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Big-data analysis of phase-formation rules in high-entropy alloys

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ABSTRACT

Big-data analysis of phase-formation rules of high-entropy alloys (HEAs) was conducted and a phase formation rule from a dynamic view was deduced. It was indicated in literatures that cooling rate has a strong influence on the phase formation of HEAs. Higher cooling rate may promote the generation of amorphous phase, and accordingly suppress the formation of intermetallics. Meanwhile, it was also shown that cooling rate had little impact on the formation of solid-solution phase. To demonstrate this rule, a series of FeCoNi(AlSiB)$_x$ HEAs ribbons were fabricated by a melt-spinning technique, and the microstructure, mechanical, and magnetic properties were also investigated. The results show that all ribbons exhibit disordered solid-solution structure. The addition of boron changes the alloy from ductility to brittleness, but without evident change of magnetic properties. The alloy in the nominal composition of FeCoNi(AlSi)$_{0.2}$ has the best combination of mechanical and magnetic properties. A distinct feature of HEAs in magnetization was noticed and explained.

1. Introduction

High-entropy alloys (HEAs) have been widely investigated due to its exceptional mechanical behaviors since the concept of HEAs was proposed by Yeh et al.[1,2]. Moreover, the magnetic properties of HEAs have aroused increasing interest[3-7]. For example, HEAs were considered as very promising candidates for soft magnets because of its unique topological structure and complex composition. However, it is still an urgent problem to efficiently pick out the appropriate components from thousands of available combinations of elements satisfied with the HEAs criteria during HEAs design and preparation process.

Phase-formation rules are frequently employed in the design process of HEAs. To date, more than twenty kinds of rules have been proposed for different conditions. However, the real fact is that only a few effective HEAs samples can be fabricated in the laboratory environment for trial-error experimentation. Traditionally, the lengthy time frame for materials to move from discovery to market is due in part to the continued reliance of materials research and development programs on scientific intuition and trial and error experimentation, which means not only countless money, intelligence and labor input, but also low efficiency and long wait. Thus, there is another problem on how to choose or propose a reasonable phase-formation rule and thus find out the controllable preparation method of HEAs.

Although trial-error and summary is commonly used to find out the potential law, nowadays, data-driven method is taking an important role in the design and preparation process for a flood of data on the topic of HEAs, which is exceeding the processing capacity of conventional database systems. Big-data is typically broken down by three characteristics[8]: Volume—how much data, Velocity—how fast that data are processed, Variety—the various types of data. The main methods of big-data analysis include visualization analysis, data mining algorithm, predictive analysis, data quality, data management, and semantic engine. Usually the workflow of big-data processing is to collect data→import data/data treatment-statistics/analysis→data mining. There is no doubt that research efficiency and accuracy will be greatly improved if the big-data are fully analyzed. The research process can be greatly accelerated with the combination of theoreti-

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cal predictions and experimental verification.

So far, big-data analysis of HEAs properties starts with a largely silent, nonchallenged step: the choice of the descriptive parameters setting. In this paper, phase-formation rules developed with the big-data analysis were taken as a main parameter which had an important impact on the HEAs magnetic properties.

2. Big-data Analysis of Phase-formation Rules

It has been verified that multi-component HEAs did not always form solid-solution[9-11]. Intermetallic compounds and amorphous phases can also be formed in many situations. In order to avoid the trial-error experiment in selecting disordered solid-solution (DSS), many calculating parameters have been deduced to predict the phase formation of HEAs. Zhang et al.[12] proposed the parameters Ω and δ (mismatch degree), regarding ΔH_{mix} (mixing enthalpy) and ΔS_{mix} (mixing entropy).

