-gaz hydrogen storage reaction

Hassen Jaafar[a,*], Luc Aymard[b], Walid Dachraoui[b], Arnaud Demortière[b], Mohieddine Abdellaoui[a]

[a] Laboratoire des Matériaux Utiles, Institut National de Recherche et d'Analyse Physico-chimique, BiotechPole Sidi Thabet, 2020 Ariana, Tunisia
[b] Laboratoire de Réactivité et Chimie des Solides - LRCS, UMR CNRS-UPJV 7314, RS2E Réseau Français sur le Stockage de l'Energie, 33 rue Saint-Leu, 80039 Amiens, France

ARTICLE INFO

Keywords:
Intermetallics AB_3 alloys
Mechanical alloying and milling
XRD analysis
TEM

ABSTRACT

We developed in the present paper the synthesis of a new AB_3-type compound $LaMg_2Ni_5Al_4$ by mechanical alloying (MA) process. X-ray diffraction analysis (XRD) was used to determine the structural properties and the phase evolution of the powder mixtures. Two different synthesis pathways have been investigated. The first starting from elemental metals and the second from a mixture of two binary compounds $LaNi_5$ ($CaCu_5$-type structure, *P6/mmm* space group) and Al(Mg) solid solution (cubic Fm-3 m space group). The results show multiphase alloys which contain $LaMg_2Ni_5Al_4$ main phase with hexagonal $PuNi_3$-type structure (R-3 m space group). Rietveld analysis shows that using a planetary ball mill, we obtain a good yield of $LaMg_2Ni_5Al_4$ compound after 5 h of mechanical alloying for both synthesis pathways. TEM analysis confirmed XRD results. SEM-EDX analysis of the final product was in agreement with the nominal chemical formula. A setup of possible solid-gaz hydrogenation reaction will be described so far at the end of this work. Electrochemical results demonstrate evidence on hydrogen absorption in the AB_3 material and the discharge capacity was equal to 5.9 H/f.u.

1. Introduction

Magnesium is the eighth most common element in the crust of earth and Mg-based intermetallic alloys are considered as good candidates for hydrogen storage applications [1].

However, its hydride has a thermodynamic problem: Mg-H binding energy it is rather estimated to be 75 kJ/mol H_2 and hydrogen sorption requires high temperature activation [2].

Early purpose was investigating possible $MgAl_2$ Laves phase formation by mechanical alloying (MA) since this alloy doesn't exist in binary Mg-Al stable phase diagram [3]. This metastable phase was found by liquid-solid quenching of an Al–30 at% Mg alloy [4].

Several studies have been done during the last decade on the mechanical alloying of Mg-Al compounds in different compositional range and on their hydrogenation properties [5–8]. Bouaricha et al. [5] studied the solid-gaz hydrogenation properties of Al(Mg) solid solutions where they noticed two sloping plateau at 11 and 20 atm for Mg:Al (58:42) at 350 °C. However the hydrogen sorption in Mg:Al (37:63) is being negligible [5]. Andreason [6] studied the hydrogenation of Mg_2Al_3 compound. A simple extrapolation of the Van't Hoff curve for ambient temperature gives very low hydrogen equilibrium pressure of about 3.10^{-4} bar.

Various functional inorganic nanostructures have been studied previously with several applications in catalysis and optoelectronic purpose. Some Intermetallic oxide materials for examples were synthesized using photocatalytic UV irradiation leading to spherical core-shell nanoparticles [12]. Other chemical one-pot reaction was reported based on the principle of Fehling's reaction resulting formation of multi-walled nanotubes with metal oxide nanoparticles [13]. However, this simple method is restrained to mono- or bimetallic compounds. Zhang and Reddy [14] studied further organometallic oxide materials using ultrasonication synthesis method giving nano-polymer composites. Recently, porous hybrid materials have attracted interest in the field of gas-sensing applications (chemical sensors and medical diagnostics); these materials were synthesized by mixing aqueous carbon nanotubes dispersion with metal oxides powder following specific muffle furnace heat treatment [15].

