



Glass-forming ability, fragility parameter, and mechanical properties of Co–Ir–Ta–B amorphous alloys



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ABSTRACT

The effect of partial substitution of Co by Ir in $\text{Co}_{57-x}\text{Ir}_x\text{Ta}_8\text{B}_{35}$ (at.%, $x = 0, 5, 10$, and 15) alloys on glass-forming ability (GFA), fragility parameter m , and mechanical properties was investigated. It was found that the glass-forming ability reflected by critical diameter for glass formation is in good agreement with the width of supercooled region (ΔT_x). The $\text{Co}_{47}\text{Ir}_{10}\text{Ta}_8\text{B}_{35}$ BMG exhibits the best GFA yielding the formation of an amorphous rod with a diameter of 5 mm, along with the largest ΔT_x value of 64 K. The Ir-bearing $\text{Co}_{47}\text{Ir}_{10}\text{Ta}_8\text{B}_{35}$ BMG presents a lower fragility parameter m than $\text{Co}_{57}\text{Ta}_8\text{B}_{35}$ BMG, which indicates better thermal stability and GFA. In addition, the $\text{Co}_{47}\text{Ir}_{10}\text{Ta}_8\text{B}_{35}$ BMG shows super-high yield strength of ~ 5200 MPa and ultimate compressive strength of ~ 5600 MPa as well as significant plasticity of $\sim 1.2\%$. The combination of good GFA, large ΔT_x and excellent mechanical properties for the new Co–Ir–Ta–B BMG is significant for scientific research and industrial application.

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1. Introduction

It is well known that bulk metallic glasses (BMGs) possess higher mechanical strength compared with similar conventional crystalline alloys, due to their disordered structures and structural uniformity [1–3]. Many efforts have been devoted to develop new BMGs with enhanced mechanical strength and glass-forming ability (GFA) for extending their industrial applicability in various fields [4–6]. Among different glass-forming systems, Fe- and Co–B-based BMGs containing high-modulus refractory metals M ($M = \text{Nb}, \text{Ta}, \text{Mo}$, etc.) usually exhibit extremely high mechanical strength, due to the nature of the strong bonding among the constituent elements as well as the unique network-like structure existing in the amorphous phase [7–9]. In recent years, a variety of Fe- and Co–B-based BMGs with superhigh fracture strengths (σ_f) over 4000 MPa have been successfully synthesized in (Fe,Co)–B–Si–Nb [10,11], (Fe,Co)–B–(Nb,Zr) [12–14], Fe–B–(Nb,Ta)–Y [15,16], (Co,Fe)–B–(Ta,Mo) [6,8], and Co–B–Si–Ta [17] alloy systems, and their maximum diameter was reported to reach 7 mm. It is noteworthy that the increment of B content seems to cause a large enhancement in mechanical strength in these glassy alloys. A few Co-based BMGs with B content more than 30 at.% have been discovered by Inoue et al. [6,8], such as $\text{Co}_{43.5}\text{Fe}_{20}\text{Ta}_{5.5}\text{B}_{31.5}$ and $(\text{Co}_{0.535}\text{Fe}_{0.1}\text{Ta}_{0.055}\text{B}_{0.31})_{98}\text{Mo}_2$, of which the σ_f was significant increased to over 5000 MPa. Very recently, we

developed a new family of Co–Ta–B BMGs with higher B content up to 35 at.%. The σ_f of these ternary Co-based BMGs is in the range of 5500–6000 MPa which is the highest value found among all BMGs reported so far [18]. Similar to the other ductile high-strength Fe- and Co-based BMGs [11,13,14], these BMGs also exhibit significant plastic strains of 0.5–1.5%. Nevertheless, the critical sizes of glass formation for the high-B Co–Ta–B alloys are only about 1–2 mm in diameter. It is hence essential to further improve their GFA, so that they are viable both for the engineering applications and fundamental studies.

In the present research, the glass former $\text{Co}_{56}\text{Ta}_8\text{B}_{35}$, which is capable of forming a 2-mm fully amorphous rod, was chosen as a starting alloy composition. Ir element in the same group as Co in the Periodic Table of the Elements was selected to be a candidate as the fourth alloying element to partially substitute for Co. It is commonly known that elements in the same group usually show similar valence electronic structures but different atomic sizes to each other. Such a method of element substitution is known as “column substitution” [19], which is derived from the empirical rules for high GFA, e.g. atomic size mismatch and negative mixing enthalpy. Acknowledged practices of column substitution include partial substitution of Sc for solvent Y in (Y–Sc)–Al–Co [20], Mg for solvent Ca in (Ca–Mg)–Ag [21], Ag for solute Cu in Mg–(Cu–Ag)–Y and Mg–(Cu–Ag)–Ni–Zn–Y [22,23], and Ti for solvent and solute Zr in (Zr–Ti)–Cu–Ni–Be and Cu–(Zr–Ti) [24,25], respectively. For the (Co,Ir)–Ta–B alloy system, the continuous change in atomic sizes in the order of $\text{B} < \text{Fe} < \text{Ir} < \text{Ta}$ and large negative mixing

