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Co dependence of Curie temperature in amorphous Fe–Co–Zr–B–Nb alloys with high glass-forming ability

B Yao[1,2], Y Zhang[2], L Si[2], H Tan[2] and Y Li[2]

[1] Department of Physics, Jilin University, Changchun 130023, People's Republic of China
[2] Department of Material Science, Faculty of Science, National University of Singapore, Singapore 117543, Singapore

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Abstract

Effects of Co substitution for Fe on the Curie temperature (T_c), glass-forming ability (GFA) and thermal stability of amorphous $Fe_{61-x}Co_xZr_5B_{30}Nb_4$ (FCZBN) alloys were studied for Co content ranging from 0 to 15 at. %. The T_c shows a sinusoid-like behaviour with increasing Co content, revealing two maxima at 3 and 12.5 at. % Co and a minimum at 7.5 at. % Co. Co content dependences of glass transition (T_g), crystallization (T_x) and reduced glass transition temperatures (T_{rg}) of the amorphous alloys are almost completely opposite to that of the T_c. The T_c decreases with increasing T_g and T_{rg}, but increases with increasing Co content. The Co content dependence of the T_c is suggested to relate to both Co content and high GFA of the amorphous alloys.

1. Introduction

Effects of Co substitution for Fe on thermal stability, glass-forming ability (GFA) and magnetic properties of Fe-based amorphous alloys have been studied widely, owing to excellent magnetic properties of the amorphous alloys. It was found that the effect of substitution of Co for Fe on the Curie temperature T_c was different for various Fe-based amorphous alloys. For some Fe-based amorphous alloys, this substitution makes the T_c either increase or decrease monotonically with increasing Co content [1–5]. For other Fe-based amorphous alloys, the T_c firstly increases up to a maximum with increasing Co content and then decreases [6–9]. The effects of this substitution on the T_c are usually attributed to changes of atomic moments of Fe and Co and/or exchange interactions of Fe–Fe, Fe–Co and Co–Co atomic couples [1–9].

It is well known that exchange interaction is related to species and interatomic distance of the nearest-neighbour magnetic atoms in amorphous alloys, based on the approximation of nearest-neighbour atoms. Since the density of random-packed atoms determines the interatomic distance, it should affect the exchange interaction of magnetic atoms in a magnetic amorphous alloy. For conventional Fe–Co based amorphous alloys, variation in exchange interaction induced by change of interatomic distance may be much smaller than that induced by change of magnetic atomic couples when the Fe atom is replaced by the Co atom, and hence

the effect of the interatomic distance on the T_c is not obvious. However, amorphous alloys with higher GFA, glass transition (T_g) and crystallization (T_x) temperatures, have much denser random-packed microstructures than the conventional amorphous alloys [10–13], leading to larger variation in the interatomic distance. This larger variation will cause a considerable influence on the exchange interaction, so that the effect of interatomic distance on the T_c becomes obvious.

Recently, it was reported that the GFA of Fe–Zr–B-based alloy could be greatly improved by the addition of some transition metals, such as Co, Ni, Ta, Nb, W, Mo, etc [14–17], so that some bulk Fe–Zr–B-based amorphous alloys with high GFA and thermostability were prepared at low cooling rate. However, their magnetic properties were different [14, 15]. Some of them are ferromagnetic at room temperature, while others are paramagnetic, which seems to be related to their GFA [15–17]. Thus, it is necessary to study correlations between GFA, T_g and T_c. However, there have been no detailed studies of the correlations so far.

In this paper, effects of Co substitution for Fe on GFA, thermal stability and magnetic properties were studied for the amorphous Fe–Zr–B–Nb alloys, and correlations between thermostability, GFA and magnetic properties were investigated in detail.

