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Relationship Between AC and DC Magnetic Properties of an Iron-Based Amorphous Alloy for High Frequency Applications

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The relationship between the effective permeability and the remanence ratio of an Fe-based amorphous alloy (Metglas 2605S3A) is investigated over a wide frequency range, in an effort to understand magnetization behavior of the alloy. In the frequency range from 1 to 200 kHz, the permeability is maximum at the remanence ratio of 0.4-0.5 and, at frequencies over 500 kHz, the correlation with negative coefficients emerges indicating that the permeability decreases with the remanent ratio, except for the ribbon coated with an insulating layer of MgO which exhibits both high values of the effective permeability and remanence ratio. It is considered from the correlation results that the boundary at which the dominant magnetization mechanism changes from domain wall motion to spin rotation is near 500 kHz. The core loss is also investigated as a function of annealing time when the samples are annealed at a fixed temperature of 435℃. The core loss in most cases decreases with the annealing time, the degree of the loss reduction increasing with the measured frequency. The present loss results indicate that the total loss may consist of the hysteresis loss and anomalous eddy current loss. The two loss components are considered to be of similar magnitudes at low frequencies while, at high frequencies, the dominant contribution to the total loss is the anomalous loss.

1. Introduction

Amorphous magnetic alloy ribbons which are usually produced by a rapid solidification process have many noble properties compared to crystalline alloys. One example of these noble properties is that the exchange interaction and magnetostriction of amorphous alloys are similar to those of crystalline alloys but the magnetocrystalline anisotropy of amorphous alloys is negligibly small. This is because the anisotropy is distributed randomly over a distance slightly larger than atomic spacing (about 1 nm) [1] resulting in a drastic reduction of the effective (or apparent) anisotropy [2]. This small effective anisotropy is one of the major and fundamental factors behind the excellent soft magnetic properties of amorphous alloys. Additionally, amorphous alloys exhibit low coercivity because of the ease of domain wall motion due to the structural uniformity (no defects) and they easily form induced anisotropies. Furthermore, amorphous alloy ribbons have very good high frequency characteristics, since the resistivity is high (3-5 times higher than crystalline alloys) and the thickness of the ribbons is small, compared to conventional crystalline alloys.

Amorphous alloy ribbons are frequently divided 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, while Co-based amorphous alloys have been used as high frequency core materials. In the late eighties, however, many efforts were made to use Fe-based amorphous alloys for high frequency 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 this purpose is $Fe_{76.5}Cr_2B_{16}Si_5C_{0.5}$ (Metglas 2605 S3A of AlliedSignal Corp., USA). It is considered to be important to understand magnetization behavior of the alloy, particularly at high frequencies, in order to use the alloy more efficiently.

The magnetization at low frequencies is known to occur mainly by domain wall motion and therefore the magnetic properties at low frequencies can be improved by removing obstacles to domain wall motion [3]. The magnetization at high frequencies, however, occurs not only by domain wall motion but also by spin rotation, the latter contribution becoming more dominant at higher frequencies [4, 5]. In an effort to understand the magnetization behavior of a Co-based amorphous alloy subjected to ac magnetic fields, the frequency dependence of relationship between the effective permeability and dc magnetic properties was recently investigated [5]. A clear correlation between the effective permeability and dc magnetic properties, particularly the remanence ratio, was found to exist and the frequency dependence of magnetization behavior was then understood from the variation of the correlation with frequency.

In this paper, a similar approach is applied to the Fe-based amorphous alloy of Metglas 2605S3A (AlliedSignal Corp., USA) and the correlation between the effective permeability and the remanence ratio is considered. Since a large number of data are required to examine the correlation more clearly, varying values of magnetic properties were obtained by annealing at various conditions which include changing the duration of annealing and post-anneal cooling method and magnetic field annealing. Magnetic properties were also obtained for the ribbons coated with an insulating oxide layer on the ribbon surface. The results for core loss which is an important magnetic property are also presented in the latter part of the paper.

