LETTER

The influence of clusters in the melt of Fe$_{80}$Si$_{10}$B$_{10}$ alloy on the subsequent glass-formation

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Abstract The structure of Fe$_{80}$Si$_{10}$B$_{10}$ alloy melt was investigated by $ab$ initio molecular dynamic simulation from the local atomic environments. It presents that Fe$_{80}$Si$_{10}$B$_{10}$ alloy can be considered as the combination of B-centered prism-like clusters and bcc-like Fe-Si solid solution. The poor glass-forming ability of the alloy has been investigated and can be attributed to the bcc-like environment around Si atoms and the relative high content of pure Fe clusters.

1. Introduction

Alloys based on the Fe–Si–B system in the amorphous and nanocrystalline states are known to have considerably high magnetic properties [1–4]. With the development of modern industrialization, continuous precision casting has become to the development trend. Investigating the heat history and metastable microstructure transformation of alloy melts is significant for the casting process that affects the crystallization kinetics in an under-cooled liquid [5], the morphology and the properties of the amorphous product [6]. So, the microstructural details of the Fe–Si–B alloy series needs to be further enlarged.

Computer simulations offer an alternate possibility to determine the structure of liquids, since one of the most significant advantages of simulation method is having the ability to give 6 partial pair distribution functions (PDFs) for Fe-based ternary alloy systems.

In this paper, the effects of metalloid-centered clusters on the stability of Fe$_{80}$Si$_{10}$B$_{10}$ alloy melts are investigated by using $ab$ initio molecular dynamics simulations. The local environments around metalloid atoms and their roles will be discussed to compare with already reported work and to boost
our fundamental understanding about the characteristic of Fe–Si–B amorphous alloys.

2. Material and methods

Although with classical Molecular Dynamics, it is easily possible to simulate structures with millions of atoms in reasonable time, an accurate simulation of the properties of Fe-metalloid type Metallic Glasses (MGs) is still a challenging problem, because obtaining an effective potential for classical Molecular Dynamics is challenging. At the same time, ab initio studies of bulk properties, such as crystallization and metadislocations, even with fast codes using density functional theory like VASP, are currently unfeasible [7], which is still confined to systems containing hundreds of atoms [8].

As focused on the local atomic structure, ab initio molecular dynamics simulations can offer a reliable statistic. In our simulations, a cubic cell containing 100 atoms with periodic boundary conditions and 3000 configurations was used for structure analysis after Fe80Si10B10 melt was equilibrated at 30 K above its liquidus temperature (1465 K) for 3 ps. For more details, see Ref. [9,10].

3. Results and discussion

We performed Voronoi Polyhedra (VPs) analysis on the atomic configuration and employed a signature to represent shapes of the atom-centered polyhedron: $n_3, n_4, n_5, n_6$. Figs. 1 and 2 exhibits the 15 most dominated atom-centered VPs diagram of the alloy melt. No signature of $<0, 0, 6, 0>$ for bcc and $<0, 12, 0, 0>$ for fcc environment, respectively, has been found yet.

The key findings taken from the Figs. 1 and 2 are as follows: (1) The preferred VPs around Si and Fe atoms are indexed as $<0, 3, 6, 4, 2>$ and its derivatives, including $<0, 3, 6, 3>$, $<0, 2, 8, 4>$, and $<0, 3, 6, 5>$. As $<0, 3, 6, 4>$ is usually considered as the deformed body-centered cubic (bcc) clusters, Si atoms have the similar bcc-like environment with Fe matrix. (2) Among B-centered VPs, $<0, 3, 6, 0>$ with the fraction of 15.8% and $<0, 2, 8, 0>$ with the fraction of 9.2% dominate in the form of prism-like VPs, which represent the 9 coordinated tri-capped trigonal prism (TTP) and the 10 coordinated Archimedean antiprism, respectively. These agree well with the results obtained by other simulation method [11] and with our previous results [12].

The sum of B-centered prism-like VPs including $<0, 3, 6, 0>$ and $<0, 2, 8, 0>$ with the fraction of 15.8% and $<0, 2, 8, 0>$ with the fraction of 9.2% dominate in the form of prism-like VPs, which represent the 9 coordinated tri-capped trigonal prism (TTP) and the 10 coordinated Archimedean antiprism, respectively. These agree well with the results obtained by other simulation method [11] and with our previous results [12].

The sum of B-centered prism-like VPs including $<0, 3, 6, 0>$ and $<0, 2, 8, 0>$ in Fe80Si10B10 melt (25.0%) is close to the corresponding value (24.5%) in Fe78Si9B13 melt [12]. As it is not difficult for $<0, 2, 8, 0>$ polyhedron to transform to $<0, 3, 6, 0>$ polyhedron, this fact reveals that the local environments around B atoms are similar at various B contents. Of course, the absolute amount of prism-like environments for B-centered VPs in Fe78Si9B13 should be larger than that in Fe80Si10B10 melt.

