INTERNATIONAL JOURNAL OF HYDROGEN ENERGY XXX (2018) 1–10

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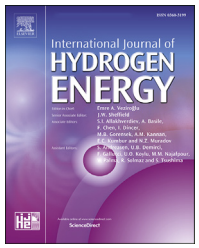

A datamining approach to classify, select and predict the formation enthalpy for intermetallic compound hydrides

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ARTICLE INFO

Article history:
Received 13 May 2018
Received in revised form
7 August 2018
Accepted 10 August 2018
Available online xxx

Keywords:
Intermetallic compounds
Datamining approach
Principal component analysis
Artificial neural network
Formability

ABSTRACT

In this paper, two techniques of datamining tools were adopted, a principal component analysis (PCA) and artificial neural network (ANN). A PCA to classify, select and identify several combinations between transition element A and B (B = Ti, Zr, Hf, Sc, Y, La and Th) and ANN to predict ΔH for ternary hydrides. Based on the datasets selected from different works, a principal component analysis (PCA) has been applied to select, classify and identify around 76 possible combinations between transition metal elements A and B. The results showed that the clustering of combinations A-B are significantly influenced by the atomic parameters of element A, such atomic radius (R_A), Pauling's electronegativity (χ_A) and atomic electron density (Z_A/R_A^3). From 76 combinations, 55 systems which have $\chi_A \geq 1.5$, $Z_A/R_A^3 > 1.28$ and $R_A < 1.46$ Å are categorized as group 1. On the other hand, 21 systems which have $\chi_A < 1.5$, $Z_A/R_A^3 < 1.28$, and $R_A > 1.46$ Å are categorized as group 2. From the first group, 46 different combinations are identified and have a negative ΔH, within 18 well-known promising binary alloys of hydrogen storage.

An (6-15-1) architecture of artificial neural network (ANN) has been developed to estimate the ΔH for the other ternary hydrides selected from different published works. The performance indices such as relative error, coefficient of determination (R^2) and mean square error (MSE) were used to control the performance of obtained results. In addition to this, the ΔH obtained from ANN model were compared with those experimental data and theoretical results available in the literature.

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https://doi.org/10.1016/j.ijhydene.2018.08.122

Introduction

The intermetallic compound hydrides are considered as an attractive material for hydrogen storage. These hydrides are promising materials both as fuel as in rechargeable batteries, fuel cells and heat storage [1,3]. They have been received wide attention due to their large number of technical applications and they opened new horizons for industrial development due to their widespread in the development of hydrogen-absorbing metal alloys [4–6]. In particular, the intermetallic compound hydrides (AB_5H_x, ABH_x, AB_2H_x, A_2BH_x and AB_3H_x, as well as solid solution alloys) are also widely investigated and have been an active research area due to their excellent hydrogen absorbing properties [7,8]. Indeed, this hydride is based on the combination between transition elements A and B. The stability of the binary formation A-B depends on both the atomic parameters of transition elements (A, B) and the processing conditions (Pressure, temperature,…). Due to the complex and non-linear effect of parameters influencing the stability and thermodynamic properties in the formation of both intermetallic compounds and their hydrides, a large number of theoretical studies and experimental works have been made [9–12], to understand, classify and study of various aspects of intermetallic compounds forming the hydrides.