2. Experimental procedures

Multicomponent alloys with nominal atomic ratios of $Fe_{61-x}Co_xZr_5B_{30}Nb_4$ (FCZBN) ($x = 0$–15) were prepared by arc-melting Fe, Co, Zr, B and Nb with purity ranging from 99.5 to 99.98% in a titanium-gettered argon atmosphere. Amorphous FCZBN ribbons (about 1.5 mm in width and 25 μm in thickness) were prepared by single roller melt-spinning technique in an argon atmosphere. The microstructure of the ribbons was characterized by x-ray diffractometry (XRD) with Cu K_α radiation and transmission electron microscopy (TEM) to be fully amorphous alloys. The onset melting (T_m) and liquidus (T_l) temperatures of the FCZBN alloys were determined by a differential temperature analyser at a heating rate of 20 °C min^{-1}. Measurements of T_g, T_x and T_c were performed by differential scanning calorimetry (DSC) at a heating rate of 20 °C min^{-1} in a flowing argon atmosphere. Apparent activation energy was measured by means of DSC at heating rates of 2.5, 5, 10, 20 and 40 °C min^{-1}. The T_c was also confirmed by measuring the change in magnetization with temperature using a vibrating sample magnetometer (VSM) at a heating rate of 0.1 °C min^{-1} in an external field of 500 Oe. Magnetization of the samples was measured by the VSM.

3. Results and discussion

Since transition from a ferromagnetic to paramagnetic phase is a second-order phase transformation, an inflection appears at the T_c in each DSC curve of the amorphous FCZBN alloys, as shown in figure 1. The T_c of the amorphous FCZBN alloy with 7.5 at. % Co was also measured by VSM to be 241 °C, which is 11 °C higher than the T_c measured by DSC. However, when the effect of the external magnetic field on the measured value of the increase in T_c is taken into account, the T_c values measured by DSC and VSM are in agreement. As shown in figure 2(a), the T_c shows a sinusoid-like changing behaviour with increasing Co content. It increases initially with increasing Co content up to 3 at. %, then decreases to a minimum at 7.5 at. %, and increases again until the Co content reaches 12.5 at. % and finally decreases with increasing Co content up to 15 at. %. This tendency is very different from Co-content dependences of the T_c observed in conventional amorphous Fe–Co–Zr–B–Nb alloys with low B content [1, 5] and other Fe–Co-based amorphous alloys [6–9, 18]. Figure 2(b) reveals

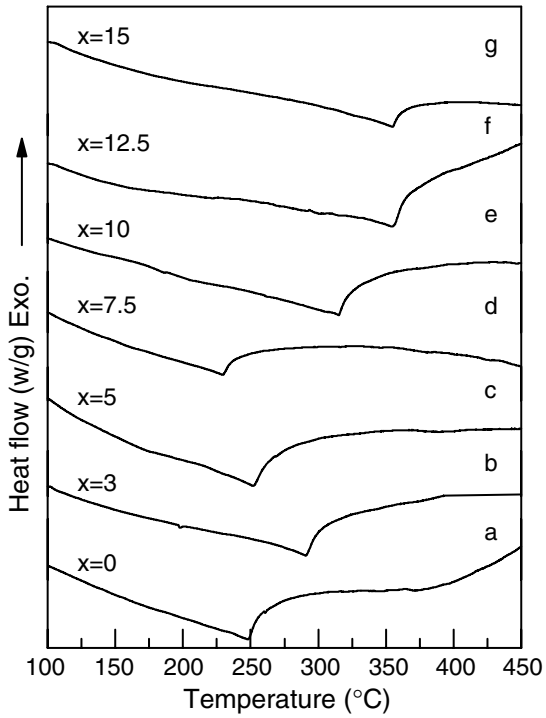

Figure 1. DSC curves for the measurement of the Curie temperature of the as-spun amorphous FCZBN alloys with various Co contents.