2. Experimental

The width and thickness of the ribbons were 4.5 mm and about 20 μm, respectively. The ribbons were wound onto toroidal cores with inner diameter of 19 mm (which makes the inner diameter of magnetic core 19 mm). The number of wound layers was fixed to be 50. The cores were annealed at a fixed temperature of 435℃ but the duration of annealing was varied widely from 5 to 210 min. Immediately after annealing, the samples were water-quenched (WQ), air-cooled (AC) or furnace-cooled (FC). The annealing was carried out using an electric furnace whose heating wire was wound non-inductively not to produce any magnetic field. The furnace for magnetic field annealing consists of the non-inductively wound furnace and a separate apparatus for applying a magnetic field. The design of the latter apparatus differs depending on the direction of applied magnetic field; a solenoid coil was used for applying a magnetic field in the transverse direction and, in the case of length direction, a Cu rod which can carry high currents up to 150 amperes was used. The current produces a longitudinal magnetic field of about 30 Oe for the core size investigated in this work. The coating of an insulating layer of MgO on the ribbon surface was done by a sol-gel method. The precursor of a $Mg(OCH_2CH_2OCH_3)_2$ solution was prepared by refluxing a mixture of magnesium ethoxide and 2-methoxyethanol for about 30 min. and hydrolysis was carried out by adding a solution of water and alcohol to the Mg alkoxide precursor. Dc magnetic properties were measured with a hysteresis loop tracer and the properties measured were the remanence ratio (B_r/B_s where B_r and B_s are the remanent and saturation flux densities, respectively), coercivity (H_c), initial permeability (μ_i) and maximum permeability (μ_m). The ac effective permeability (μ_e) was measured by using an impedance analyzer at an applied field of 10 mOe and over a wide frequency range of 100 Hz to 1 MHz. The core loss was measured with a B-H analyzer (Iwatsu Co. Model No. SY-8216) at frequencies up to 200 kHz and at the flux densities of 0.3, 0.5 and 0.7 T.

3. Results and Discussion

3.1 Magnetic Properties at Various Conditions

In Fig. 1 are shown the results for μ_e as a function of frequency for the samples annealed for various periods from 0 (as-cast state) to 210 min. It is seen from the figure that the effective permeability of as-cast sample is very low and remains relatively constant in the whole frequency range, but the permeability is improved significantly by the annealing. The frequency dependence of the permeability differs with annealing time. For the samples annealed for 20 min and shorter, the effective permeability is very high at low frequencies but decreases rapidly with the frequency, exhibiting very low values of permeability at high frequencies. This tendency is more prominent for the samples annealed for shorter period of time. On the other hand, the effective permeability of the samples annealed for 60 min and longer is low at low frequencies but it remains nearly constant up to 10-200 kHz, after which it decreases steeply with the frequency. The frequency at which the permeability decreashes steeply increases with annealing time and the magnitudes of the frequency are 10, 50 and 200 kHz at the annealing periods of 60, 120 and 210 min, respectively. This

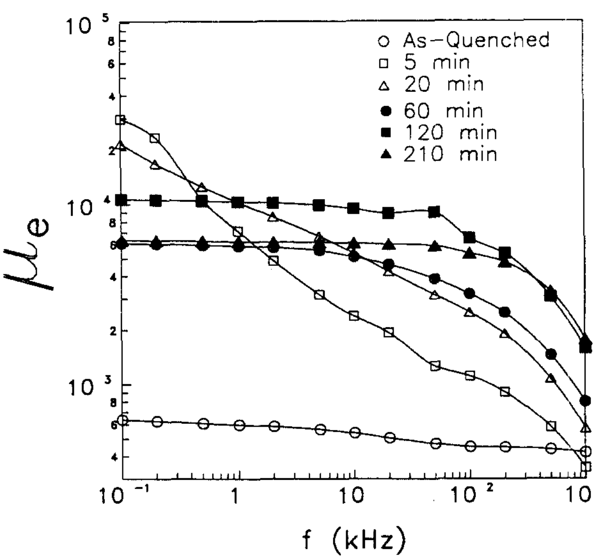


Fig. 1 The value of effective permeability of Metglas 2605S3A at various annealing times as a function of frequency. The annealing temperature is 435℃.

indicates that the high frequency permeability characteristics improve with annealing time. For example, the effective permeability at 1 MHz is highest at the longest annealing time of 210 min and it decreases with decreasing annealing time monotonically, the permeability of the sample annealed for 5 min being even smaller than that of as-cast sample at this frequency.

