Alloy Designs of High-Entropy Crystalline and Bulk Glassy Alloys by Evaluating Mixing Enthalpy and Delta Parameter for Quinary to Decimal Equi-Atomic Alloys

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The values of mixing enthalpy (δmix) and Delta parameter (δ) were calculated with 73 elements from Miedema's model for multicomponent equi-atomic alloys to investigate the possibilities of the alloys to be formed into high-entropy (H-E) alloys or high-entropy bulk metallic glasses (HE-BMGs). The equi-atomic alloys from about 15 million (73C5) quinary to 621 billion (73C10) decimal systems were evaluated by referring to a δmix-δ diagram for zones S and B's for H-E alloys with disordered solid solutions and BMGs, respectively, reported by Zhang et al. The results revealed that the number of quinary equi-atomic alloys plotted in zone S is 28405 (~0.19% in 73C5), whereas those in zones B1 and B2 for conventional and Cu- and Mg-based BMGs, respectively, were 1036385 and 21518 (~6.90 and ~0.14%), respectively. This kind of statistical approach using δmix-δ diagram will lead to finding out unprecedented H-E alloys and HE-BMGs.


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1. Introduction

This paper principally describes high-entropy (H-E) alloys1,2 and bulk metallic glasses (BMGs)3 as well as relevant high-entropy bulk metallic glasses (HE-BMGs)4-8 and H-E glassy alloys.9 These four kinds of alloys are common in that they are multicomponent solid solutions, but H-E alloys and BMGs are completely different for their compositional features and atomic structures. Specifically, H-E alloys are principally equi-atomic or near equi-atomic crystalline alloys with five or more constituent elements and with either bcc, fcc or their mixture1,2 structure. On the other hand, BMGs are usually composed of constituent elements with three or more that are mixed non-stoichiometric in composition and are formed into non-crystalline alloys that exhibit a glass transition in behavior with temperature.3 Interestingly, H-E alloys and BMG are frequently developed under alloy designs with a same concept, although they exhibit different features mentioned above. Historically, the alloy design of H-E alloys and BMGs dates back to the early studies of metallic glasses that were performed in 1970s. For instance, it was reported by Chen in 1980 that principal factors for the formation of metallic glasses contain atomic size differences and heat of mixing (mixing enthalpy) as well as eutectic compositions, kinetic factors, valence electron concentration and so forth. Practically, H-E alloys and BMGs have been designed by referring to the factors and their threshold values. Here, the factors5 are selected from the following physical and thermodynamic quantities: (1) the number of constituent elements, (2) atomic size differences and (3) mixing enthalpy. The first factor is described as N, the second is frequently with Delta parameter (δ),11 percentage in atomic radius differences, mismatch entropy (S0)12 or topological instability (A) for multicomponent alloys13 based on a model for binary alloys,14 and the third with δmix. On the other hand, the criteria for H-E alloys and BMGs were separately given by determining numerical or thermodynamic values to the factors with aim to use the criteria as an alloy design. For instance, it was reported that H-E alloys requires N ≥ 5, and are plotted in zone S in a δmix-δ diagram.11 On the other hand, most of the BMGs satisfy N ≥ 3, the percentage in atomic radius differences ≥ 12%, and δmix/KJ mol−1 < 0.3 as well as zones B1 and B2 in the δmix-δ diagram. Thus, the alloy designs for H-E alloys and BMGs have common factors, but different criteria. Accordingly, the difference in criteria as well as different compositional features and atomic structures of H-E alloys and BMGs leads to the development of H-E alloys and BMGs separately. Hence, H-E alloys and BMGs have long been believed to be different metallic materials as a common knowledge.

However, this common knowledge has been broken by appearing HE-BMGs, which has both features of H-E alloys and BMGs. Specifically, HE-BMGs have recently been found in sequence in 2011 in a Zn20Cu10Sr20Yb20(Li0.55Mg0.45)20 alloy5 and a Pd20Pt20Cu20Ni20P20 alloy6 and the related two alloys2 of Sr20Cu20Yb20Mg20Zn20 and Sr20Cu20Yb20Mg20-Zn10Cu10 to the literature5 and an Er20Tb20Dy20Ni20Al20 alloy.7 Furthermore, a Zr20Ti20Cu20Ni20Be20 alloy8 has very recently been reported as the fifth type of HE-BMG. Historically, these HE-BMGs were found after several year intervals when the first HE-BMG was reported in 2002 in a Ti20Zr20Hf20Cu20Be20 alloy.4 The lacks of finding HE-BMGs during 2003 to 2010 and 2012 suggest a sort of difficulty in finding out new HE-BMGs, which may be due to the difference in nature between H-E alloys and BMGs. Thus, it is of great importance to analyze the features of H-E alloys,
BMGs and HE-BMGs statistically under unified conditions along their alloy designs.