Co-content dependence of magnetizations of the amorphous FCZBN alloys measured at room temperature (M_{RT}), which is similar to that of the T_c, implying that the Co-content dependence of the T_c measured by DSC is correct. T_g, T_x and reduced glass transition temperature T_{rg} ($T_{rg} = T_g/T_l$) versus Co content plots are shown in figure 2(c). It is well known that T_{rg} is usually used to characterize GFA [19–21] of amorphous alloys, while T_g and T_x are used to characterize their thermal stability. Figure 2(c) indicates that the Co-content dependence of thermal stability is similar to that of GFA in the amorphous FCZBN alloys.

It is known that a very small amount of crystallite in an amorphous alloy may significantly affect the physical properties of the amorphous alloy, such as thermal stability and magnetic properties, etc. Although no crystallite is detected by XRD and TEM in the amorphous FCZBN ribbons, it is still necessary to study the effect of volume fraction transformed on the Curie temperature to confirm that the Co-content dependence of the T_c is due to instinctive properties of the amorphous FCZBN alloys or due to the effect of a very small amount of crystallite that was not detected. An amorphous FCZBN alloy with 5 at. % Co was run repeatedly in DSC in the following way: heating at 20 °C min^{-1} from room temperature to 640 °C, then isothermally annealing for 5 min at this temperature and finally uncontrolled cooling to room temperature. Since the temperature 640 °C is located in a supercooled liquid region ΔT_x ($\Delta T_x = T_x - T_g$), the amorphous FCZBN alloy crystallizes very slowly and its volume fraction transformed increases gradually and slowly after each run. Figures 3(a)–(e) show typical DSC curves of amorphous FCZBN alloys recorded at the first, second, fourth, sixth and eighth runs respectively, indicating that after all runs the Curie temperature of the amorphous FCZBN alloy changed less than 1 °C, except after the first run when it increased 7 °C, which is attributed to relaxation and

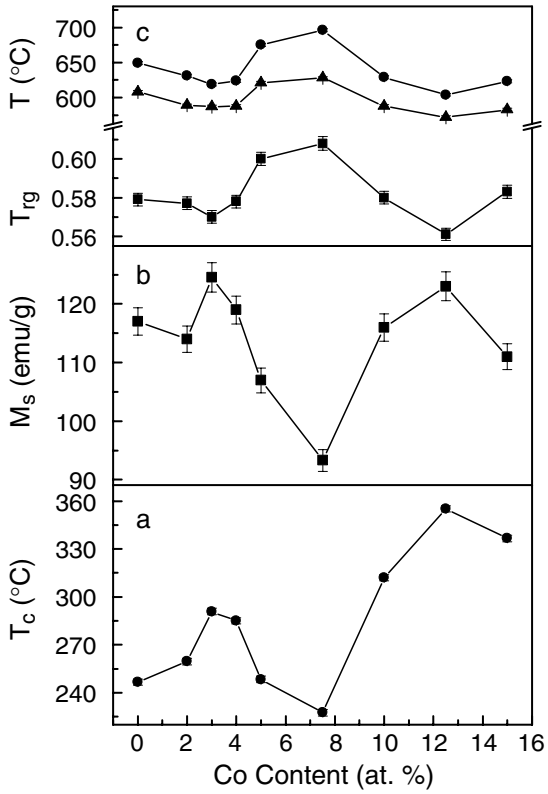

Figure 2. Variations of T_c (a), magnetization (b) and T_g, T_x and T_{rg} (c) of the amorphous FCZBN alloys as a function of Co content. ▲ T_g; ● T_x; ■ T_{rg}.