It would be of interest to see how dc magnetic properties change with annealing time. Only a brief summary of them is presented here, the detailed results being reported elsewhere [3]. The value of remanence ratio is 0.47 for as-cast sample and increases significantly to 0.75 at an annealing time of 5 min and then decreases monotonically with the further increase of annealing time. The coercivity of as-cast sample is very large (135 mOe) but is significantly reduced to 14 mOe at 5 min's annealing. As annealing time is increased further, the coercivity increases monotonically with annealing time. The initial permeability increases continuously with annealing time. The maximum permeability is highest at an annealing time of 5 min and thereafter it monotonically decreases with annealing time. It is seen from results for these dc magnetic properties and the permeability shown in Fig. 1 that the samples annealed for short periods of time (5 and 20 min) exhibit good dc magnetic properties but the permeability is relatively poor, particularly at high frequencies. The opposite trend is observed for the samples annealed for longer periods of time. This fact indicates that the main factors affecting dc magnetic properties and the ac effective permeability differ with each other.

The amount and type of residual stress and induced anisotropies are affected by cooling rate immediately after annealing. It is likely that high cooling rate causes more residual stress but less induced anisotropies. Anisotropies induced by magnetization will only be formed during cooling since the annealing temperature (435℃) is higher than the Curie temperature of the alloy (358℃). The cooling methods include water-quenched, air-cooled or furnace-cooled and the annealing time is fixed at 120 min. Dc magnetic properties obtained at different cooling methods are summarized in Table Ⅰ. Dc magnetic properties

Table Ⅰ. Dc magnetic properties of Metglas 2605S3A annealed at 435℃ for 2 hr and then water-quenched, air-cooled or furnace-cooled.

	Hc (mOe)	B_r/B_{10}	μ_m	μ_i
WQ	92.0	0.571	70426	10532
AC	84.0	0.472	65212	14318
FC	100	0.606	64211	7879

are affected only slightly as can be seen from the Table. The value of coercivity is 84 mOe when the sample is air-cooled which is compared to the values of 92 and 100 mOe for the water-quenched and furnace-cooled samples, respectively. The high coercivity values for the water-quenched may be due to the presence of large residual stress and those for the furnace-cooled samples due to the formation of large amount of clusters and/or domain wall stabilization. The remanence ratios are 0.47, 0.57 and 0.61, respectively, for the air-cooled, water-quenched and furnace-cooled samples. The high remanence ratio of furnace-cooled sample is due to large induced anisotropies. The highest initial permeability is observed for the air-cooled sample while the highest maximum permeability for the water-quenched sampie. The maximum permeability of furnace-cooled sample is low because of high coercivity, although the remanence ratio is high. In Fig. 2 are shown the results for the frequency dependence of effective permeability at the various cooling methods. The effective permeability of water-quenched sample is highest over the whole frequency range and that of furnace-cooled sample is lowest except for the very high frequencies of 500 and 1000 kHz. The loss is also affected only slightly by the cooling rate as can be seen from Table Ⅱ. The lowest loss is observed for the air-cooled sample.

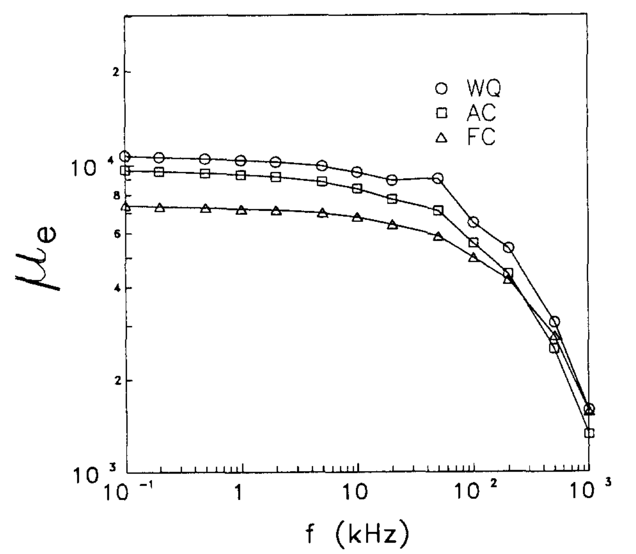


Fig. 2 The value of effective permeability of Metglas 2605S3A at various post-anneal cooling methods as a function of frequency. The samples were annealed at 435℃ for 2 hr.