The alloying ability of intermetallic compounds can be evaluated by its enthalpy of formation (ΔH). A negative ΔH means an exothermic process, and the intermetallic compounds can be formed and stable. For the predicting stable intermetallic compounds and its hydrides several theoretical methods have been reported to estimate the ΔH, such the Miedema's model [10,13], a semi-empirical model [11,14] and density functional theory (DFT) calculation [9]. Herbst [10] has used Miedema's model to estimate ΔH for binary $AH_{x\ (x=1\text{-}3)}$ and $AB_nH_{x\ (n=1,2,3,5)}$ type hydrides and predict the hydrogen content of binary and ternary hydrides. Griessen et *al.* [11] have calculated by means of a semi-empirical model based on the electronic structure of the host alloys the heats of formation of the transition metal hydrides A_5BH_x, A_2BH_x, ABH_x, AB_2H_x and AB_5H_x, where A = Sc, Ti, V, Y, Zr, Nb, La, Hf, Ta, Ni, Pd and B is transition metal, and have also predicted 44 different compounds to react with hydrogen. Watson and Bennett [15] have predicted the ΔH for 276 equiatomic composition of transition metal alloys by an electron band theory model based on the bandwidth, Fermi level position and number of electrons in the band. Also, by mean of the DFT calculation, J.Wang et *al* [9] have estimated the enthalpies of formation for Al-X (X = Co, Cu, Hf, Mg, Mn, Ni, Sr, V, Ti, Y and Zr) systems. However, the abundance of physical parameters of two transition elements A and B are probably responsible for the difficulties in the study of the formability of A-B system. It is difficult to develop them in the form of mathematical equations. Van Mal [16] proposed some criteria for classification of ternary transition metal hydrides according to ΔH.

Therefore, it is important to understand the effect of atomic parameters on the formability of intermetallic compounds and its hydrides. In this context, much recent informatics methods have been developed. In which a new knowledge system is built by collecting and classifying information with the help of calculations and databases [2]. Among this informatics method which attracts an increasing care of the material science researches is the data mining (DM). DM approach becomes a significant technique in the building knowledge-based system by inductive inference [17]. The most popular DM approaches are partial least square (PLS) (Linear regression), artificial neural network (ANN) and principal component analysis (PCA).

Following specific interest, a recent example of this type of approaches comes from Benyelloul et *al.* [18–20] where the authors demonstrate an application of ANN and PCA for the task of predicting several physical parameters and thermodynamic properties. In other hand, Jin Guo et *al* [21] have used the partial least square (PLS) method to classify and select the binary transition metal alloys. They found that the stability and the formability of hydrides of binary transition metal alloys are significantly influenced by electron density.

However, it is shown that the atomic parameters (such as electron density, atomic radius, Pauling's electronegativity and molar volume) effect strongly on the formation of binary alloys A-B systems and it is mandatory to study this effect.

Hence, two distinct types of datamining methods have been proposed in this study. Firstly, a PCA is used to select and classify several combinations between transition element A and B favourable for stability. Secondly, an ANN is used to predict ΔH from several ternary hydrides.

In this view, our paper is structured as follows. In section Computational methods, we give a brief overview of basic techniques used in datamining. In the following section concerning the results and discussion, we analysed the obtained results. Finally, a conclusion of the present work is given.

Computational methods

Principal component analysis

Principal component analysis (PCA) is one of most technique in exploratory data analysis in DM approach, it is the technique for data study which involves many variables and can effectively solve the correlation problem [22,23]. PCA is used to reduce a large set of variable that still contains most of the information in the large data set [24]. The original matrix data is decomposed and projected into two plots, the sample are classified in the score plot and the descriptors in terms of their separation of the samples in the loading plot following the principle component (PCs) axis [18]. The details of the PCA can be summarized as follow steps [23,25].

Step 1: The original multiple quality characteristic matrix:

$$X^0 = \begin{bmatrix} X_{11} & . & . & X_{1n} \\ . & . & . & . \\ . & . & . & . \\ X_{m1} & . & . & X_{mn} \end{bmatrix} \quad (1)$$

where m is the number of trials and n is the number of the quality characteristic.

Step 2: Data standardization

$$\tilde{X}_{ij} = \frac{X_{ij} - \tilde{X}_j}{S_j} \quad (2)$$

Please cite this article in press as: Djellouli A, et al., A datamining approach to classify, select and predict the formation enthalpy for intermetallic compound hydrides, International Journal of Hydrogen Energy (2018), https://doi.org/10.1016/j.ijhydene.2018.08.122