From the effective cluster packing model [13,14], the preferred VP type is controlled by the effective atomic size ratio between the solute and solvent atoms, $R^*$. Of course,
\(\{0, 3, 6, 4\}\) and its derivatives including \(\{0, 2, 8, 2\}\) and \(\{0, 2, 8, 1\}\) can also considered as the deformed icosahedral ordering. With decreasing \(R^\prime\), the preferred VP type changes from the 12 coordinated deformed icosahedral type \((R^\prime \approx 0.902, \text{ for Fe-Si with } R^\prime = 0.929)\) [15], and then to the TTP type \((R^\prime \approx 0.732, \text{ as in Fe-B with } R^\prime = 0.661)\). The atom sizes are obtained from reference [16]. Here, the packing cluster type shows agreement with the change of \(R^\prime\).

The results from Voronoi polyhedra analysis have suggested that Si has the same local environment with Fe, which is consistent with our previous results [12]. The dissolved Si atoms slightly change the initial bcc-like dominating structure of Fe matrix, and lead the Si-centered VPs to preferring the bcc-like structure, which damages the stability of alloy melt. The B atoms are mostly located at the center of prism-like VPs including \(\{0, 3, 6, 0\}\) and \(\{0, 2, 8, 0\}\), which is fully consistent with the structural model for MGs proposed by Bernal [17]. Therefore, the structure of \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\) can be described as B-centered prism-like clusters embedded in a bcc-like Fe–Si solid solution.

The details of neighboring information between various species through the definition of chemical short range order (CSRO) in Ref. [18] are listed in Table 1. For an M-centered species through the definition of chemical short range order bcc-like Fe–Si solid solution.

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The details of neighboring information between various species through the definition of chemical short range order (CSRO) in Ref. [18] are listed in Table 1. For an M-centered CSRO in which other same M atoms can be found, it is called S-type CSRO. If the atoms in the nearest-neighbor shell are all Fe atoms, then this CSRO is called a P-type CSRO.

According to the plot of critical ribbon thickness against Fe–Si–B composition offered by Masumoto [19], the critical thickness of \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{13}\) is at least 3 times as large as that of \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\). So, there are probably three causes resulting in this difference.

For \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\) melt, it has higher Si and lower B content than \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{13}\) melt. More importantly, around Si and B atoms are bcc-like and prism-like environments, which are unfavorable and favorable for the melt stability, respectively.

Furthermore, the pure Fe clusters account for 3.5\%, which is larger than that of \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{13}\) (0.6\%). According to the viewpoint [22] that when a type of clusters, such as the pure Fe clusters dominates in \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\), it will precipitate first from amorphous state under heat treatment, which will decrease the glass-forming ability (GFA).

The poor solute–solute avoidance for Si and B atoms may also play an important role for the stability of Fe–Si–B alloy series. According to the results of Table 1, the fraction of S-type CSROs for Si-centered and B-centered in \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\) is larger that of \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{13}\). This fact suggests that the solute–solute avoidance for the former is poor, which is unfavorable for the stability of alloy melt [18].

The structure of \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\) in amorphous state is similar to that of liquid melts, except the predictable splitting of the second peaks in PDFs originating from the greater correlation length. To calculate the GFA of MGs, i.e., the frozen metallic liquids, the crystallization tendency \((\Delta T_c = T_c - T_h)\) is often used [23,24]. So, when heated from amorphous state, this retained bcc-like structure is inherently similar to and should be inclined to transform to bcc phases including FeSi, Fe(Si) and \(\alpha\)-Fe(Si) [21,25], which promotes the crystallization of Fe–Si–B MGs and reslutantly damages the GFA. These simulations are consistent with the experimental phenomena: Fe–Si–B MGs are always precipitated the Si-contained bcc-type crystals first, even at different Si and B component ratios [21,25]. Furthermore, no supercooled liquid region appears in Fe–Si–B ternary alloys, demonstrating the poor stability of transformation from amorphous structure to a crystalline phase. Therefore, it is easier to understand the poor GFA of Fe–Si–B which can only be found in the ribbon form when taking these factors into account.

4. Summary

The structure of \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\) alloy melt was investigated by \textit{ab initio} molecular dynamic simulation. It is found that \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\) alloy can be considered as the combination of B-centered prism-like clusters and bcc-like Fe–Si solid solution. Compared with \(\text{Fe}_{78}\text{Si}_{9}\text{B}_{13}\), the worse GFA is resulted from the more bcc-like environment around Si atoms and the less prism-like environment around B atoms. The relative higher pure Fe clusters and poor solute–solute avoidance may also decrease the stability of \(\text{Fe}_{80}\text{Si}_{10}\text{B}_{10}\) alloy. These simulations are consistent with the reported experimental phenomena.

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References


