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MAGNETIC PROPERTIES OF METAL-METAL AMORPHOUS ALLOYS

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INTRODUCTION

During the last decade, magnetic properties of a large number of melt-quenched ferromagnetic amorphous alloys have extensively been studied from fundamental and practical points of view. Most of these studies, however, have been directed towards alloy systems of 3d transition metals with about 15-30 at.% of metalloïd elements of III, IV and V groups as glass-forming elements.

Recently, (Fe, Co and/or Ni)-(Zr or Hf) alloys containing less than 15 at.% Zr or Hf have been for the first time produced in the amorphous state by rapidly quenching from the melt. The formation range of amorphous phase, the mechanical properties as well as thermal stability have been reported earlier^(1,2). In particular, most of (Fe,Co,Ni)₉₀-Zr₁₀ alloys exhibit low-field magnetic properties superior to those of TM-metalloïd alloys⁽³⁾.

We report here the magnetic properties of these new amorphous alloys based on 3d transition metals, (V,Cr,Mn,Fe,Co,Ni), alone or combination, and Zr or Hf as the only glass former element.

EXPERIMENTAL

Amorphous alloy ribbons (1 mm wide, 20 µm thick) were prepared by the melt spinning technique. These ribbons were confirmed by X-ray diffraction to be amorphous. The temperature dependence of magnetization was measured in the range 4.2-800 K and in a magnetic field of 6~10 kOe by a pendulum magnetometer. The densities of the ribbons were measured by improved Archimedeam method using tetrabromoethane (Br₂CHCHBr₂).

RESULTS

The experimental results are summarized in Figs. 1 and 2, where the average magnetic moment, n_B per TM atoms and the Curie temperature, T_c, are plotted as a function of the average outer electron concentration, N, of TM atoms, respectively. Data for TM₈₀P₁₀B₁₀⁽⁴⁾ are also included for comparison.

The curves of TM₈₀P₁₀B₁₀ amorphous alloys show that n_B and T_c change in the similar manner to the Slater Pauling curve. The n_B for the (Fe-Co-Ni)₉₀Zr₁₀ amorphous alloys with N more than 8.5 also shows a similar decreasing to the general features. However, quite different behaviours in n_B for TM-metalloïd alloys less than N=8.5 can be noticed. The n_B for Fe₉₀Zr₁₀ (N=8) is significantly small, compared with Fe₈₀P₁₀B₁₀, and thus n_B for TM₉₀Zr₁₀ increases rapidly with increasing N and reaches a maximum value near N=8.4. The maximum value is 1.85 µ_B for Fe-Ni-Zr and 2.05 µ_B for Fe-Co-Zr, respectively. It is notice that n_B for N=8 is the ever reported minimum moment of 1.29 µ_B/Fe atom in

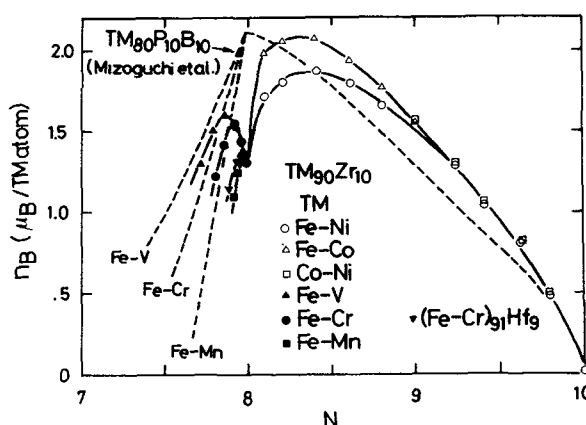


Fig. 1 The average saturation moment of amorphous TM-M alloys(TM; Ni, Co, Fe, Mn, Cr and/or V, M; Zr or Hf) plotted as a function of the average outer electron concentration of a TM atom

As seen in Fig. 1, the transition metals such as Mn, Cr and V which number of 3d-electrons is less than Fe greatly reduce the net moment of the TM-metalloïd amorphous alloys. It is, however, seen the interesting behaviour that the replacement of Fe by Mn, Cr or V in TM₉₀Zr₁₀ amorphous alloys first increases n_B contrary to TM-metalloïd alloys and then decreases greatly. The value of N where the n_B takes a maximum value decreases with decreasing atomic number of TM atoms.

