

PREPARATION AND STRUCTURAL STUDY OF MECHANICAL ALLOYED OF $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$

X. F. ZHANG, X. Q. HUANG, R. W. PENG, G. Q. WANG and S. Y. ZHANG

*Department of Physics and National Laboratory of Solid State Microstructures,
Department of Electronic Engineering, Nanjing University, Nanjing 210093, China,
The Science and Technology University of the PLA, Nanjing 210016, China*

Received 4 September 2004

The amorphous alloys of $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$ are successfully obtained by using the mechanical alloying technique. The sample is analyzed by X-ray diffraction, transmission electron microscopy and differential scanning calorimetry. The DSC result of the powder sample milled for 120 h shows a complete amorphous phase and a wide supercooled liquid region ($T_x - T_g \simeq 80$ K).

Keywords: Amorphous alloys; supercooled liquid region.

1. Introduction

After the discovery of the excellent soft magnetic properties of Fe-Si-B-Nb-Cu,¹ a class of noncrystalline with soft magnetic properties has been produced by controlling crystallization of amorphous precursors which are in ribbon form by melt spinning and planar flow casting methods. The limitation in shape and geometry of the materials prepared by rapid solidification make this family of materials difficult to be applied.² Recently, efforts have been devoted to the development of bulk metallic glasses which have been obtained in Zr, Ti, Ni, Cu and Mg based systems, and more recently in Ca-Mg-Cu and Ca-Mg-Ag-Cu systems.

On the other hand, in order to predict alloy compositions with high glass forming ability (GFA), a number of attempts have been made to understand the mechanism of amorphization. Shen *et al.* were successful in obtaining a amorphous rods of $\text{Fe}_{67}\text{Cr}_4\text{Mo}_4\text{Ga}_4\text{P}_{11}\text{B}_5\text{C}_5$ metallic glasses with a diameter of 4 mm.^{3,4} So it is highly desirable to develop a material (and/or method) having similar magnetic properties to those noncrystalline and morphology adequate to the fabrication of the devices for which the use of the available material is convenient.

As one of the useful methods of material processing, mechanical alloying (MA) is an effective and inexpensive way to produce large-scale quantities of amorphous alloy powders.^{5,6} It is well known that the powder obtained by MA can be pressed to any necessary bulk shape. So it is important to investigate the amorphization of such multicomponent system that may show good soft magnetic properties by subsequent

heat treatment. In this paper, we select the precursors of $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$ on the following three empirical rules:¹ (1) multicomponent alloy system consisting of more than three constituent elements; (2) significant difference in atomic size ratios above 12% among the three main constituent elements; (3) negative heats of mixing among their elements.

2. Experiment

The Fe–V (52 : 48, 200 mesh) binary powder is mixed with the elemental powders of Fe, Co, Cu and B (>99% purity, 100 – 200 mesh) to give the desired component and sealed in a cylindrical stainless steel container under argon atmosphere. The mechanical alloying is performed in a planetary ball mill with a ball-to-powder weight ratio of 30 : 1, and at a rotation speed of 260 rev/min. The containers are opened only at the end of each run. A little methanol is added to prevent excessive welding in the chamber that allowing a high degree of mixing. The structure of the obtained powders are examined by *Dmax* – γA X-ray diffractometer of CuK radiation. Transmission electron microscopy (TEM) and electron diffraction patterns of the samples are obtained by *JEM* – 200cx. The thermal property of the sample is studied with a differential scanning calorimetry (DSC) at a heating rate of 15 K/min.

3. Result and Discussion

Figure 1 shows the XRD patterns of the $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$ powders of different milling times. It can be seen that with the increasing of milling time, the peaks become broaden and the intensity of the peaks decrease. These results indicate

