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Study on soft magnetic properties of $Fe_{36}Co_{36}B_{19.2}Si_{4.8}Nb_4$ metallic glass

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Abstract. $Fe_{36}Co_{36}B_{19.2}Si_{4.8}Nb_4$ BMG rods with diameter of 3mm and amorphous ribbons with thickness of 100μm were prepared with methods of copper mold suction casting and melt-spinning, respectively. These metallic glasses were annealed at different temperatures in vacuum, and the changes of structures and magnetic properties were analyzed. The hysteresis loops were tested by B-H loop tracer and the magnetic domain structures were characterized with MFM. Results show that annealing between T_g and T_x leads to the best soft magnetic property, and this is referred to be induced by the migration of magnetic domain walls.

1. Introduction

Since bulk glassy alloys were synthesized in variety of alloy systems [1-4], great efforts have been devoted to promoting the final application of bulk metallic glasses (BMGs). These alloys show extraordinary mechanical, physics and chemical properties [5-9], among which Fe-based BMGs are drawing increasing attention due to their superior soft magnetic properties [10, 11]. They can be processed directly into sophisticated micro-cores and components such as sensors, showing broad prospects for applications in the fast growing high-tech field. Besides, the low cost of raw materials makes them more easily to be widely applied comparing to other alloy systems. Therefore, further study on the micro-mechanism of their outstanding soft magnetic properties is necessary. Magnetic domains of BMGs which have seldom been studied before were characterized by MFM in this article, and it may be of significance for understanding the origin of the soft magnetic property.

2. Experimental

Ingots of $Fe_{36}Co_{36}B_{19.2}Si_{4.8}Nb_4$ alloy were prepared by arc melting the mixtures of constituent elements in a titanium gettered argon atmosphere, and the purities for all elements are higher than 99.9wt%. The alloy compositions represent nominal atomic percent. Alloy rods with diameter of 3mm were prepared by means of copper mold suction casting and ribbons with thickness of 100μm were prepared by melt-spinning. Structural identification of the alloy rods was carried out by means of X-ray diffractometry (XRD) on the cross-section using the Cu K_α radiation (λ=0.15406nm). Differential scanning calorimetry (DSC) were employed to study the thermodynamic behaviors of the metallic glasses (MGs) at a heating rate of 0.67K/s. Magnetic properties were measured with a B-H loop tracer under a field of 800A/m. The prepared metallic ribbons were annealed under different temperatures for 1h after enclosed in quartz tubes with vacuum, and temperature in the annealing furnace was calibrated with commercial temperature measuring pens. Then the changes of structures and magnetic properties with annealing were analyzed. At last, the magnetic domain structures for rod samples were characterized with magnetic force microscopy (MFM).

3. Results and discussion

3.1. Structural and thermodynamic analysis

The alloy rods with diameter of 3mm are shown in Fig.1. Their XRD spectrum is shown in Fig.2. The broad and continuous peak with the absence of sharp crystallized peak implies fully amorphous structure. Then DSC analysis was carried out to study the thermodynamic behaviors of the BMGs, which is shown in Fig.3. The glass transition temperature (T_g) and crystallization temperature (T_x) of the BMGs are 545℃ and 590℃ from the DSC spectrum, respectively.