The purposes of the present study are to analyze the equiatomic multicomponent alloys for their values of $\Delta H_{\text{mix}}$ and $\delta$ on the $\Delta H_{\text{mix}}$-$\delta$ diagram and to derive the candidates for H-E alloys and HE-BMGs in the multicomponent equi-atomic alloys.

2. Methods

The values of $\Delta H_{\text{mix}}$ were calculated based on eq. (1) where its right-hand side contains an assumption that $\Delta H_{\text{mix}}$ was calculated as quadratic formula with maximum or minimum at the equi-atomic composition in an A–B alloys system. In reality, the equi-atomic composition can be described as either AB, A50B50 or A0.5B0.5, respectively, for the compound-, atomic percentage- or atomic fraction-type. The factor four on the right-hand side of eq. (1) accounts for the value of $\Delta H_{\text{mix}}$ at the binary equi-atomic alloy, A0.5B0.5, with $c_i = c_j = 0.5$.

$$\Delta H_{\text{mix}} = 4 \sum_{i \neq j} \sum_{i=1}^{N} \Delta H_{ij} c_i c_j. \quad (1)$$

In calculating $\Delta H_{\text{mix}}$, actual values of $\Delta H_{ij}$ were acquired from the author’s previous work.15 On the other hand, $\delta$ was calculated by eqs. (2) and (3).11

$$\delta = \sqrt{\sum_{i=1}^{N} c_i (1 - \frac{r_i}{\bar{r}})^2} \times 100, \quad (2)$$

$$\bar{r} = \sum_{i=1}^{N} c_i r_i. \quad (3)$$

Here, $r_i$ is the atomic radius of the $i$-th constituent element and $\bar{r}$ the average atomic radius of the alloy defined by eq. (3). The $r_i$’s of the 73 elements were acquired from literature,10 and are also from the author’s previous work.15

Statistical analysis was performed for multicomponent equi-atomic systems with $N$ element ($5 \leq N \leq 10$). The number of alloys computed for their $\Delta H_{\text{mix}}$ and $\delta$ was amounted to be the combination of 73 elements taken from zones S, B1, and B2 and C where S is for H-E alloys, S’, B1 and B2 for BMGs without and with including Cu- and Mg-based alloys, respectively, and C for intermediate compounds. The details of the zones S, S’, B1 and B2 and C are reported in literature.11 The present study, in particular, focuses on zones S and B’s. It was reported11 that zone S was determined by Zhang and Zhou17 as

$$(0.7 <) \delta \leq 4.6, \quad (4)$$

$$(\delta - 12.2)^2 + (\Delta H_{\text{mix}} + 31.2)^2 = 47, \quad (5)$$

On the other hand, the authors originally formulated zones B1 and B2, which are drawn as ellipses in the literature.11 The zone B1 was approximated with eq. (6), as an ellipse with the center ($\delta$, $\Delta H_{\text{mix}}$) = (12.2, -31.2). In the same way, zone B2 for Cu- and Mg-based BMGs, which is a rotated ellipse to the $\delta$-axis, was investigated by applying a linear transformation for an ellipsoid with a center of ($\delta$, $\Delta H_{\text{mix}}$) = (9.10, -12.7) along the line, $\Delta H_{\text{mix}} = 7.808 - 83.5$. The rotating angle is about 82.69 degree, which was calculated by $\tan^{-1} 7.8$. The actual zones S and B’s will appear in figures in the next Section.