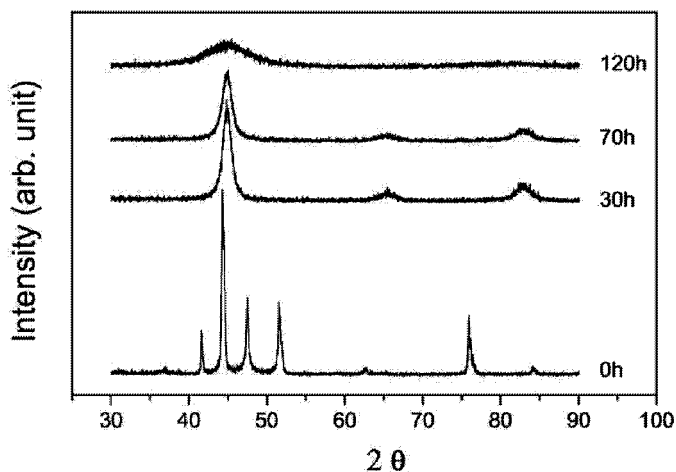


Fig. 1. X-ray diffraction patterns of $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$ powders after various milling time.

that the increasing of milling time would decrease the crystalline size and increase the internal strain. When the milling time up to 120 h, no evidence of any crystalline peaks can be seen in the figure, which indicate that the crystallite alloy has been transformed into amorphous powder.

The variation of the full width at the half maximum (FWHM) of the reflection profiles with different angle can be analytically represented:

$$\beta(2\theta) = \sqrt{U \tan^2 \theta + V \tan \theta + W}. \quad (1)$$

By applying the Scherrer equation

$$D = \frac{0.9\lambda}{\beta(2\theta) \cos \theta}, \quad (2)$$

the average crystalline size (D) can be easily calculated from the above equation.

Figure 2 shows the milling time dependence of the average crystalline size of the studied powders. It is can be seen that the sizes of crystalline become almost constant after 30 h.

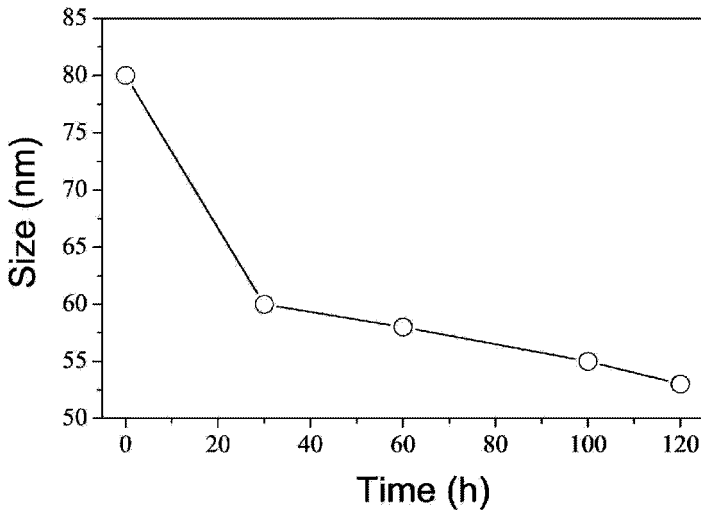


Fig. 2. Critical Average crystalline size of $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$ for various milling times.

Figure 3 is a TEM bright-field image and an electron diffraction pattern of the $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$ powder. From this figure, we can see the halo ring in the pattern, which confirms the sample is amorphous. Here, we try to give a reasonable explanation of our results. It is noted that Fe has a similar atom size to Co, the partial Co replaced by Fe will cause disordered structure in the sample and the corresponding grain sizes are much smaller than the exchange coherence length. In this newly obtained powder, the magnetic anisotropy is randomly averaged out by

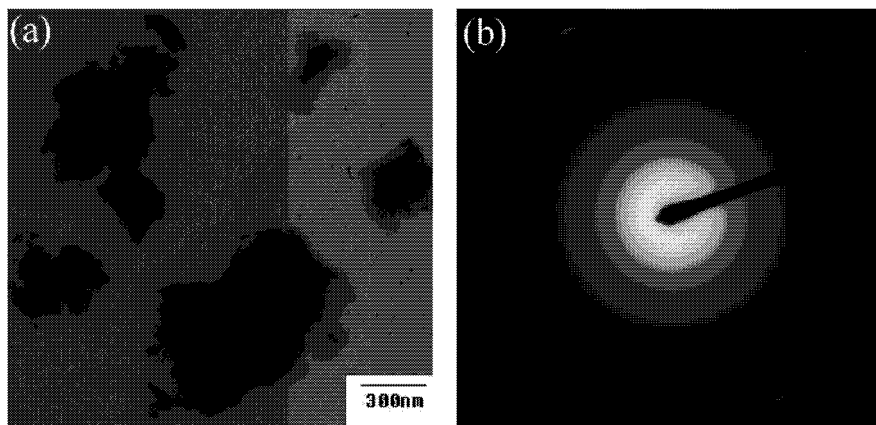


Fig. 3. TEM micrograph images and corresponding electron diffraction of $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$ which milled for 100 hours.