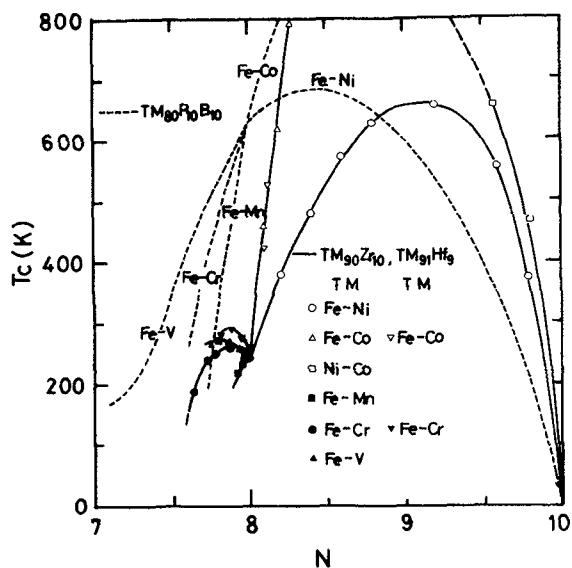


Fig. 2 The Curie temperature of amorphous TM-M alloys plotted as a function of the average outer electron concentration of TM atom

In addition to this unusual feature of n_B , T_c of Fe-rich alloys also behaves differently from the conventional TM-metalloid amorphous alloys. T_c takes 243K for $Fe_{90}Zr_{10}$, which is very small compared with T_c for Fe-metalloids. For $(Fe-Ni)_{90}Zr_{10}$ amorphous alloys, T_c changes in a parabolic way with respect to N , showing a maximum value at $N=9.4$. In the case of Fe-Co alloys, a large content of Co more than $N=8.3$ (30 at%) raises T_c beyond the crystallization temperature, T_x ,⁽¹⁾ and the compositional dependence of T_c has the similar tendency to that of Fe-Ni alloys. On the other hand, Mn, Cr and V simply reduce the T_c of amorphous Fe-based metalloid alloys. In the case of $TM_{90}Zr_{10}$ amorphous alloys, however, a small addition of these elements results in the increase of T_c , although T_c decreases in the high concentration range.

In Fig.3, the reduced magnetization, $M(T)/M(0)$, as a function of reduced temperature, T/T_c , is illustrated for Fe-Zr and Fe-Hf amorphous alloys together with those of $Fe_{88}B_{12}$, Fe-P-C⁽⁵⁾, crystalline α -Fe and Fe-Ni invar alloy. The curves for the present alloys are very closed to that of $Fe_{88}B_{12}$, being much less convex upward than that of α -Fe. These curves trends of approximately the same order as that of crystalline Fe-Ni invar alloy. It has also been found that the replacement of Fe by Mn, Cr or V makes the reduced magnetization curve less convex upward than those of the binary $Fe_{90}Zr_{10}$ and $Fe_{91}Hf_9$ amorphous alloys.

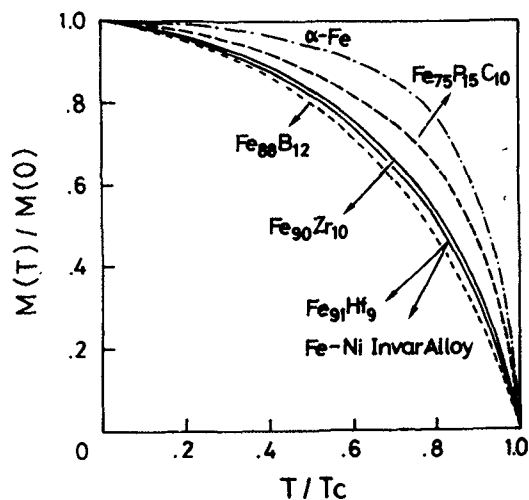


Fig. 3 Reduced magnetization curves of amorphous Fe-(Zr or Hf) alloys together with those of amorphous Fe-B and Fe-P-C and those of crystalline α -Fe and Fe-35.8 Ni alloys

The detailed compositional dependences of n_B and T_c in the systems of Fe-Zr and Fe-Hf are shown in Fig. 4. Both n_B and T_c decrease monotonically with decreasing Zr or Hf content. If our data is combined with those of Kazama et al.⁽⁶⁾ for Fe-Zr alloys prepared by a sputtering method, it appears that both n_B and T_c show a peak around critical value $x=13$ ⁽⁷⁾. Since this variation in n_B is similar to the magnetic moment vs. concentration curve for Fe-Ni invar alloys. It is of interest to apply the Weiss' hypothesis, namely two magnetic states on Fe, for explaining the presently found variation in n_B . In regard to this anomalous behaviour in n_B and T_c , the following results concerning density should be noticed.