3. Results

First, the quinary equi-atomic alloys from 73 elements were investigated for their features on the $\Delta H_{\text{mix}}$-$\delta$ diagram. An especial attention was paid to the alloys plotted in zone S for H-E alloys. Figure 1 shows the $\Delta H_{\text{mix}}$-$\delta$ diagram acquired from literature11 on which the quinary equi-atomic alloys in zone S are plotted with small open circles where zone S is described by eqs. (4) and (5). It was found that the number of equi-atomic alloys plotted in zone S was amounted to be 28405 (~0.19%) for all the possible equi-atomic quinary alloys from $\gamma_7$Cr. The 28405 alloys sorted in the atomic number order start with L12Mg2Al2Ti20Ag20 alloy where ($\delta$, $\Delta H_{\text{mix}}$/kJ mol$^{-1}$) = (4.18, -2.88) and end with Re20Au20Hg20Bi20U20 alloy where ($\delta$, $\Delta H_{\text{mix}}$/kJ mol$^{-1}$) = (4.53, -6.56), in which it is without saying that representative H-E alloys found to date, such as CoCrFeNiCu13 are also included. The plots of quinary equi-atomic alloys cover zone S almost densely, but there are a small area with less densely-plotted near a line $\delta = 0.7$. This suggests that the boundary of the plots of $\gamma_7$Cr quinary equi-atomic alloys is near the origin of the $\Delta H_{\text{mix}}$-$\delta$ diagram.

Figure 2 shows the plots of the equi-atomic alloys from binary to quinary systems on the $\Delta H_{\text{mix}}$-$\delta$ diagrams, in which every values of $\Delta H_{\text{mix}}$ and $\delta$ are plotted with symbols: (a) cross for binary, (b) triangle for ternary, (c) square for quaternary and (d) pentagon for quinary alloys. Figure 2(a) for binary equi-atomic alloys shows that $\Delta H_{\text{mix}}$ and $\delta$ are densely plotted at a zone with $-30 \leq \Delta H_{\text{mix}}$/kJ mol$^{-1} \leq 50$.
and $0 \leq \delta \leq 20$. Some of the plots are out of the frame in Fig. 2(a) in a range $\Delta H_{\text{mix}} < -200$ kJ mol$^{-1}$. Furthermore, the plots in Fig. 2(a) appear to be divided into two areas roughly by a line: $\Delta H_{\text{mix}} = 58 - 220$, although the boundary is unclear. However, this boundary becomes clear in the ternary equi-atomic alloys shown in Fig. 2(b). In a similar way, the boundary of the area at the bottom-right side to the line ($\Delta H_{\text{mix}} = 58 - 220$) becomes more clear in Fig. 2(c) than that in Fig. 2(b). Besides, another boundary denoted by a line expressed by $\Delta H_{\text{mix}} = 58 - 170$ appears in Fig. 2(d) for quinary equi-atomic alloys. These results suggest that the plots in Fig. 2(d) are composed of two rectangles rotated and overlapped, which is shown in two lines in Fig. 2(d). The area at the upper-left side has a boundary denoted by a line with a formula of $\Delta H_{\text{mix}} = 58 - 220$, whereas the area at the bottom-right side is by another line described as $\Delta H_{\text{mix}} = 58 - 170$. The area at the bottom-right side is presumably due to the inclusion of the metalloid elements and its relatively small atomic radius than the metallic ones. The inclusions of metalloids with smaller atomic radius than metallic ones increase according to eq. (2).

Then, the features of $\Delta H_{\text{mix}}$ and $\delta$ were analyzed systematically by taking into account the change of $N$ from 5 to 10. Figure 3 shows the tendencies that (a) $\gamma_5C_N$, (b) alloys in zone S and (c) its percentage increase with increasing $N$ from 5 to 10. Figure 3 clearly shows that curves (a) and (b) increase with increasing $N$. However, curve (a) increases at a higher rate than curve (b), leading to decrease in curve (c) with increasing $N$. The decreasing tendency of curve (b) is presumably due to the increasing tendency of $\delta$ calculated by eq. (2) with increasing $N$. This increasing tendency makes the plots out of zone S in the $\Delta H_{\text{mix}}-\delta$ diagram for $\delta > 4.6$. Figure 3 demonstrates that candidates for H-E alloy reach to one million for octal equi-atomic alloys ($N = 8$), and that its percentage is nearly 0.01% in $\gamma_5C_8$. Hence, $N = 8$ can be an experimental and engineering goal when considering the additional tasks for experiments and for obtaining engineering products.