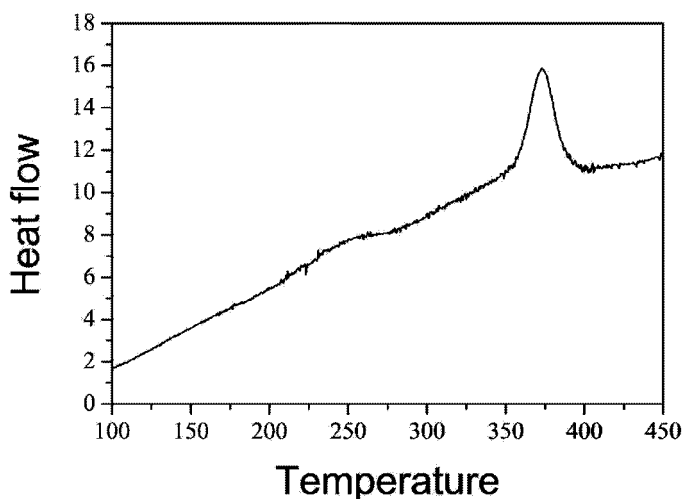


Fig. 4. The DSC cure of the amorphous $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$.

exchange interaction, therefore, there is only a small net-effect on the magnetization process.

Figure 4 presents the DSC curve for the amorphous powder at a heat rate of 15 K/min. Noted that two peaks appear in the figure. The first peak is inconspicuous show the glass transition temperature $T_g \simeq 270$ K, the second peak shows the onset crystallization temperature $T_x \simeq 350$ K. A large value of $T_x - T_g \simeq 80$ K is indicative of a high thermal stability of the supercooled liquid. In practice, such a large $T_x - T_g$ range is very important. It is well known that when heating a glass above T_g , the viscosity of the glass decreases extremely fast and the crystallization

does not start until the temperature reaches T_x . In addition, heating a glass within the $T_x - T_g$ range may enable us to shape the glass under a small applied pressure.

4. Conclusion

First, by using the ball-milling technique (milling time 120 h, rotation speed 260 rev/min), we have successfully obtained the amorphous phase material of $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$.

Second, if the milling time is less than 100 h and the rotation speed is lower than 260 rev/min, the results show that partial amorphization takes place. This process is more effective when the precursors are the elemental powders, the newly formed disorder phase and the residual crystalline phases reached equilibrium. Even if the milling time increase, complete amorphization cannot be obtained.

Third, the dependence of glass forming ability (GFA) on metalloid content suggested that the GFA of $\text{Co}_{50}\text{Fe}_{20}\text{Cu}_2\text{V}_8\text{B}_{20}$ could be optimized by adjusting the compositions of the two metalloids: vanadium and boron.

Acknowledgments

This work was supported by the grants from National Nature Science Foundation of China (90201039, 10274029, 10021001, 10374042), the State Key Program for Basic Research from the Ministry of Science and Technology of China, the provincial NSF of Jiangsu and partly by the Ministry of Education of China and Fok Ying Tung Education Foundation.

References

1. Y. Yoshizawa, S. Oguma, and K. Yamaguchi, *J. Appl. Phys.* **64**, 6044 (1988).
2. A. Inoue, T. Zhang, and A. Takeuchi, *Appl. Phys. Lett.* **71**, 464 (1997).
3. T. D. Shen and R. B. Schwarz, *Appl. Phys. Lett.* **75**, 49 (1999).
4. T. D. Shen, U. Harms, and R. B. Schwarz, *Mat. Sci. Forum.* **383**, 441 (2002).
5. J. J. Sonol, *Mater. Sci. Forum.* **269**, 503 (1998).
6. Y. L. Ji, G. H. Wang, F. Li, G. Q. Wang, and S. Y. Zhang, *J. M. Sci. Lett.* **19**, 1641 (2000).