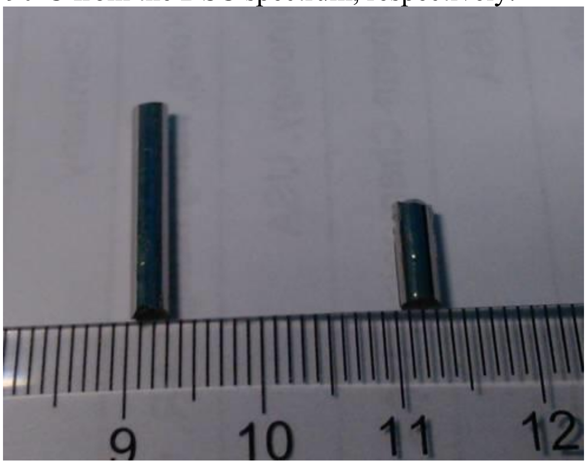

Figure 1. $Fe_{36}Co_{36}B_{19.2}Si_{4.8}Nb_4$ alloy rods with diameter of 3mm

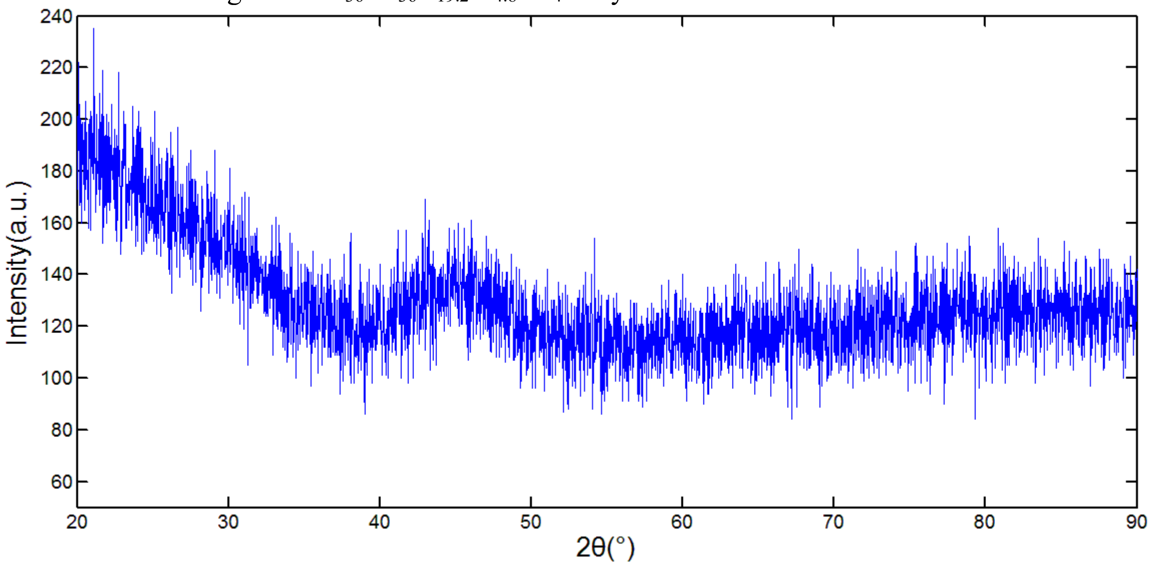

Figure 2. XRD spectrum of $Fe_{36}Co_{36}B_{19.2}Si_{4.8}Nb_4$ alloy rod

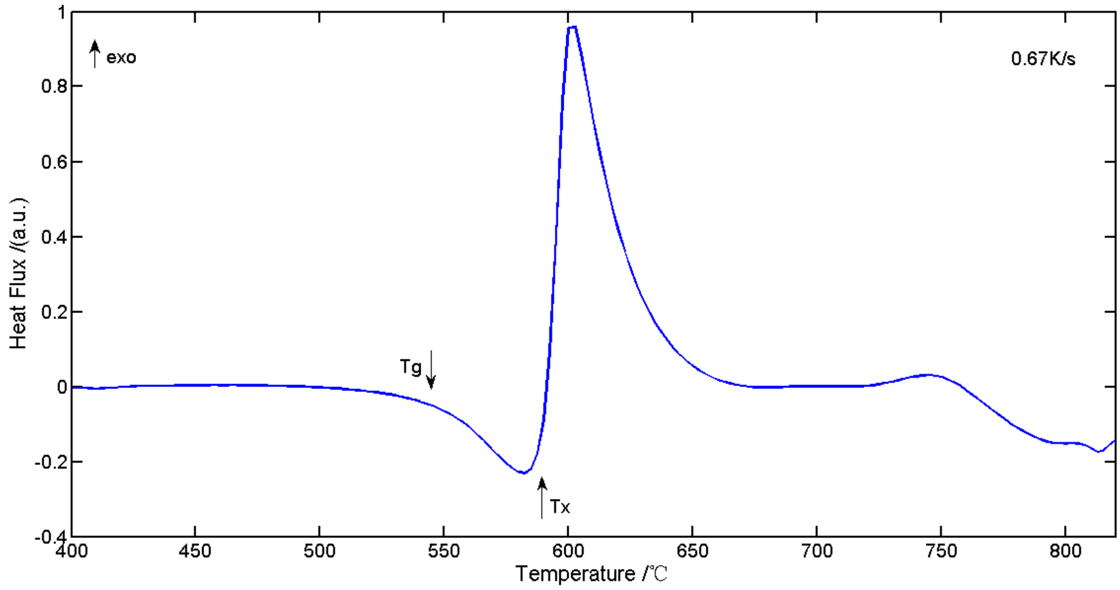

Figure 3. DSC spectrum of $Fe_{36}Co_{36}B_{19.2}Si_{4.8}Nb_4$ BMG rod

3.2. Soft magnetic properties analysis

Since the soft magnetic properties of amorphous alloys can be improved by annealing due to structural relaxation, the amorphous ribbons were annealed at 5 temperatures (200°C, 350°C, 508°C, 580°C, 650°C) in vacuum. These temperatures are below T_g (200°C, 350°C, 508°C), between T_g and T_x (580°C), and above T_x (650°C), respectively. And the annealing time is 1h for all of the ribbons. Then XRD tests were carried out with all these samples, as shown in Fig.4. It is clear from the diagram that samples annealed below T_g keep the amorphous structure, while a certain degree of nano-crystallization occurs when the annealing temperature is higher than T_g.

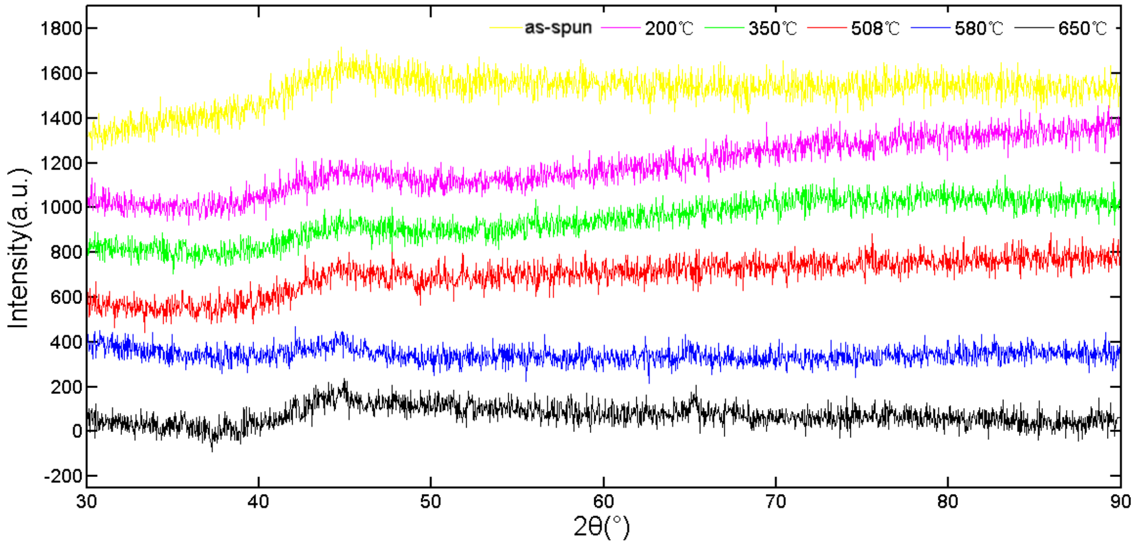

Figure 4. XRD spectrums comparison of $Fe_{36}Co_{36}B_{19.2}Si_{4.8}Nb_4$ amorphous ribbons annealed at different temperatures

B-H magnetic hysteresis loops of the annealed ribbons were measured with B-H loop tracer, as shown in Fig.5. The inset at the right bottom of Fig.5 is an enlarged view near zero, which clearly demonstrates the coercive forces of these samples. Comparing the hysteresis loop of the as-spun sample with those of 200°C, 350°C and 508°C annealed samples, which all keep amorphous structure, it's found that the 508℃ annealed sample possesses the minimum of coercive force and the maximum of magnetization, which imply the best soft magnetic properties. This result shows that annealing slightly below T_g leads to the best soft magnetic properties without changing the amorphous structure, which is consistent with literatures reporting that annealing at T_g-50℃ is conducive to improve soft magnetic performance of BMGs. However, for the samples annealed at 200℃ and 350℃, the changing trends of magnetization and coercivity with annealing are not the same. Their coercive forces are both larger than that of the as-spun sample, while the 200℃ sample possesses lower magnetization than that of the as-spun sample, and the 350℃ sample higher. Considering that the coercive force is the most fundamental property of soft magnetic materials, it can be judged that the soft magnetic properties were slightly decreased after annealing at 200℃ and 350℃. The low-temperature annealing effect on soft magnetic properties of MGs needs to be further studied.