increase of chemical short-range order in the amorphous FCZBN alloy. However, intensity of the inflection at Curie point decreased slowly with increasing run times. Figures 4(a)–(d) show XRD profiles of the as-spun FCZBN ribbon and the ribbon run after the third, fifth and seventh times respectively. Obviously, the amorphous FCZBN alloy crystallizes gradually in a eutectic mode to α-Fe (Co) and a Fe–Co–Zr–Nb–B phase, the structure of which is not yet identified. Its volume fraction transformed increases with increasing run times, whereas the volume fraction of the amorphous FCZBN alloy decreases and becomes very small after the seventh time. On comparison of figures 3 and 4, we infer that the intensity of the inflection decreases on decreasing the volume fraction of the amorphous alloy (or increasing the volume fraction transformed), and becomes very weak when the volume fraction of the amorphous alloy is very small. However, the Curie temperature changes very little. The above results indicate that the Curie temperature of the amorphous FCZBN alloys is not affected by the volume fraction transformed. The Co-content dependence of the T_c, shown in figure 2(a), is a natural property of the amorphous FCZBN alloy. The same conclusion was also obtained in the amorphous FCZBN alloy with 3, 7.5 and 12.5 at. % Co. In addition, it was also found that T_g and T_x do not change with the volume fraction transformed for the amorphous FCZBN ribbons.

It is well known that the Curie temperature depends on the atomic moment and exchange interaction of magnetic atoms. However, it was reported that the average atomic moments

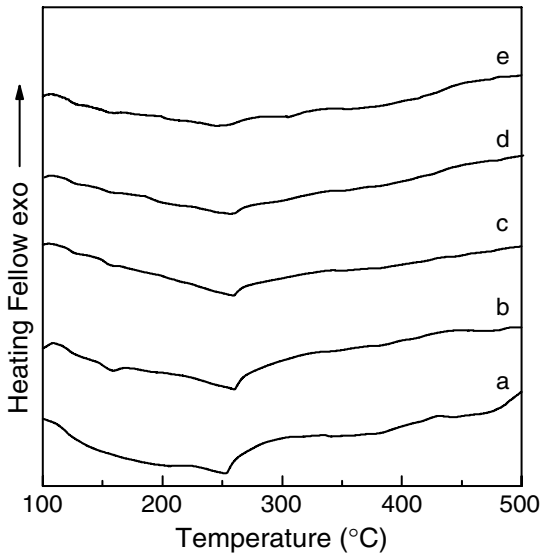

Figure 3. DSC curves of the amorphous FCZBN alloy with 5 at. % Co recorded at the first (a), second (b), fourth (c), sixth (d) and eighth (e) runs.

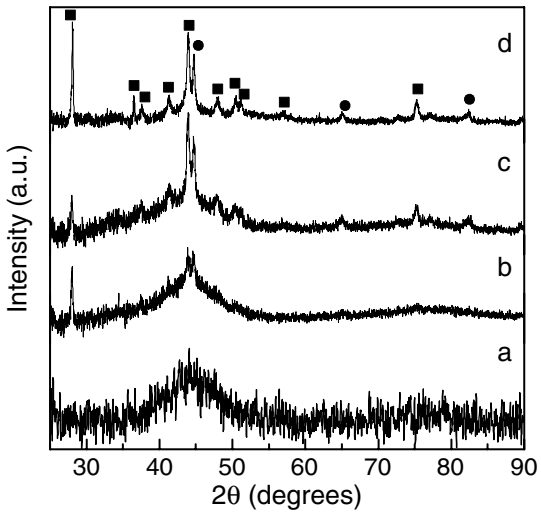

Figure 4. XRD profiles of the amorphous FCZBN alloy with 5 at. % Co (a) and the amorphous alloy run after three (b), five (c) and seven times (d). ● α-Fe (Co); ■ Fe–Co–Zr–B–Nb.

of Fe and Co atoms change little in Co content; less than 20 at. % for Fe-rich Fe–Co-based amorphous alloys [1–4]. Therefore, the variation of T_c is dominated by exchange interaction in the amorphous FCZBN alloys.