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enthalpies among the constituent elements (i.e. the Ir–Co, Ir–B, and Ir–Ta are –3, –11 and –52 kJ/mol, respectively) caused by the partial substitution of Ir for Co indicate their high GFA. On the basis of the above analysis, we investigated in detail the GFA and thermal properties of $\text{Co}_{57-x}\text{Ir}_x\text{Ta}_8\text{B}_{35}$ ($x = 0, 5, 10$, and 15). It was found that the new $\text{Co}_{47}\text{Ir}_{10}\text{Ta}_8\text{B}_{35}$ BMG with a critical diameter of 5 mm as well as a wide supercooled liquid region up to 64 K can be synthesized by copper mold casting. Furthermore, the fragility parameter m and mechanical properties for the Co–Ir–Ta–B BMGs were discussed.

2. Material and methods

A Co–B pre-alloy with B content of 40 at.% was prepared by induction melting pure Co (99.9 mass%) and pure B (99.99 mass%) in a high purity argon atmosphere. Master alloys with nominal compositions of $\text{Co}_{57-x}\text{Ir}_x\text{Ta}_8\text{B}_{35}$ (at.%, $x = 0, 5, 10$, and 15) were then prepared by arc melting the mixings of pure Co, Ta (99.99 mass%) metal and Co–B pre-alloy in a Ti-gettered high purity argon atmosphere. The master alloy ingots were remelted several times to ensure homogeneity. From these ingots, alloy ribbons with a cross section of 0.02×1.5 mm were prepared by the single roller melt-spinning method. Cylindrical rods with different diameters ranging from 1 to 5 mm were produced by injection mold casting method.

The amorphous structure of as-cast rods and as-spun ribbons was determined by X-ray diffraction (XRD) using a Dmax2200PC Rigaku X-ray diffractometer with Cu K α radiation. Thermal analysis was carried out using a NETZSCH 404 C differential scanning calorimetry (DSC) under a continuous argon flow. The value of fragility parameter m was calculated using the effective activation energy and Vogel–Fulcher–Tammann parameters, which can be described based on the dependence of glass-transition temperature on heating rate [26–28]. For the analysis of fragility parameter, a series of DSC scans related to the glass-transition behavior were obtained at different heating rates ranging from 0.083 to 0.83 K/s. Uniaxial compression testing was conducted at an ambient temperature using mechanical testing machine at a strain rate of $4.2 \times 10^{-4} \text{ s}^{-1}$. The testing samples with a gauge aspect ratio of 2:1 were cut from the as-cast 1 mm cylindrical rods. The compression tests were performed at least in triplicate for each BMG. Fracture morphologies were observed with JEOL 7600 scanning electron microscopy (SEM).

3. Results and discussion

3.1. Glass-forming ability and thermal properties

Fig. 1 shows the XRD patterns for the as-cast $\text{Co}_{57-x}\text{Ir}_x\text{Ta}_8\text{B}_{35}$ (at.%, $x = 0, 5, 10$, and 15) glassy alloy rods with their critical diameters for glass formation (d_c). Only a broad diffuse peak without distinct crystalline peaks can be observed for each pattern, indicating that these samples consist of fully amorphous structure. Therefore, the critical diameters of glass rods, which is commonly used to evaluated the glass-forming ability (GFA) of amorphous alloys, are 2, 3, 5, and 3 mm for alloys with Ir addition of $x = 0, 5, 10$, and 15 at.%, respectively. The present results demonstrate that

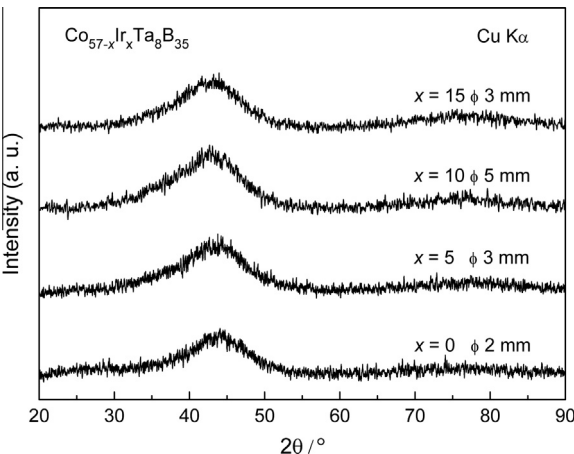


Fig. 1. XRD patterns of $\text{Co}_{57-x}\text{Ir}_x\text{Ta}_8\text{B}_{35}$ (at.%, $x = 0, 5, 10$, and 15) BMGs.