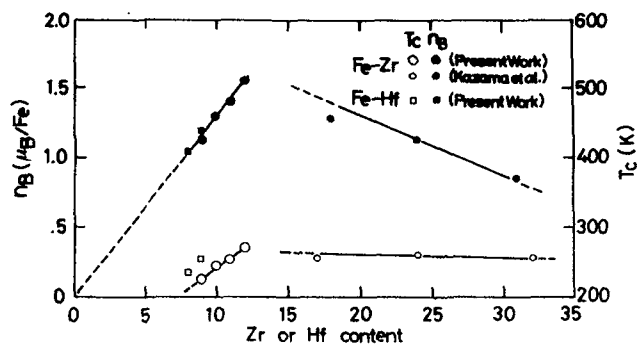


Fig. 4 Saturation moment and Curie temperature as a function of Zr or Hf content

Fig. 5 illustrates the density, D , for amorphous $(\text{Fe-Co})_{90}\text{Zr}_{10}$, $(\text{Fe-Co})_{80}\text{B}_{20}$ ⁽⁸⁾ and $(\text{Fe-Co})_{75}\text{Si}_{15}\text{B}_{10}$ ⁽⁹⁾ alloys as a function of cobalt concentration $\text{Co}/(\text{Co}+\text{Fe})$. Three significant features may be noted. First, the density increases in the order of (Si,B)-, B- and Zr-(Fe-Co) alloys, or with increasing the total amount of (Fe-Co) content. Second, the density changes almost linearly against Co content, but with two different slopes which can be identified at a critical concentration (c.c). The declines for three alloys are nearly the same in the Co rich region but different in the Fe rich region. Third, the slope of the density vs. Co content in the Fe rich region becomes small in the order of Si-B, B and Zr-alloys.

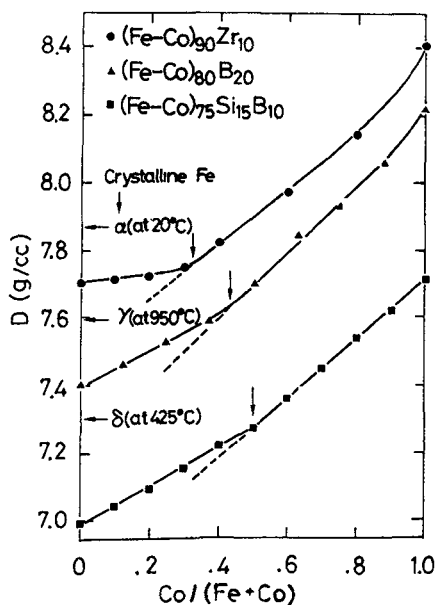


Fig. 5 Composition dependence of the density for amorphous Fe-Co based alloys

It seems that the density in the Fe rich region is abnormally large, compared with the value obtainable by extrapolating the linear portion in the Co rich region to $\text{Co}=0$. This large density is thought to be arising with the substitution of Co by Fe atoms accompanying with a considerable structural relaxation such that the amorphous structure makes much denser. This view is supported by the result of X-ray study on structure⁽¹⁰⁾, that is, the coordination number of Fe-Fe atoms in $\text{Fe}_{90}\text{Zr}_{10}$ alloy is 11.8 which is bigger than 10.8 for $\text{Co}_{90}\text{Zr}_{10}$ alloy.

The density for amorphous $(\text{Fe-TM})_{90}\text{Zr}_{10}$ alloys is plotted as a function of the average outer

electron concentration, N , of TM atoms in Fig. 6. The density of these alloys for N more than 8 have been already explained. The behavior in density around $N=8$ should be noticed. The replacement of Fe by small amounts of Mn, Cr or V for $(\text{Fe-TM})_{90}\text{Zr}_{10}$ alloys does not change the value of density significantly. This is particularly noticeable in the case of Cr and V atoms. However, when the replacement is large, the density of $(\text{Fe-TM})_{90}\text{Zr}_{10}$ alloys is greatly reduced in the sequence of Cr, V and Mn.