Previous studies have reported that the T_c increases monotonically with increasing Co content in Fe–Co–Zr–B–Cu and Fe–Co–Zr–B–Nb–Cu amorphous alloys with low B content, which has been attributed to the fact that exchange interactions of Co–Co and Fe–Co atomic couples are stronger than those of the Fe–Fe atomic couple [1, 5]. However, this mechanism

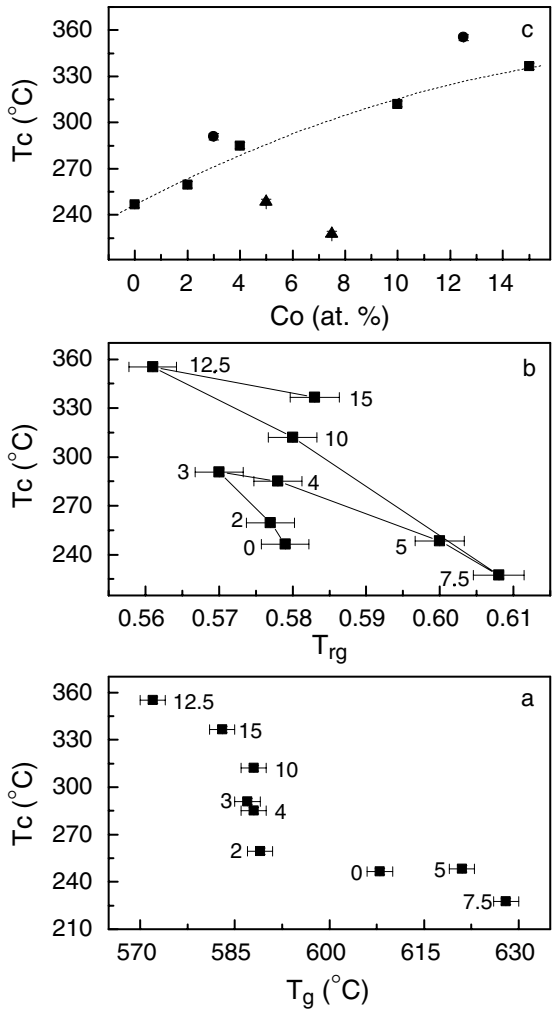

Figure 5. Plots of T_c as a function of T_g (a), T_{rg} (b) and Co content (c) for the amorphous FCZBN alloys. The number beside each symbol in (a) and (b) represents Co content of the amorphous FCZBN alloy corresponding to the symbol, and in (c) ■, represents the amorphous FCZBN alloys with close T_{rg}.

hardly explains the change of T_c observed in the present experiment. Therefore, the T_c should be affected by additional factors besides exchange interactions of Co–Co, Fe–Co and Fe–Fe atomic couples in the amorphous FCZBN alloys.

Comparison of figures 2(a) and (c) shows that the Co-content dependence of the T_c is almost completely opposite to that of T_g, T_x and T_{rg}, which implies that some correlation may exist between the T_c and T_g, T_x and T_{rg} in the amorphous FCZBN alloys. Figure 5(a) shows a T_c versus T_g plot of the amorphous FCZBN alloys, the number beside each symbol represents Co content of the amorphous FCZBN alloy corresponding to the symbol. It indicates that the T_c decreases with increasing T_g. For amorphous alloys with close values of T_g, such as the amorphous alloys with Co contents of 2, 3, 4 and 10 at. %, T_c increases with increasing Co content. Figure 5(b) shows a plot of T_c as a function of T_{rg},