During the last years, research in our group turned to the development of AB_3 type alloys [9–11] from mixture of AB_5 and AB_2 alloys in 1:2 proportion. $LaNi_5$ has relatively high hydrogen equilibrium pressure at room temperature (~ 2 bar) [16]. Thus, the choice of AB_2 system is very important in order to have the appropriate properties for reasonable hydrogen storage AB_3 material.

* Corresponding author.

https://doi.org/10.1016/j.jssc.2018.01.015
Received 4 September 2017; Received in revised form 11 January 2018; Accepted 14 January 2018
Available online 04 February 2018
0022-4596/ © 2018 Elsevier Inc. All rights reserved.

Structural and electrochemical properties of Mg-based intermetallics like Mg_xNi_{100-x} obtained by mechanical alloying has been widely investigated previously: we established kinetics of hydrogen desorption relationship between the mechanical alloying conditions and the microstructural state of end products and also correlation between the microstructural state and the electrochemical properties [17–21].

In this paper, we report on the optimization of mechanical alloying process for Mg:Al(33:66) solid solution and the synthesis of a new AB_3-type compound $LaMg_2Ni_5Al_4$ in two different pathways. The approach established here leads to new material with higher hydrogen storage capacity and improved stability comparatively to the classical AB_2 and AB_5-type precursors. Moreover, these hydridable intermetallic materials provide higher capacity than non-chemical storing method like hydrogen liquid or compressed gas and can be used as H_2 tank or reservoir for Full cell [22–24]. However, due to superior Aluminum content, these materials are expected to be suitable for thermal Batteries research with promising possible outcome towards high temperature cells and for other lithiated composite electrode [25–27].

2. Materials and methods

For Al(Mg) solid solution, 1 g mixture of elemental Al (Fisher, 99.8% granular 20 mesh, France) and Mg (VWR, 99.8%, France), with an atomic ratio of 2:1, was sealed into a stainless steel vial (45 cm^3 in volume) with 5 stainless steel balls (12 mm in diameter and 7.16 g in mass) in a glove box filled with purified argon gas. The ball-to-powder weight ratio was equal to 36:1. The MA experiments were performed at room temperature using a Fritsch "Pulverisette P7″ planetary ball mill. The disc and vial rotation speeds were respectively equal to 350 and 700 rpm.

For synthesis of $LaMg_2Ni_5Al_4$ alloy from elementary pure metals, 1 g mixture of elemental La (Aldrich, 99.9% ingot under oil, France) Mg (VWR, 99.8%, France), Ni (Acros, 99.9%, France) and Al (Fisher, 99.8% granular 20 mesh, France) was introduced under the same previous conditions with disc rotation speed of 550 rpm with the following atomic proportions La at% = 8.3%, Mg at% = 16.7%, Ni at % = 41.7% and Al at% = 33.3%.

Same MA experiments were tested for synthesis of $LaMg_2Ni_5Al_4$ alloy obtained from binary precursors $LaNi_5$ and Al(Mg) in molecular ratio of 1:2.

Crystallographic characterization of synthesized powders was carried out by XRD using a (θ – 2θ) Panalytical XPERT PRO MPD diffractometer operating with Cu Kα radiation (λ = 0.15406 nm).

Phase identification was carried out using the X′Pert HighScore Plus software connected to the Inorganic Crystal Structure Database ICSD [28].

XRD diffractogram refinement was done using Rietveld method through FullProf Suite Program (2.05) [29]. Samples for electron microscopy were prepared by dispersing the powder in ethanol and depositing it on a holey carbon grid. Selected area electron diffraction (SAED) patterns and High-resolution transmission electron microscopy images (HRTEM) were obtained on FEI Tecnai F20 transmission electron microscope.

Typical procedure for hydrogenation storage test: 30 mg of AB_3 compound obtained from pure elements is placed in hydrogenation vessel set to vacuum for 1 h. A pressure of 11 bars was injected and the sample was heated at temperature of 280 °C for 24 h.