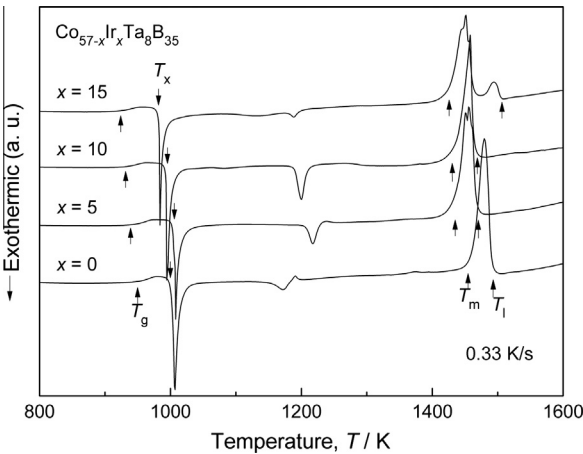


Fig. 2. DSC curves of $\text{Co}_{57-x}\text{Ir}_x\text{Ta}_8\text{B}_{35}$ (at.%, $x = 0, 5, 10$, and 15) amorphous alloys.

the introduction of Ir with proper amounts is effective in improving the GFA of the Co–Ta–B alloys.

Fig. 2 shows the DSC curves of $\text{Co}_{57-x}\text{Ir}_x\text{Ta}_8\text{B}_{35}$ (at.%, $x = 0, 5, 10$, and 15) glassy alloys at a heating rate of 0.33 K/s. As shown in Fig. 2, each trace of scan exhibits an endothermic event characteristic of the glass transition and a supercooled liquid region, followed by two characteristic exothermic events of crystallization processes. The glass-transition temperature (T_g) gradually decreases from 949 K to 925 K with the increase of Ir content from 0 to 15 at.%. The onset temperature of crystallization (T_x) slightly increases from 1001 K to 1002 K as Ir content increases from 0 to 5 at.%, and then decreases to 925 K with an increase of Ir content up to 15 at.%. The melting behaviors of these Co-based alloys are also shown in Fig. 2. It can be seen that the melting temperature (T_m) shifts toward a relatively lower temperature region with the addition of Ir. The liquidus temperature (T_l) decreases with increasing Ta content, reaching a minimum value of 1461 K at 10 at.% of Ir, and then followed by a remarkable increase with the further increase of Ta content. The characteristic temperatures including T_g , T_x , T_m , and T_l are listed in Table 1.

Based on the characteristic temperatures, the glass-forming criteria, e.g. the width of supercooled liquid region ($\Delta T_x = T_x - T_g$) [29], the reduced glass transition temperature ($T_{rg} = T_g/T_l$) [30], and γ parameter [$\gamma = T_x/(T_g + T_l)$] [31], were calculated. These parameters as well as d_c are also summarized in Table 1. As presented in the table, ΔT_x increases from 52 K to 64 K with an increase of Ir contents from 0 to 10 at.%, and then decreases to 57 K with a further addition of Ir content up to 15 at.%. Generally, a larger ΔT_x implies a higher resistance against crystallization, and therefore enable the production of BMG with a larger d_c . In the present Co–Ir–Ta–B alloys, the trend of change for ΔT_x is in good agreement with the experimental results of GFA reflected by d_c . In addition, T_{rg} and γ parameter can also be increased by appropriate addition of Ir

Table 1
The critical diameters (d_c) and thermal parameters for $\text{Co}_{57-x}\text{Ir}_x\text{Ta}_8\text{B}_{35}$ (at.%, $x = 0, 5, 10$, and 15) BMGs, including the glass transition temperature (T_g), onset temperature of crystallization (T_x), melting (T_m) and liquidus temperature (T_l), together with supercooled liquid region, supercooled liquid region ($\Delta T_x = T_x - T_g$), reduced glass transition temperature ($T_{rg} = T_g/T_l$), and γ parameter [$\gamma = T_x/(T_g + T_l)$].

Alloys	d_c (mm)	T_g (K)	T_x (K)	T_m (K)	T_l (K)	ΔT_x (K)	T_{rg}	γ
$\text{Co}_{57}\text{Ta}_8\text{B}_{35}$	2	949	1001	1461	1492	52	0.636	0.410
$\text{Co}_{52}\text{Ir}_5\text{Ta}_8\text{B}_{35}$	3	941	1002	1437	1469	61	0.641	0.416
$\text{Co}_{47}\text{Ir}_{10}\text{Ta}_8\text{B}_{35}$	5	929	993	1430	1461	64	0.636	0.415
$\text{Co}_{42}\text{Ir}_{15}\text{Ta}_8\text{B}_{35}$	3	925	982	1425	1505	57	0.615	0.404

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