revealing that the T_c decreases with increasing T_{rg} in each of the four content ranges 0–3, 3–7.5, 7.5–12.5 and 12.5–15 at. % Co. But the decreasing rate of the T_c is different for different Co-content ranges. The rate is higher for Co contents ranging from 0 to 3 at. % and 7.5 to 12.5 at. %, for which T_{rg} increases with decreasing Co content, compared with Co contents of 3 to 7.5 at. % and 12.5 to 15 at. %, for which T_{rg} increases with increasing Co content. For the amorphous FCZBN alloys with close values or the same T_{rg}, such as the amorphous alloys with Co contents of 0, 2, 4, 10 and 15 at. %, as shown in figure 5(b), the T_c (indicated by ■ in figure 5(c)) increases with increasing Co content monotonically, similar to previously reported results [1, 5]. By fitting the points marked by the solid squares, a T_c versus Co-content curve, shown by a broken line in figure 5(c), was obtained. It gives a tendency T_c with Co content for amorphous alloys having T_{rg} of about 0.58. It suggests that the T_c increases monotonically with increasing Co content for amorphous FCZBN alloys with the same or close values T_{rg}. However, for amorphous FCZBN alloys with T_{rg} less than 0.58, such as the amorphous FCZBN alloys with Co content of 3 or 12.5 at. %, the T_c (denoted by ● in figure 5(c)) is always higher than the Curie temperature (T_c') evaluated from T_c versus Co-content curve at its corresponding Co content, such as 3 or 12.5 at. % Co. For the amorphous FCZBN alloys with T_{rg} much larger than 0.58, such as the amorphous FCZBN alloy with Co content of 5 or 7.5 at. %, T_c (denoted by ▲ in figure 5(c)) is far less than T_c' evaluated from T_c versus Co-content curve at the same Co content, such as 5 or 7.5 at. % Co. These results indicate that the T_c decreases with increasing T_{rg} for amorphous FCZBN alloys under the same Co content. It was also found from figure 5(c) that some amorphous FCZBN alloys with higher Co content and T_{rg} have less T_c than those with lower Co content and T_{rg}, while some amorphous FCZBN alloys with higher Co content and T_{rg} have a larger T_c compared with those having lower Co content and T_{rg}. For example, the T_c of the amorphous FCZBN alloy with 7.5 at. % Co and T_{rg} of 0.608 is less than that of the amorphous FCZBN alloys with Co content less than 5 at. % and T_{rg} less than 0.58, while T_c of the amorphous FCZBN alloy with 10 at. % Co and T_{rg} of 0.58 is larger than that of the amorphous FCZBN alloy with 3 at. % Co and T_{rg} of 0.57, as shown in figure 5(c). In the former, T_{rg} dominates T_c, implying that T_{rg} has more effect on the T_c than Co content. While in the latter, Co content dominates T_c, implying that Co content has more influence on T_c than T_{rg}. On the basis of the analysis mentioned above, it is concluded that the T_c of the amorphous FCZBN alloys is determined by Co content and T_{rg}, and that the T_c increases with increasing Co content but decreases with increasing T_{rg}. Co-content dependence of the T_c is determined by a competition of effects of Co content and T_{rg} on it.

It was reported that T_g, T_x and GFA are related to the atomic interaction and the dense random-packed structure of amorphous alloys [10–13]. Many experimental results have demonstrated that an amorphous alloy with high T_g, T_x and GFA has highly dense random-packed structure or high density [10–13]. Thus, it can be deduced from figure 2(c) that the amorphous FCZBN alloy with high T_{rg} should have a highly dense random-packed structure and the amorphous FCZBN alloy with 7.5 at. % Co should have the largest density. This can be attributed to the negative heats of mixing of Co–Zr and Co–Nb, which are much larger than those of Fe–Zr and Fe–Nb and differences in heat of mixing between Co–Zr(Nb) and Fe–Zr(Nb) change with atomic ratios of Fe and Co to Zr and Nb and reach maxima at atomic ratios of 1 : 1 to 1.5 : 1 [22]. For amorphous FCZBN alloys without Co, the atomic ratio of Fe to the number of B and Fe is about 2 : 3, and so, on average six Zr and Nb atoms should become nearest-neighbour atoms of Fe. Co usually occupies the position of Fe in Fe–Zr (or Nb) clusters to form Co–Zr(Nb) clusters upon the substitution of Co for Fe, and the atomic ratio of Co to Zr and Nb is 1.25 : 1 in the amorphous FCZBN alloy with 7.5 at. % Co. Therefore, the amorphous FCZBN alloys should have the largest negative heat of mixing on replacement of Fe by 7.5 at. % Co, and

