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THE EFFECTS OF ANNEALING ON THE DC MAGNETIC PROPERTIES OF AN IRON-BASED AMORPHOUS ALLOY

Y. S. Choi*, D. H. Kim*, S. H. Lim, T. H. Noh and I. K. Kang
*R & D Division, Hankook Core Co. Ltd., Chonan, Chungnam, 333-810, Korea
Division of Metals, KIST, P. O. Box 131, Cheongryang, Seoul, 136-130, Korea

Abstract-The iron-based Metglas 2605S3A amorphous alloy ribbons are annealed at 435°C for various periods from 5 to 210 min, and the effect of annealing is investigated on the dc magnetic properties of the ribbon. Typical square-type hysteresis loops are observed for the ribbons annealed for 5 min, indicative of the nearly complete removal of residual stresses which are produced during melt-quenching. As the annealing time increases, the coercivity increases and the shape of hysteresis loops transforms to round type and finally to sheared one at the longest annealing time of 210 min. These results may be explained by the formation of clusters with chemical short order and very fine crystallites (at the annealing time of 210 min), and the diffusion-induced stresses during the formation of the clusters. For the samples annealed for 5 min, very good dc properties of the squareness ratio, coercivity and maximum permeability are observed, but, rather unexpectedly, the initial permeability is found to be very low. These results are considered to be due to a simple domain structure consisting of very small number of 180° domains.

I. INTRODUCTION

Amorphous alloy ribbons, which are usually produced by rapidly quenching liquid melts onto a rotating wheel, were first reported by Duwez and his coworkers in 1960 [1] and were reported to exhibit some noble properties unheard of conventional crystalline alloys. Among the properties of amorphous alloy ribbons of particular interest are the excellent soft magnetic properties which are related to the very small magnetocrystalline anisotropy resulting from the absence of atomic long range order and the ease of domain wall motion due to structural uniformity. Amorphous alloy ribbons are usually devided into Fe- and Co-based alloys. Fe-based amorphous alloys, which usually exhibit higher magnetic flux density but rather poorer soft magnetic properties than Co-based amorphous alloys, have mainly been used as line frequency transformer cores whilst Co-based amorphous alloys used as high frequency core materials. In the late eighties, however, many research efforts were made to use Fe-based amorphous alloys for high frequency (about several kHz) applications such as high frequency transformers and choke cores by developing new alloys and controlling the microstructure through a suitable heat treatment. One of the Fe-based alloys developed for high frequency applications is $Fe_{76.5}Cr_2B_{16}Si_5C_{0.5}$ (Metglas 2605S3A) manufactured by AlliedSignal Corp. USA. It seems, however, that the changes in the microstructure and magnetic properties of the alloy on annealing are not fully known. The aim of the present work is to investigate the effects of annealing on the magnetic properties of the amorphous ribbons of Metglas 2605S3A. The results on dc magnetic properties only are presented in this article.

II. EXPERIMENTAL

The amorphous ribbons of wide width were supplied by AlliedSignal Corp and they were slit to 4.5 mm width. The thickness of the ribbons was below 20 μm. The ribbons were wound onto a torodial core with inner diameter of 19 mm. The number of wound layer was fixed to 50. The cores were annealed at a fixed temperature of 435°C but the duration of annealing was varied widely from 5 to 210 min. Immediately after annealing, the samples were water-quenched. The dc magnetic properties were measured with a hysteresis loop tracer and the measured properties were the remanence ratio (B_r/B_s where B_r and B_s are the remanent flux density and saturation flux density, respectively), coercivity (H_c), initial permeability (μ_i) and maximum permeability (μ_m).

III. RESULTS AND DISCUSSION

In Figs.1(a)-(f) are shown the hysteresis loops for the samples annealed for 0 (as-cast), 5, 20, 60, 120, and 210 min, respectively. In order to see the variation of the loop with annealing time accurately, the maximum applied magnetic field was fixed to 100 mOe. From the figures it is seen that, in the case of as-cast sample, the value of coercivity is very large and the flux density increases very slowly with the applied magnetic field, in particular, no appreciable increase in the flux density being observed up to 60 mOe of applied magnetic field in the virgin curve. The poor

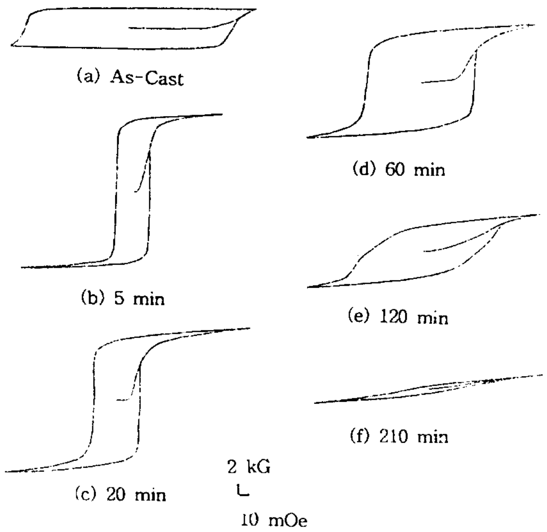

Fig.1 Dc hysteresis loops of Metglas 2605S3A as a function of annealing time. The annealing temperature is 435°C.

