

Enhanced glass-forming ability of Fe-based bulk metallic glasses prepared using hot metal and commercial raw materials through the optimization of Mo content

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The glass-forming ability of Fe-based metallic glass $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ ($x = 0\text{--}6.5$ at.%), which can be produced in large quantities and cost-effectively using hot metal and commercial raw materials, was significantly enhanced by optimizing the Mo content, leading to the formation of a fully amorphous rod of 6 mm in diameter with high compressive fracture strength (~ 3.3 GPa). It is concluded that the enhanced glass-forming ability is attributed to the suppression of the primary α -(Fe, Mo) phase by the optimum amount of Mo substitution for Fe.

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Among bulk metallic glasses (BMGs) having attractive properties for structural and functional applications, Fe-based BMGs with high glass-forming ability (GFA) are of great interest [1–4]. Recently, Fe-based BMGs prepared at the laboratory scale using high-purity elements have been highlighted as potentially being of use in practical applications. Indeed, the Fe-based BMGs can be cast into amorphous rods of more than 1 cm diameter [5–8]. Significant GFA enhancement of the Fe-based BMGs has been achieved by the addition of small amounts of Y, Er or Tm [5–8], as well as the use of high-purity elements. However, alloy design strategies using high-purity raw materials and novel elements to produce high GFA may not be the most economical approach to developing Fe-based BMGs for the commercial markets. That is, for extensive practical applications of Fe-based BMGs, the BMGs need to be produced in large quantities and cost-effectively through relevant materials-processing routes. The productivity, unit cost and variety of materials processing needed to produce Fe-based BMGs can be limited by using novel and pure elements.

Some trials to fabricate cost-effective Fe-based BMGs using hot metal produced by blast furnace and industrial

raw materials by tuning the composition have been reported [9,10]. Basic alloy design strategies in the trials are to decrease the melting temperature of the Fe-based alloys by minor additions of metallic and metalloid elements to the hot metal so that the melt can be frozen in an amorphous state at a relatively low cooling rate. Meanwhile, since heterogeneous nucleation sites inevitably exist in the melt during commercial materials processing, finding elements that effectively suppress the formation of the primary phase upon solidification is more important in the alloy design for high GFA than finding elements that reduce the heterogeneous nucleation sites. In this study, based on this scenario, we develop Fe-based BMGs having high GFA in the alloy system Fe–C–Si–B–P–Cr–Al–Mo. Small amounts of Mo have been systematically substituted for Fe to enhance the GFA of the alloy $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ ($x = 0\text{--}6.5$). The effects of Mo substitutions on GFA and crystallization behaviors were discussed based upon the thermal and microstructural analysis results.

A series of $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ alloys with $x = 0, 2.5, 4.5$ and 6.5 were prepared using hot metal, industrial ferro-alloys and commercially pure Si, Mo, Al (>95%) through an arc-melting process under an argon atmosphere. The chemical compositions, including oxygen and nitrogen contents, of

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Table 1. Chemical compositions (wt.%) of raw materials used

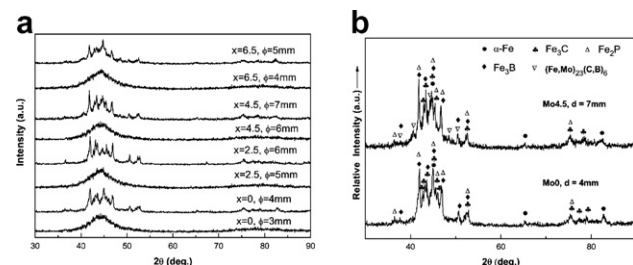
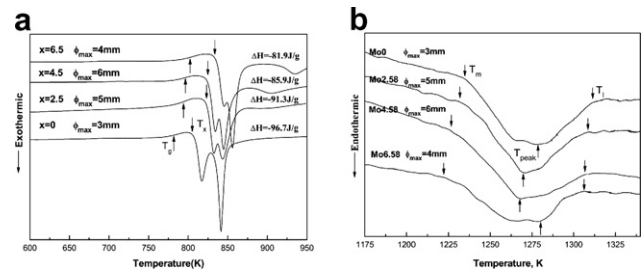
Materials	C	Si	P	B	Cr	Mn	Al	V	Cu	O	N
Hot metal	4.58	1.37	0.11	–	–	–	–	–	–	–	–
Fe–P	0.015	0.25	28.08	–	–	–	–	0.21	0.11	0.0334	0.0022
Fe–B	0.17	0.65	0.038	19.75	–	0.21	0.05	–	–	0.0156	0.002
Fe–Cr	1.43	0.325	0.016	–	55.94	–	–	–	–	0.2415	0.0131