Finally, the authors analyzed the possibility to have HE-BMG in quinary equi-atomic alloys based on the results on the $\Delta H_{\text{mix}}-\delta$ diagram. Figure 4 demonstrates the quinary equi-atomic alloys plotted in zones B’s. The number of quinary equi-atomic alloys in zone B$_1$ is $1036385$ ($\sim 6.90\%$ in $\gamma_5C_5$), which is composed of Li$_2$B$_{20}$B$_{20}$Nb$_{20}$Rh$_{20}$ to Hg$_{20}$Pb$_{20}$B$_{20}$Pb$_{20}$Pu$_{20}$ alloys in the order of atomic number arrangement with $(\delta, \Delta H_{\text{mix}}/\text{kJ mol}^{-1}) = (17.67, -32.96)$.
to (8.69, −29.28), respectively. The number of 1036385 is considerably greater than those in zone S with 28405 (~0.19%). However, it appears that most of the quinary equi-atomic alloys in zone B1, such as Hg20Pb20Bi20Th20Pu20 alloy, are different from glass-formers. On the other hand, the number of quinary equi-atomic alloys in zone B2 is amounted to be 18690 (~0.126% in 73(C3)) where the alloys ordered with atomic number are from Lzb0Be20Al20Nb20Ta20 to Tlz0Pb20-Thz0U20Pu20 with (δ, ΔHmix/kJ mol−1) = (9.65, −8.32) to (8.92, −16.48), respectively. Among the 18690 alloys, the number of alloys containing Cu and/or Mg is 3446, which literary can be regarded as HE-BMGs with prototypical BMGs in Cu- and Mg-based systems, although further additional conditions are required to narrow down the appropriate candidates. These results merely indicate that the number of quinary equi-atomic alloys tends to depend on extent of zones S and B’s in the ΔHmix−δ diagram in the order of B2 < S < B1. This tendency contradicts to a fact that one faces with difficulty in finding new HE-BMGs in recent years. In other words, the number of candidates for HE-BMGs especially for zone B1 = 1036385 are overestimated considerably due to lacks of other necessary conditions for forming HE-BMGs. Thus, the authors discuss the additional necessary conditions for the equi-atomic alloys to narrow down the candidates for HE-BMGs in the next Section, together with peculiarities of some H-E alloys and HE-BMGs.

4. Discussion

The authors discuss the following items: (1) necessary conditions for forming HE-BMGs on the basis of the authors’ previous results on classifications of BMGs15,18 and (2) an Al0.5TiZrPdCuNi H-E alloy7 and the Ti20Zr20Hf20Cu20Ni204) and Zn-containing HE-BMGs5,7 for their peculiarities in positions of the plots in the ΔHmix−δ diagram.