4.3 mg of hydrogenated powder was then mixed with 2.9 mg of Super-P carbon black (C_{sp}) for better conductivity and the overall mixture was placed in two electrode Swagelok cell: Li° as negative electrode, AB_3 as positive electrode and $LiPF_6$ (1 M) in Ethyl carbonate / Dimethyl carbonate (50/50) as electrolyte. Acquisition data are collected from a galvanostatic controller connected to feedback software MacPile 3.32. Swagelok cell preparation was performed in a glove box filled with purified argon gas.

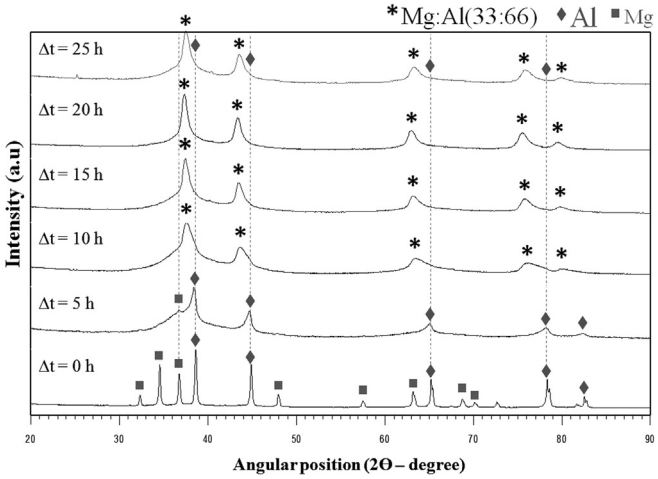

Fig. 1. XRD patterns of the mechanically alloyed Mg-Al mixture after different milling time.

3. Results and discussion

3.1. Characterization of Al(Mg) solid solution

Mg-Al mixtures prepared within different milling durations were examined using XRD for structural studies. Fig. 1 shows diffractograms of different samples (pristine, 5 h, 10 h, 20 h and 25 h). After 5 h milling, it seems that no reaction occurred and XRD diffraction pattern shows the peaks corresponding to initial face-centered cubic Aluminum and hexagonal Magnesium that obviously changed partly from crystalline to amorphous state.

After 10 h milling time, a new phase was observed. Identification by HighScore, connected to ICSD data base, shows that this phase corresponds to the Al(Mg) solid solution where Mg is incorporated in Al unit cell. This phase seems to be stable even for longer milling time.

Refinement of XRD pattern of the sample obtained after 15 h milling time is shown in Fig. 2 where the values of atomic occupation of Mg and Al in the different sites are corrected to consider the atomic ratio of the two initial elementary metals (1:2). This phase is refined in the cubic Fm-3 m space group and the vertical markers correspond to the allowed Bragg reflections for Mg:Al(33:66).

All refinement results including cell parameters, mass fraction and Rietveld refinement reliability factors are shown in Table 1. Theoretical weight fractions before milling are respectively 31.03% and 68.96% for Mg and Al.

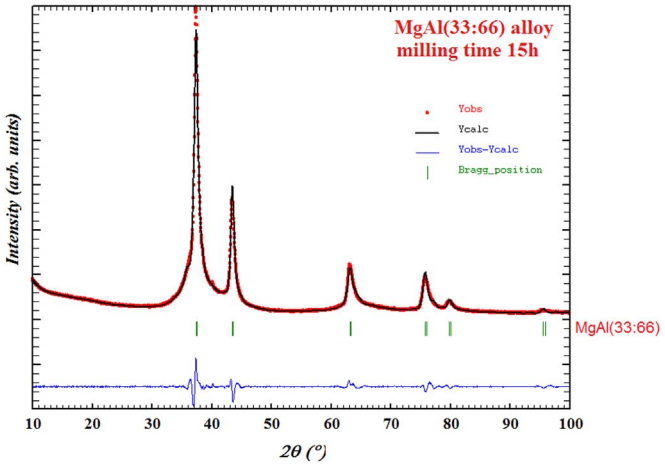

Fig. 2. Calculated (black line) and observed (red line) X-ray diffraction patterns for the Mg:Al(33:66) alloy obtained after 15 h milling time.

Download English Version:

https://daneshyari.com/en/article/7757812

Download Persian Version:

https://daneshyari.com/article/7757812

Daneshyari.com