*Commercial pure elements (Al, Si, Mo) > 95%

above-mentioned hot metal and ferro-alloys are shown in Table 1 in mass percentages. To evaluate the GFA of the alloys, a piece of ingot was levitation-melted and quenched into a splat with the thickness of 50–80 μm in a splat quencher. Also, the ingots were suction-cast into a rod using copper molds with cylinder cavities of 1–7 mm diameter and a height of around 50 mm. Microstructures of as-cast or annealed samples were investigated by X-ray diffraction (XRD) with Cu radiation ($\text{Cu K}\alpha$, $\lambda = 0.1541 \text{ nm}$) and transmission electron microscopy (TEM). Glass transition and crystallization of amorphous samples were examined by differential scanning calorimeter (DSC; Perkin Elmer Diamond) under an Ar atmosphere at the heating rate of 40 K min^{-1} . Mechanical properties of room temperature were measured with an Instron testing machine at a strain rate of 10^{-4} s^{-1} under a compression mode.

Figure 1 shows XRD patterns of suction-cast $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ alloys with $x = 0, 2.5, 4.5$ and 6.5 . It is well known that the GFA of an alloy increases as the maximum diameter of the fully amorphous rod of the alloy increases. The XRD patterns in Figure 1(a) reveal that a fully amorphous rod with a maximum diameter of 6 mm can be obtained with 4.5 at.% Mo substitution for Fe. That is, the GFA of the alloy $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ increases with x up to 4.5 and then decreases with an x of 6.5. Hence, it is feasible to conclude that the substitution of an optimum amount of Mo for Fe effectively enhances the GFA of the alloy $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$. The Bragg peaks obtained from rods with diameters larger than the maximum diameters of fully amorphous rods are attributed to the crystallization of many different phases up on solidification. It should be noted that $(\text{Fe, Mo})_{23}(\text{C, B})_6$ phase can be clearly indexed in the XRD pattern of the alloy with $x = 4.5$ but not in the case of the alloy with $x = 0$ (Fig. 1(b)).

Figure 2 shows DSC and differential thermal analysis (DTA) heating traces of fully amorphous rods with the maximum diameters of the alloys $\text{Fe}_{71.2-x}\text{C}_{7.0}$

**Figure 1.** The XRD patterns of suction-cast rods of $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ ($x = 0, 2.5, 4.5$ and 6.5).**Figure 2.** DSC and DTA heating traces of fully amorphous rods with the maximum diameters of the alloys $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ alloys with $x = 0, 2.5, 4.5$ and 6.5 .

$\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$. With the substitutions of Mo for Fe, the glass transition temperature (T_g) and the onset temperature (T_x) of the first crystallization event increases, indicating that the amorphous phase can be stabilized at high temperatures. Clear supercooled liquid regions, $\Delta T_x = T_x - T_g$, are observed in the DSC traces. The extent of the supercooled liquid region is often regarded as a parameter to assess GFA of an alloy. As summarized in Table 2, the alloys containing Mo present wider supercooled liquid regions than the alloy without Mo. This is coincident with the fact that the partial substitution of Mo for Fe improves the GFA of the $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ alloy, as inferred from the XRD results shown in Figure 1. However, it should be pointed out that the extents of supercooled regions remain around 29 K as the Mo content increases, while the maximum GFA was obtained in the alloy with 4.5 at.% Mo. Parameters T_{rg} and γ , which often used to estimate the GFA of an alloy, monotonically increase with the increase of Mo content, indicating that these parameters are not suitable for the GFA estimation of the $\text{Fe}_{71.2-x}\text{C}_{7.0}\text{Si}_{3.3}\text{B}_{5.5}\text{P}_{8.7}\text{Cr}_{2.3}\text{Al}_{2.0}\text{Mo}_x$ alloys. Rather, the Gibbs free energy difference between the liquid and solid phases tends to be in good agreement with the GFA changes of the alloys. The Gibbs free energy difference listed in Table 2 was obtained through thermodynamic calculations using the DTA heating traces shown in Figure 2(b). (Detailed calculation methods are explained elsewhere [10].)

To understand the change of crystallization behaviors with the Mo substitution for Fe, the splat-quenched ribbons with $x = 0$ and 4.5 were heated to temperatures at which the first exothermic event ends, i.e. 828 and 838 K, respectively, and immediately cooled to room temperature in DSC. The XRD pattern of the heat-treated alloy with $x = 0$ displays relatively sharp diffraction peaks, corresponding to a body-centered cubic phase (α -Fe) with the broad diffraction maxima typical for an amorphous phase (Fig. 3(a)). In the case of the alloy

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