4.1 Necessary conditions for forming HE-BMGs

It is considered that appropriate necessary conditions are required to the results shown in Fig. 4 to narrow down the candidates for the H-E BMGs from the equi-atomic alloys that possess ΔHmix and δ in zones B’s. As a candidate for the condition, one can list relationships between seven classes of BMGs (C-I to C-VII) and their main constituent element and its relative atomic size. Table 1 summarizes seven classes of BMGs (C-I to C-VII), sub-groups comprising BMGs, representative BMGs and their descriptions with sub-groups15,18 and representative HE-BMGs that relate to classes of BMGs. As summarized in Table 1 from our previous work18, the main constituent element of BMGs tends to be the one with the greatest atomic radius for C-I, C-V and C-VII, the intermediate for C-II, C-III and C-IV, and the smallest for C-VI. In addition, BMGs belonging to C-II, C-III and C-IV require further conditions for ΔHmix of ΔHmix between the atomic pairs excepting for the main elements should be two times or larger and negative in magnitude than ΔHmix between atomic pairs including the main element. For instance, an Fe70Zr10B20 alloy belonging to C-II exhibits atomic radii (r) of rFe (0.124 nm) > rZr (0.162 nm) > rB with ΔHmixFe−Zr of 6 kJ mol−1, whereas that of Y−Cu is 71 kJ mol−1. In a similar way, a Mg70Y10Cu20 alloy belonging to C-IV exhibits rY (0.182 nm) > rMg (0.160 nm) > rCu (0.128 nm) and ΔHmixMg−Cu−Y of 25 and −26 kJ mol−1, respectively, whereas that of Zr−B is −71 kJ mol−1. In a similar way, a Mg70Y10Cu20 alloy belonging to C-IV exhibits rY (0.182 nm) > rMg (0.160 nm) > rCu (0.128 nm) and ΔHmixMg−Cu−Y of 25 and −26 kJ mol−1, respectively, whereas that of Y−Cu is 71 kJ mol−1. In a similar way, a Mg70Y10Cu20 alloy belonging to C-IV exhibits rY (0.182 nm) > rMg (0.160 nm) > rCu (0.128 nm) and ΔHmixMg−Cu−Y of 25 and −26 kJ mol−1, respectively, whereas that of Y−Cu is 71 kJ mol−1. Thus, Fe70Zr10B20 and Mg70Y10Cu20 alloys satisfy that rFe and rMg are intermediate in each alloy and that ΔHmixFe−Zr and ΔHmixY−Cu are larger and negative in magnitude two or more times than that for the other atomic pairs. These relationships of BMGs among class, sub-group and relative atomic size of the main constituents as well as ΔHmin restrict BMG formations as glass-forming system and provides a hint to develop HE-BMGs from prototypical BMGs.
In reality, most of HE-BMGs found to date appear to be developed by undertaking a procedure to share the fraction of the main constituent element of prototypical BMGs with the other chemically-similar elements in the same sub-group as summarized in Table 1. This process is well-satisfied for Pd20Pt20Ni20Cu20P20 HE-BMG from a prototype of Pd40Ni40-P20 BMG when 40 at% Pd is replaced by 20 at% Pd and 20 at% Pt, whereas Ni is by Ni and Cu. In a similar way, Er20Tb20Dy20Ni20Al20 HE-BMG would be found from a prototypical Zr60Al15Ni25 BMG by approximating the Ni and Al contents ~20 at% roughly and by replacing Zr with 60 at% for lanthanides. In strong contrast, it is difficult to share the main constituent element from prototypical BMGs belonging to C-II and C-III, such as Fe-based BMGs. An explanation of this difficulty is due to the fact that substituting Fe with \( r_{Fe} = 0.124 \text{ nm} \) for the other elements, excepting for Co and Ni with \( r_{Co} = r_{Ni} = 0.125 \text{ nm} \) violates a rule regarding intermediate atomic radius of the main constituent element for BMGs in C-II and C-III. Here, it should be noted Cu has nearly the same atomic radius (\( r_{Cu} = 0.128 \text{ nm} \)) but Fe-Cu sub system is an immiscible system, which is against a glass-former, with \( \Delta H_{mix} \) of Fe-Cu is +13 kJ mol\(^{-1}\). In reality, \( r \)'s of late-transition metals with 4d and 5d orbitals ranges larger than Fe, such as \( r_{Fe} = 0.137 \text{ nm} \) and \( r_{Cu} = 0.144 \text{ nm} \), which increase the atomic radius when replacing Fe with late-transition metals with 4d and 5d orbitals. Thus, absence of alternative constituent elements replaceable with Fe is a characteristic of BMGs in C-II and C-III, leading to difficulty in finding out new HE-BMG. In a similar way, it is almost impossible to have HE-BMGs from BMGs belonging to C-VII due to less possibility to replace Ca and Mg with other alkaline and alkaline earth elements. Accordingly, candidates of new HE-BMGs will be found principally from prototypical BMGs classified into C-I. The above-mentioned additional conditions, which are somewhat difficult to be dealt with computationally, will considerably attribute to decrease the number of candidates for HE-BMGs, presumably by the factor of ten thousands or less, from 1036385 (~6.90% in \( \gamma_7\text{Ca} \)) to \( \gamma_1\text{Cu} \) given from Fig. 4.

As for H-E alloys, additional conditions also should be taken into account to squeeze the candidates from the statistical results. However, to the best of the authors’ knowledge there exist only a few conditions available at present, such as valence electron concentration (VEC)\(^{(20)}\) or unified parameter \( \Omega^{(21)} \) from thermodynamic quantities including \( \Delta H_{mix} \) and others. Thus, the present study does not proceed into the details about the necessary conditions for H-E alloys.