Comparing all samples it can be found that the 580℃ sample (represented with the yellow line) possesses the lowest coercive force and the maximum of magnetization. The result shows that annealing between T_g and T_x improves the soft magnetic properties at the maximum extent. Combined with the XRD spectrums, nano-crystallization has occurred in this sample and a nanocrystallized-amorphous composite structure is formed. After annealing at 650℃, the coercivity starts to increase, and the magnetization decreases, implying decreasing of soft magnetic performance, which is due to the complete nano-crystallization in the sample when the annealing temperature is higher than T_x.

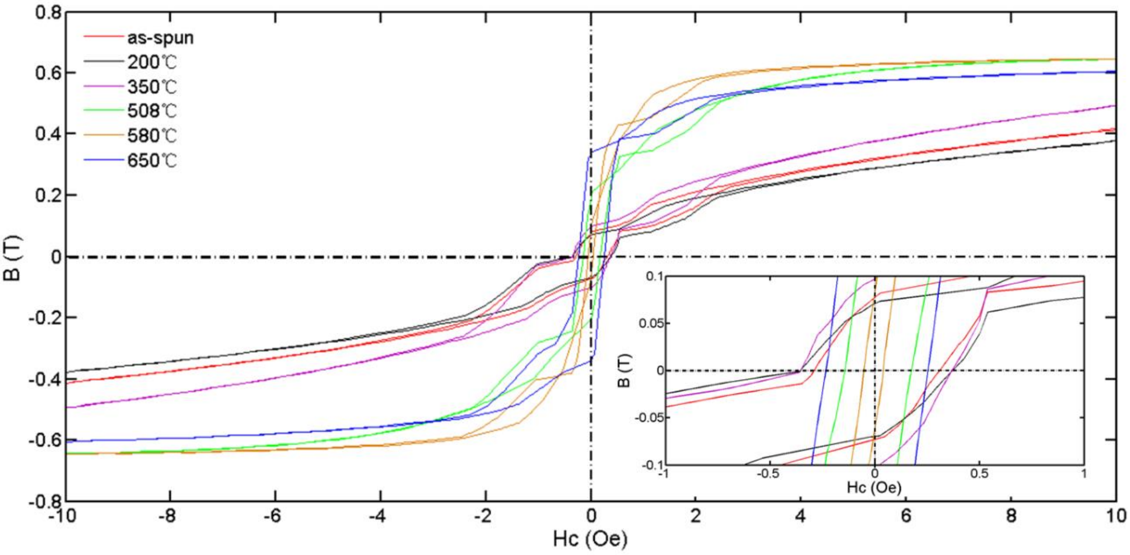

Figure 5. B-H hysteresis loops comparison of $Fe_{36}Co_{36}B_{19.2}Si_{4.8}Nb_4$ amorphous ribbons annealed at different temperatures. The inset shows enlarged hysteresis loops.

Results above indicate that a certain degree of crystallization to form nanocrystallized-amorphous composites will be the best in helping improve the soft magnetic properties.

3.3. MFM analysis

Since the 580℃ annealed sample possesses the best soft magnetic properties, we made further study on it with MFM, in order to find micro-mechanism for its excellent soft magnetic properties. Figure 6 shows the topography and magnetic domain images of as-cast and 580℃ annealed rod samples. The scanning range of the instrument is only within 20μm, so it's difficult to scan one pinpoint location before and after annealing. But we scanned the surface on multiple locations for each sample, and found that the domain patterns remain similar on the whole for both samples. It can be seen from the Figure 6 that the surface topography is fairly flat for both samples, but the magnetic domain structures differ greatly. Before annealing, the magnetic domains of the as-cast sample are quite localized, and the magnetic domain walls warp remarkably. However, most of the magnetic domains grow wider and the magnetic domain walls become straight clearly after annealing. This shows the distinct migration of magnetic domain walls, which leads to the growing of magnetic domains, and also makes straight of the warped magnetic domain walls during the moving process. Annealing provides a driving force for the movement of magnetic domain walls, which may explain this migration. The wider magnetic domains increase the magnetization of a single magnetic domain, which increases the magnetization of the whole sample in the end. And the wider magnetic domains also reduce the total number of domain walls on the other hand, which leads to more easily respond to an external magnetic field, that is, reduces the coercive force of the sample. Thus, this sample shows better soft magnetic performance at the macro level.