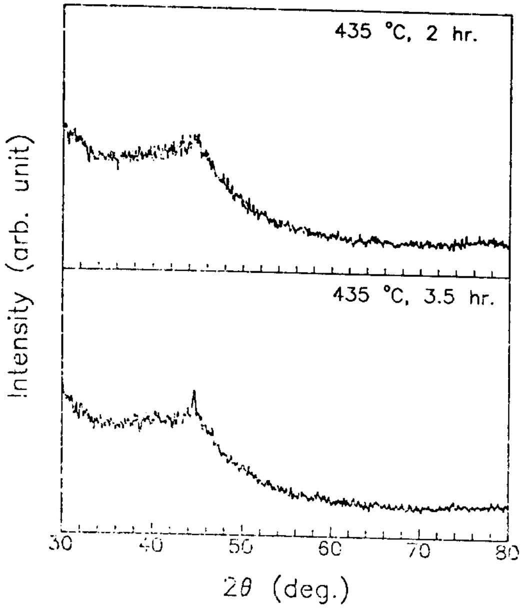

Fig.2 X-ray diffraction patterns of Metglas 2605S3A annealed at 435°C for 120 and 210 min.

soft magnetic properties of the as-cast sample is due to the remaining stress induced during rapid quenching and the coupling of the stress to magnetostriction resulting in magnetoelastic interaction which hinders the domain wall motion. It is known that the magnitude of magnetoelastic interaction is proportional to the product of the remaining stress and the magnetostriction [2]. A significant change occurs in the shape of hysteresis loop for the samples annealed for a short period of 5 min; coercivity is very small and the loop changes to the typical square type. This result is condidered to be due to the relief of the remaining stress rather substantially by the short period of annealing and hence the ease of domain wall motion. As the duration of annealing increases, the shape of hysteresis loop gradually transforms from square type into round one and finally the typical sheared loop is observed for the samples annealed for 210 min. The results may be explained by the two contributions which occur with the annealing; the stress relief which facilitates the domain wall motion and the formation of clusters with chemical short range order and/or precipitates which retards the domain wall motion. It is expected that the latter contribution dominates as the annealing time increases. Since no precipitates are detected by x-ray diffraction and transmission electron microscopy (TEM) except for the samples annealed for 210 min, which seems to be reasonable because the annealing temperature (435°C) is far lower than the crystallization of 535°C, the change in the dc magnetic properties for the samples annealed for more than 20 min may be explained by the formation of clusters with chemical short range order and hence the difficulty of domain wall motion. It was reported by Kronmuller [3,4] that the difficulty of domain wall motion is proportional to the square root of the size of clusters. Since the size of clusters is expected to increase with the annealing time, the results shown in Figs.1 for the change in the hysteresis loops as with the annealing time may thus be understood. However, the contribution by the clusters is known to be rather small, for example, the contribution to the coercivity being less than 10 mOe. More factors, therefore, should be taken into account to explain the large increase in the coercivity. One possibility is the re-introduction of stress induced by the atomic migration to form the clusters. The atomic migration induced stress often occurs for Fe-based amorphous alloy ribbons and its contribution to coercivity is reported to be large [5]. The coercivity increase of more than 50 mOe was reported by Noh et al for Fe$_{76}$Mo$_7$P$_{17}$ alloys by the re-introduced stress accompanying the atomic migration for the cluster formation during a low temperature structural relaxation [5]. X-ray diffraction patterns for the samples annealed for 210 min, as shown in Fig.2, indicate the existence of a very small amount of precipitates, which may explain the significant difference in the shape of hysteresis loop of the samples annealed for 210 min, compared to the samples annealed for the shorter periods of time. It is reported that sheared hysteresis loops similar to that shown in Fig.1(f) are often observed for the samples with precipitates, because the precipitates cause the domain structure to be refined as well as they hinder the domain wall motion [6,7].