### 4.2 Peculiarities of Al0.5TiZrPdCuNi H-E alloy and Ti20Zr20Hf50Cu20Ni20 and Zn-containing HE-BMGs

In contrast to decreasing the number of candidates for HE-BMGs by adopting additional necessary conditions mentioned above, peculiarities of the alloys contribute to increasing the candidates of HE-BMGs as well as H-E alloys. For instance, the Al0.5TiZrPdCuNi H-E alloy,\(^{(9)}\) Ti20Zr20Hf50Cu20Ni20\(^{(4)}\) and Zn-containing HE-BMGs\(^{(5,7)}\) are considerably different from conventional H-E alloys in points that former three alloys have larger and negative \( \Delta H_{mix} \) and larger \( \delta \) values than the latter alloys in zone S. This is due to the facts that the Al0.5TiZrPdCuNi H-E alloy was developed through a procedure of fining new H-E glassy alloy and that Ti20Zr20Hf50Cu20Ni20 and Zn-containing HE-BMGs are composed of glass-forming elements. This indicates that glassy alloys usually possess larger and negative \( \Delta H_{mix} \) and larger \( \delta \) values than conventional H-E alloys and that BMGs are considerably different from conventional H-E alloys. In reality, the Al0.5TiZrPdCuNi H-E alloy is plotted significantly different from zone S. A possible reason for this has recently statistically analyzed\(^{(22)}\) with the help of the plots of 6150 ternary amorphous alloys in the \( \Delta H_{mix}-\delta \) diagram. The statistical analysis revealed that the Al0.5TiZrPdCuNi H-E alloy has a set of \( \Delta H_{mix} \) and \( \delta \) values where the amorphous alloys are less formed in the \( \Delta H_{mix}-\delta \) diagram, which is similar to zone S for H-E alloys.
that is located out of the area of amorphous formation. On the other hand, the Ti_{20}Zr_{20}Hf_{20}Cu_{20}Ni_{20} HE-BMG is peculiar in terms of combinations of elements discussed in Table 1. In reality, the Ti_{20}Zr_{20}Hf_{20}Cu_{20}Ni_{20} can be written as (d_{s}f_{h0}(d_{s}p)_{30} with s-d_{s}f-d_{s}p-p description in Table 1, which has opposite fractions to (d_{s}p)_{30}(d_{s}f)_{30} from BMGs belonging to C-VI, such as Cu_{60}Zr_{30}Ti_{10} alloy. Besides, one cannot rule out a possibility that new HE-BMGs will be found unprecedented area than the conventional zones S, S’, C, B_1 and B_2, such as Zn-containing HE-BMGs^{5,7} plotted apart from the above zones in the ΔH_{mix}-δ diagram in Fig. 4. These peculiar alloys are difficult to be dealt with in the present study for finding out new H-E alloys and HE-BMGs, since the present study utilizes statistical analysis based on the zones S and B’s, which have already been known as the ones for H-E alloys and BMGs due to a certain amount of enough number of the alloys. However, the statistical approach in the present study has a potential to include these peculiar alloys for their H_{mix} and δ values when the number of the alloys becomes large enough to form specific zones in near future. Thus, the authors reached the tentative conclusion that statistical approaches using ΔH_{mix}-δ diagram for zones S and B’s will be useful to find out new HE-BMGs as well as H-E alloys with the help of additional necessary conditions to narrow down the statistically-obtained candidates. Further detailed results will be presented somewhere in near future.

5. Conclusions

Multicomponent equi-atomic alloys ranging from quinary to decimal systems were evaluated for mixing enthalpy (ΔH_{mix}) and Delta parameter (δ) with 73 elements from Miedema’s model to find out candidates for high-entropy (H-E) alloys and high-entropy bulk metallic glasses (HE-BMGs). The alloys were plotted in ΔH_{mix}-δ diagram including zone S for H-E alloys with disordered solid solutions and zone B’s for BMGs determined by Zhang et al. The results revealed that the number of alloys plotted in zone S is 28405 (~0.19% in 37_2C_3) for quinary equi-atomic alloys and 4236606 (~0.0007% in 73_1C_10) for decimal system, whereas those in zones B_1 and B_2 for conventional and Cu- and Mg-based BMGs, respectively, for quinary equi-atomic alloy were 1036385 and 21518 (~6.90 and ~0.14% in 37_2C_3). This kind of statistical analysis will be useful for searching for new HE-BMGs as well as H-E alloys when additional necessary conditions are provided to narrow down the candidates for the statistically-obtained results.

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