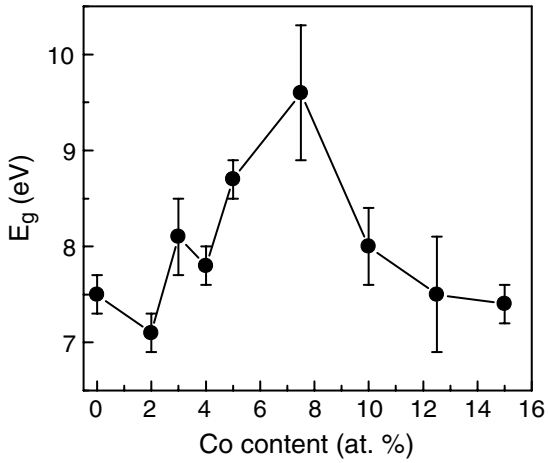

Figure 6. Plot of the apparent activation energy of glass transition of the amorphous FCZBN alloys as a function of Co content.

so have the densest random-packed structure. To confirm the above qualitative analysis, apparent activation energies of glass transition (E_g) of the amorphous FCZBN alloys were measured by using Kissinger's method [23], and were plotted in figure 6 as a function of Co content. Figure 6 shows that the Co dependences of E_g are similar to those of T_{rg} in figure 2(c). It is known that E_g is related to the atomic diffusion ability of amorphous alloys near glass transition [24–26]. The higher the apparent activation energy, the more difficult the atomic diffusion, implying smaller atomic free volume and a denser atomic random-packing of the amorphous alloy [27, 28]. Therefore, the results of both figures 2(c) and 6 indicate that the amorphous FCZBN alloy with higher T_{rg} has a denser atomic random-packed structure, in agreement with experimental results reported previously [10–13, 29]. The density of the amorphous FCZBN alloy is much higher near a Co content of 7.5 at. % compared with other Co contents. As such, the interatomic distance of the Fe–Fe pair in the amorphous FCZBN alloys decreases with increasing T_{rg} and greatly decreases near 7.5 at. % Co.

Previous studies have demonstrated that the exchange interaction of Fe–Fe atomic pairs in Fe-rich Fe-based amorphous alloys decreases as the interatomic distance of an Fe–Fe couple reduces [30–32], resulting in a decrease of T_c. A similar phenomenon was also observed in the amorphous $Fe_{91-x}Zr_5B_xNb_4$ (FZBN) [33], an alloy system similar to the amorphous FCZBN used in this study. For the amorphous FZBN alloys, the T_c increases on increasing the B atoms (or decreasing Fe content). This is due to the increase in interatomic distance of Fe–Fe pair caused by decrease in Fe content and incorporation of B atoms into the framework composed of Fe, Zr and Nb [34–37], which results in increase of exchange interaction of Fe–Fe pair with increasing B content. Therefore, the interatomic distance of Fe–Fe pair has an important influence on the exchange interaction in the Fe–Zr–B–Nb alloy system. For the amorphous FCZBN alloys, as mentioned above, interatomic distance of the Fe–Fe pair decreases with increasing T_{rg}. Therefore, if only the exchange interaction of the Fe–Fe couples is taken into account, the T_c should decrease with increasing T_{rg} in the amorphous FCZBN alloys and reach a minimum in the amorphous alloy with Co content of 7.5 at. %.