In order to examine the variation of the magnetic properties with the annealing time more clearly, the change in the remanence ratio, coercivity, initial permeability and maximum permeability is investigated as a function of annealing time and the results are shown in Figs.3-6. The value of remanence ratio is 0.47 for the as-cast samples and it increases significantly to 0.75 when annealed for 5 min. When the annealing time is increased further, the remanence ratio decreases monotonically with the annealing time and, for the samples annealed for 210 min, it becomes as low as 0.27 which is much samller than that of as-cast samples. Since, according to Chikazumi [8], the remanence ratio is a measure of the distribution of the magnetization direction of domains, some information on domain orientation may be deduced from the results for remanence ratio. Ribbon-type samples mainly consist of 180° type domains and, if the domains are distributed randomly within the samples, the remanence ratio is calculated to be 0.5 [8]. If the domains are aligned in the length and the transverse directions only, the remanence ratios are 1 and 0, respectively [8]. The remanence of as-cast samples (0.47) is close to 0.5, indicating random distribution of domains. This may result from the fact that the domain orientation is mainly determined by magnetoelastic interactions due to the remaining stress. More domains are considered to be in the length direction for the samples annealed for 5 min. This is because the domain orientation is not affected greatly by magnetoelastic interactions due to the relief of the remaining stress but by the shape anisotropy and the induced anisotropies during annealing. Although no magnetic field or stress is applied to the samples during annealing, the domain orientation will be in the length direction below the Curie temperature due to the shape anisotropy and hence the induced anisotropies will be formed in the same direction. The preferred orientation in the length direction gradually decreases with the annealing time and, at the annealing of 120 min, the domain orientation appears to be random. This may be related to the formation of clusters and/or atomic migration induced stress, which are likely to supress the directional ordering responsible for the induced anisotropies or refine the domains. The samples annealed for 210 min which have a small amount of precipitates exhibit low remanence ratio,

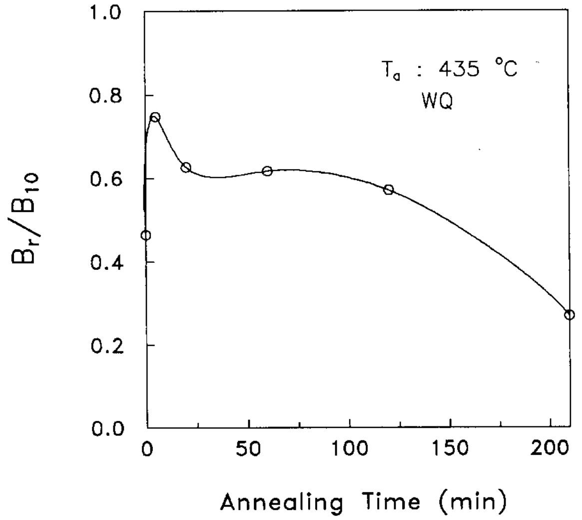

Fig.3 The value of squareness ratio of Metglas 2605S3A as a function of annealing time. The annealing temperature is 435°C.

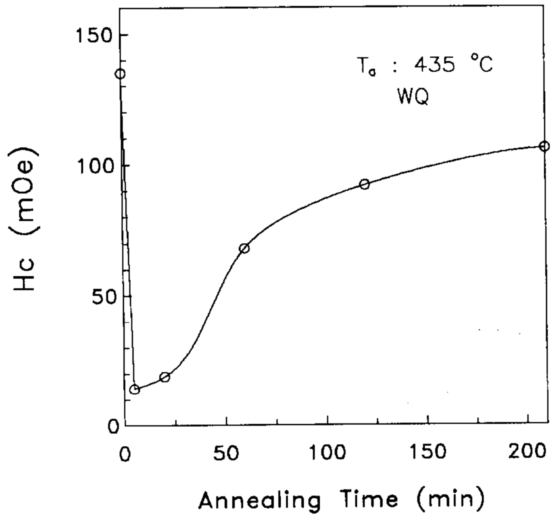

Fig.4 The value of coercivity of Metglas 2605S3A as a function of annealing time. The annealing temperature is 435°C.

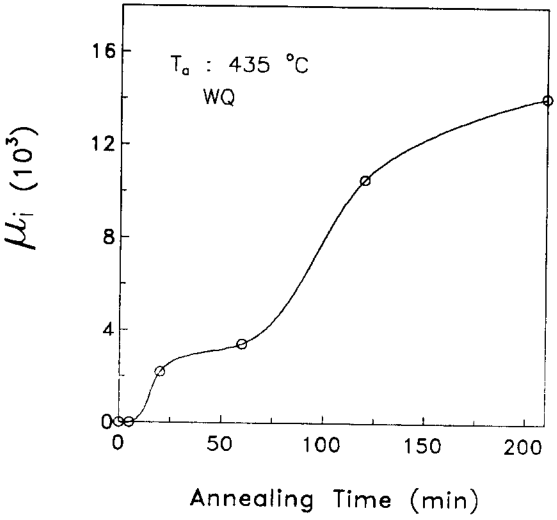

Fig.5 The value of dc initial permeability of Metglas 2605S3A as a function of annealing time. The annealing temperature is 435℃.

indicating more domains are aligned in the transverse direction.