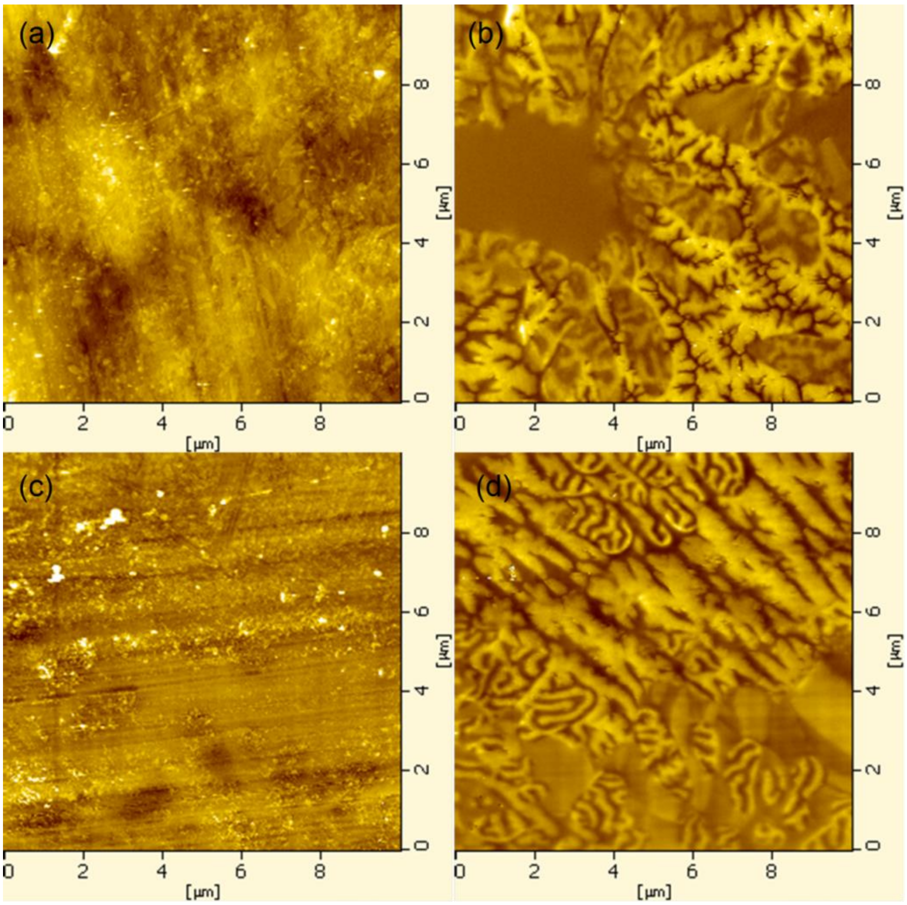

Figure 6. Topography (a, c) and magnetic domain (b, d) images of as-cast (a, b) and 580℃ annealed (c, d) BMG rods

Therefore, we can use the same principle of magnetic domain wall migration to explain the worse soft magnetic properties of the 650℃ annealed ribbons. Since 650℃ is higher than T_x, nano-crystallization in this sample is complete after annealing for 1h, which increases the total number of internal grain boundaries. The increase of grain boundaries impedes the movement of magnetic domain walls, thus impedes the magnetic domains growing, and also increases the number of magnetic domain walls, resulting in worse soft magnetic properties.

4. Conclusions

We prepared BMG rods with diameter of 3mm and amorphous ribbons with thickness of 100μm. The amorphous ribbons were annealed at different temperatures in vacuum, and structures and magnetic properties were analyzed after annealing. Results show that annealing between T_g and T_x leads to the formation of nanocrystallized-amorphous composites, which possess the best soft magnetic properties. MFM analysis was carried out with as-cast and 580℃ (T_g < 580℃ < T_x) annealed rod samples. Annealing results in the migration of magnetic domain walls, which makes the magnetic domains become wider and the warped magnetic domain walls become straight. The wider magnetic domains increase the magnetization of the whole sample and also reduce the total number of domain walls, which further reduces the coercive force. The sample show better soft magnetic properties in the end.

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