$$\Omega=\frac{T_m \Delta S_{mix}}{|\Delta H_{mix}|} \tag{1}$$

$$\delta=\sqrt{\sum_{i=1}^{n} c_i\left(1-\frac{r_i}{\bar{r}}\right)^2} \tag{2}$$

$$\Delta H_{mix}=\sum_{i=1, i\neq j}^{n} \Omega_{ij} c_i c_j \tag{3}$$

$$\Delta S_{mix}=-R\sum_{i=1}^{n}(c_i \ln c_i) \tag{4}$$

$$T_m=\sum_{i=1}^{n} c_i (T_m)_i \tag{5}$$

where, Ω_{ij} ($=4\Delta H_{AB}^{mix}$) is the regular solution interaction parameter between the ith and jth elements; c_i and c_j are the atomic percentage of the ith and jth components, respectively; r_i is the atomic radius of ith atom; $\bar{r}$ is the average radius of ith to nth atoms; ΔH_{AB}^{mix} is the enthalpy of mixing of binary liquid alloys listed in Table 1; R is the gas constant, 8.314 J · K^{-1} · mol^{-1}; and $(T_m)_i$ is the melting point of the ith component of alloy.

Table 1
Chemical mixing enthalpy ΔH_{mix}^{AB} of different elements (kJ · mol^{-1})

Element	Fe	Co	Ni	B	Al	Si
B	−26	−24	24	0	0	−14
Al	−11	−19	−22	0	0	−19
Si	−35	−38	−40	−14	−19	0

According to their research results, $\Omega \geqslant 1.1$ and $\delta \leqslant 6.6\%$ should be the criterion for forming solid-solution phase. Fig. 1[12] presents the relationship between Ω and δ, in which the zone marked B means the zone that mainly forms bulk metallic glasses (BMGs), and the zone marked I mainly forms intermetallic compounds. There is also a transition zone which forms both the random solid solutions and the intermetallic compounds.

Following Zhang′s method, parameters were calculated and presented in Table 2 below. The results are also shown in Fig. 1.

Guo et al.[13] summarized the relationship between the HEAs structure and the valence electron concentration (*VEC*). Their work suggested using the

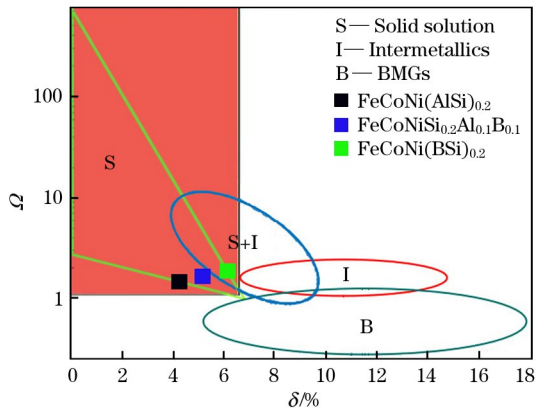

Fig. 1. Phase-formation map based on Ω and δ for HEAs.

Table 2
δ, ΔH_{mix}, ΔS_{mix}, Ω parameters and valence electron concentration (*VEC*) for CoFeNi(AlBSi)$_x$ alloy systems

Alloy	δ	ΔH_{mix}/(kJ · mol^{-1})	ΔS_{mix}/(J · K^{-1} · mol^{-1})	Ω	*VEC*
FeCoNi(AlSi)$_{0.2}$	4.325	−12.750	11.753	1.579	8.351
FeCoNiSi$_{0.2}$Al$_{0.1}$B$_{0.1}$	5.125	−11.754	12.075	1.807	8.348
FeCoNi(BSi)$_{0.2}$	6.189	−10.877	11.753	1.957	8.351

VEC ($VEC=\sum_{i=1}^{n} c_i (VEC)_i$, where $(VEC)_i$ is the *VEC* of the ith element) to predict the BCC or FCC structure solid solutions of HEAs. According to their conclusion, single FCC structure solid solution exists at $VEC \geqslant 8.0$, mixed FCC and BCC solid solution co-exist at $6.87 \leqslant VEC < 8.00$, and sole BCC structure solid solution exists at $VEC < 6.87$. Fig. 2[13] presents the relationship between the structure and *VEC* and the *VEC* of the CoFeNi(AlBSi)$_x$ series alloys was also calculated, which is shown in Table 2 and marked in Fig. 2[13].

It is obvious that the calculated parameter Ω, δ and *VEC* of the CoFeNi(AlBSi)$_x$ series alloys agree with these HEAs solid solution formation criterion above very well, and then it can be concluded that CoFeNi(AlBSi)$_x$ series alloys tend to form solid solution with melt-spinning method. However, the pa-