On the basis of the above analysis, it is concluded that the Co-content dependence of the T_c is determined by the competition of two kinds of changing behaviours of T_c; one is that the T_c increases with increasing Co content, which is attributed to the fact that the exchange

interactions of Fe–Co and Co–Co atomic couples are stronger than that of the Fe–Fe couple [1, 5]; and another is that the T_c decreases with increasing T_{rg} or GFA, which is due to the decrease in exchange interaction of the Fe–Fe couple with decreasing interatomic distance of the Fe–Fe couple. Amorphous FCZBN alloys with Co contents of 0, 2, 4, 10 and 15 at. % and with values the same or close to T_{rg} of 0.58, have close interatomic distance of Fe–Fe pairs. Consequently, T_c behaviour is determined by Co content and increases with increasing Co content. However, for the amorphous FCZBN alloys with Co content of 3 and 12.5 at. %, T_{rg} is less than 0.58, and so they should have a larger interatomic distance of the Fe–Fe couple compared with those for T_{rg} of 0.58 under the same Co content, resulting in them having a higher T_c compared with those having T_{rg} of 0.58 and the same Co content, as shown in figure 5(c). For the amorphous FCZBN alloys with 5 and 7.5 at. % Co, T_{rg} is greater than 0.58, and so the interatomic distance for a Fe–Fe couple should be much smaller than that for amorphous FCZBN alloys with T_{rg} of 0.58, resulting in a decreasing value of the exchange interaction induced by decrease in the interatomic distance that is much larger than the increase in the value of the exchange interaction induced by an increase in Co content. Therefore, the amorphous FCZBN alloys with 5 and 7.5 at. % Co have a much lower T_c compared with amorphous FCZB alloys irrespective of the Co content of the amorphous FCZBN alloys, as shown in figure 5(c). On the basis of the conclusions mentioned above, the experimental phenomenon that the rate of decrease of the T_c with T_{rg} is different for different Co-content ranges shown in figure 5(b) can also be explained. For Co content ranging from 0 to 3 and 7.5 to 12.5 at. %, T_{rg} increases with decreasing Co content, and both decrease of Co content and increase in T_{rg} result in a decrease of T_c. For Co content ranging from 3 to 7.5 and 12.5 to 15 at. % T_{rg} increases with increasing Co content; however, increase in T_{rg} results in a decrease of T_c, but increase of Co content increases the T_c. Therefore, the decreasing rate is larger for Co content ranging from 0 to 3 and 7.5 to 12.5 at. % compared with the Co content ranging from 3 to 7.5 and 12.5 to 15 at. %.

Similar results were also reported for other Fe-rich Fe-based amorphous alloys [16, 17]; for example the Ta content dependence of the T_c is opposite to that of T_g and GFA in amorphous $Fe_{56}Co_7Ni_7Zr_{10-x}Ta_xB_{20}$ and $Fe_{56}Co_7Ni_7Zr_{10-x}Nb_xB_{20}$ alloys [16]. The bulk $Fe_{60}Co_8Zr_{10}Mo_5W_2B_{15}$ amorphous alloy has the highest T_g and GFA in the Fe-based amorphous alloy to the best of our knowledge, but its T_c is the lowest in the reported Fe–Co-based amorphous alloys [15].

4. Conclusions

The T_c of the amorphous FCZBN alloys show a sinusoid-like changing behaviour with increasing Co content, revealing two maxima at 3 and 12.5 at. % and a minimum at 7.5 at. %. This behaviour is almost completely opposite to Co-content dependence of the T_g, T_x and T_{rg} over the same composition range. T_c decreases with increasing T_g and T_{rg}, but increases with increasing Co content. The Co-content dependence of the T_c is determined by competition of effects of Co content and T_{rg} on the T_c. The effect of Co content can be attributed to the exchange interactions of Fe–Co and Co–Co couples, which are stronger than those of Fe–Fe, resulting in an increase in T_c with increasing Co content. The effect of T_{rg} is due to decrease in the exchange interaction of Fe–Fe couple with decreasing interatomic distance, caused by an increase in the density of the amorphous FCZBN alloy with increasing GFA, which results in a decrease of T_c with increasing T_{rg}.

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