The coercivity of as-cast samples is very large (135 mOe) but is significantly reduced to 14 mOe by the annealing for 5 min. As the annealing time is increased further, the coercivity monotonically increases with the annealing time. These results can be explained by the relief of the remaining stress nearly completely at the annealing time of 5 min and, at the longer annealing time, by the formation of clusters with chemical short order, precipitates and the atomic migration induced stress.

The initial permeability is very low for the as-cast samples and the samples annealed for 5 min and, at the further increase of annealing time, it increases monotonically with the annealinhg time. These results for the initial permeability are not in accord with the known fact that coercivity and permeability have the reciprocal relationship. The low initial permeability of as-cast samples is well understood by the existence of remaining stress and is well in accord with the other dc magnetic properties. However, it seems not straightforward to understand the low initial permeability of the samples annealed for 5 min considering low coercivity (Fig.4) and high maximum permeability (Fig.6) of these samples. This result may be related to the domain structure. Since the stress is nearly absent and the clusters are not likely to form at such a short period of time, it is quite probable that the domain structure of samples annealed for 5 min is simple, a small number of domains (2 domains at the extreme case) being aligned in the length direction, which is is supported by the value of remanence ratio. The magnetization of these samples is expected to occur by the motion of 180° walls and, because of the simple domain structure, domain walls are hard to move at very small magnetic fields where the initial permeability is measured. Even a small movement of domain walls will only contribute to a small amount of magnetization. As a result of this, a very small value of the initial permeability is considered to be observed for the samples annealed for 5 min. However, a megnetic field higher than critical value is applied, domains will move fast because of small hindrances to wall motion, resulting in the observed low coercivity and high maximum permeability. As the annealing time increases, the clusters or precipitates are formed causing the domain refinement and, resultantly, domains will move rather easily at very small magnetic fields and/or the area of magnetization reversal will be large for a given distance of domain movement, resulting in the increased initial permeability.

It is seen from Fig.6 that the maximum permeability is highest at the annealing time of 5

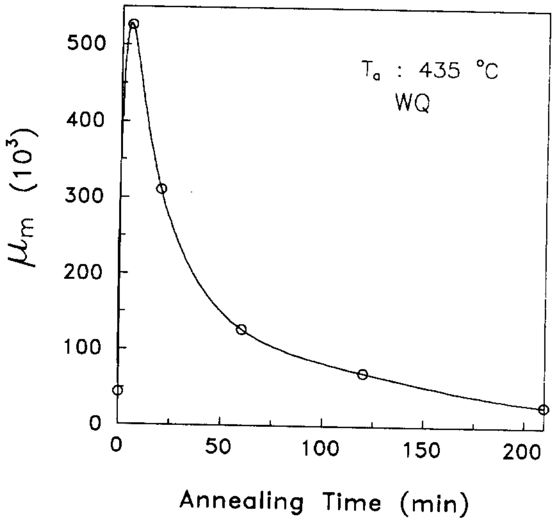

Fig.6 The value of the maximum permeability of Metglas 2605S3A as a function of annealing time. The annealing temperature is 435℃.

permeability is highest at the annealing time of 5 min and thereafter it monotonically decreases with the annealing time. The maximum permeability is high when the coercivity is low and the remanence ratio is high.

IV. CONCLUSIONS

The effects of annealing on the dc magnetic properties of the Fe-based amorphous alloy ribbon, Metglas 2605S3A (AlliedSignal Corp.) are investigated. The ribbons were annealed at a fixed temperature of 435°C but the duration of annealing was varied widely from 5 to 210 min. The residual stresses induced during melt-quenching are considered to be removed nearly completely at the annealing time of 5 min, resulting in typical square-type hysteresis loops, low coercivity and maximum permeability. As the duration of annealing increases further, the coercivity increases and the shape of hysteresis loops transforms to round type and finally to sheared one at the longest annealing time of 210 min. These results has been explained by considering the formation of clusters with chemical short order and very fine crystallites (at the annealing time of 210 min), and the atomic migration induced stresses during the formation of the clusters. Of particular interest is the very low initial permeability observed for the samples annealed for 5 min and this result is discussed by the simple domain structure of 180° domains.

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