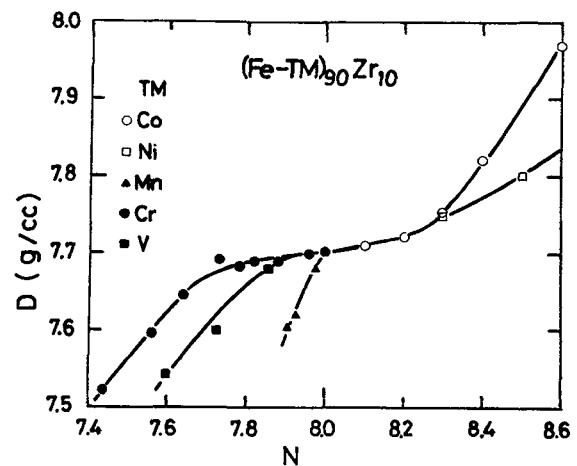


Fig. 6 The density of amorphous TM-M alloys plotted as a function of the average outer electron concentration of a TM atom

The small change in density near $N=8$ may be attributed to structural relaxation due to the TM atom situated interstitially among Fe atoms. On the other hand, the large reduction of density due to the high content of Mn, Cr and V may be occurred with the substitution of Fe by three elements.

Discussion

The sharp decreases in n_B and T_c for amorphous $\text{TM}_{90}\text{Zr}_{10}$ and $\text{TM}_{91}\text{Hf}_9$ alloys with increasing iron concentration, observed here, is also found in the case of the crystalline Fe-Ni invar alloy. In regard to this similarity, it should be noted that the amorphous quaternary alloys of $(\text{Fe}_{1-x-y}\text{Co}_x\text{Ni}_y)_{90}\text{Zr}_{10}$ for $x+y$ less than 0.28 have, in fact, a clear invar characteristics in a wide temperature range.⁽¹¹⁾ Although no definitive theory to explain the invar effect has yet been established, it seems of little doubt that the invar effect is associated with a negative exchange interaction among some of the magnetic moments⁽¹²⁾. It is known that the spin moments on Fe atoms have a tendency to the negative coupling when the Fe atoms are densely packed.

The concentration dependence of the density for $\text{TM}_{90}\text{Zr}_{10}$ alloys implies that Fe atoms are densely randomly packed in these amorphous alloys. That is, Fe-rich portion in the amorphous alloys seems to have a considerable amount of local atomic cluster based on nearly 12-fold coordinated dense random packing structure. This cluster would seem to have the Fe-Fe pairs closely packed to each other, therefore, such Fe-Fe pairs may have presumably a negative exchange interaction. Assuming the negative exchange interaction among Fe atoms, the decrease in n_B with decreasing Zr or Hf as found in Fig. 4 can be explained by the increase in the number of this type of Fe-Fe pairs.

On the case of $(\text{Fe}_{1-x}\text{Co}_x \text{ or } \text{Fe}_{1-x}\text{Ni}_x)_{90}\text{Zr}_{10}$ alloys, the number of Fe-Fe pairs with negative exchange interaction decrease with increasing x , while the number of Fe-Co or Fe-Ni pairs with positive exchange interaction increase. Therefore, the n_B increases sharply with increasing x .

On the other hand, in the case of amorphous $(\text{Fe-TM})_{90}\text{Zr}_{10}$ alloys (TM=Mn, Cr or V), as shown in Figs. 1 and 2, replacing Fe by a small amount of TM atoms increases both T_c and n_B . This decrement may be tentatively explained as follows. As already mentioned in Fig. 6, no significant change in density can be observed by replacing of Fe by Cr and V in the $(\text{Fe-Cr or V})_{90}\text{Zr}_{10}$ alloys. The mechanism for the small change in density, even though the atomic weight of Cr or V is smaller than that of Fe, may be explained such that the atoms of Cr or V are placed interstitially among Fe atoms so as to increase the total number of Fe and Cr (or V) atoms per unit volume. TM atoms situated interstitially among Fe atoms would slightly increase the atomic distance of surrounding Fe atoms. This slight expansion may change the magnetic interaction of Fe-Fe pair from negative to positive. Therefore, the values of n_B and T_c increase with addition of a small amount of Co or V. The decrease in n_B in the high content of Mn, Co and V is attributed to the antiferromagnetic coupling between Fe and TM atoms.

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