In Fig. 3 are shown the results for effective permeability as a function of frequency for the samples annealed under no magnetic field and magnetic fields in the transverse and length directions. The duration of annealing is 120 min and the samples are furnace-cooled after annealing in order to form large induced anisotropies. It is seen from the figure that the highest permeability is observed for the samples annealed without magnetic field while, the lowest permeability for the samples annealed under magnetic field in the length direction. The frequency at which the permeability begins to drop steeply is also low being about 2 kHz when field-annealed in the length direction and this value is compared with 50 and 100 kHz in the cases of no field anneal and field anneal in the transverse direc-

Table Ⅱ. The core losses of Metglas 2605S3A annealed at 435℃ for 2 hr and then water-quenched, air-cooled or furnace-cooled.

Frequency	Cooling condition	Core loss (W/kg)		
		0.3 T	0.5 T	0.7 T
	WQ	0.58	1.58	3.42
1 kHz	AC	0.50	1.46	3.31
	FC	0.78	1.94	3.68
	WQ	9.55	27.52	56.6
10 kHz	AC	8.31	25.0	53.3
	FC	16.88	37.23	72.34
	WQ	295.85	860.38	1626.3
100 kHz	AC	276.54	847.15	1579.02
	FC	386.92	1083.39	1936.58
	WQ	761.86	2331.38	
200 kHz	AC	645.43	2153.74	
	FC	976.54	2867.82	

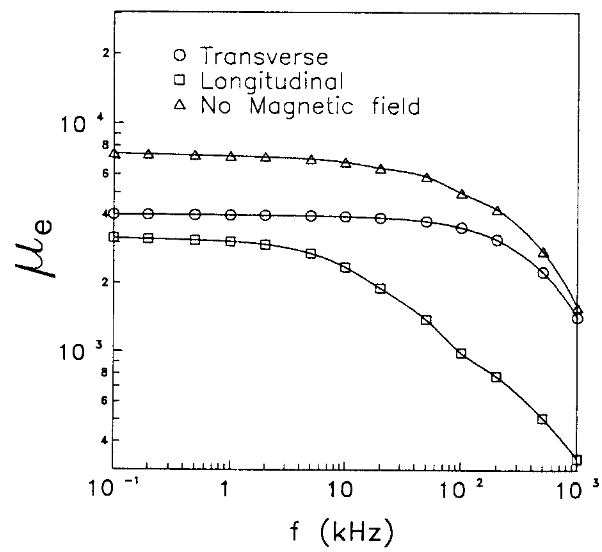


Fig. 3 The value of effective permeability of Metglas 2605S3A at various post-anneal cooling methods as a function of frequency. The samples were annealed at 435℃ for 2 hr.

tion, respectively.

The effects of surface coating are also investigated and the results are shown in Fig. 4. The results are for the samples annealed for 120 min and then water-quenched. The permeability of coated sample is slightly lower than that of uncoated sample at nearly all frequencies. The irregular behavior in the permeability versus frequency curves at about 50 kHz is considered to be due to the magneto-mechanical resonance which frequently occurs for alloys with large magnetostriction [6].

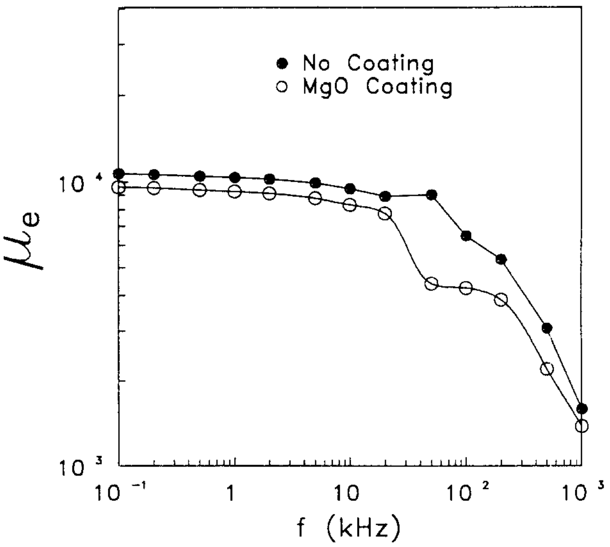
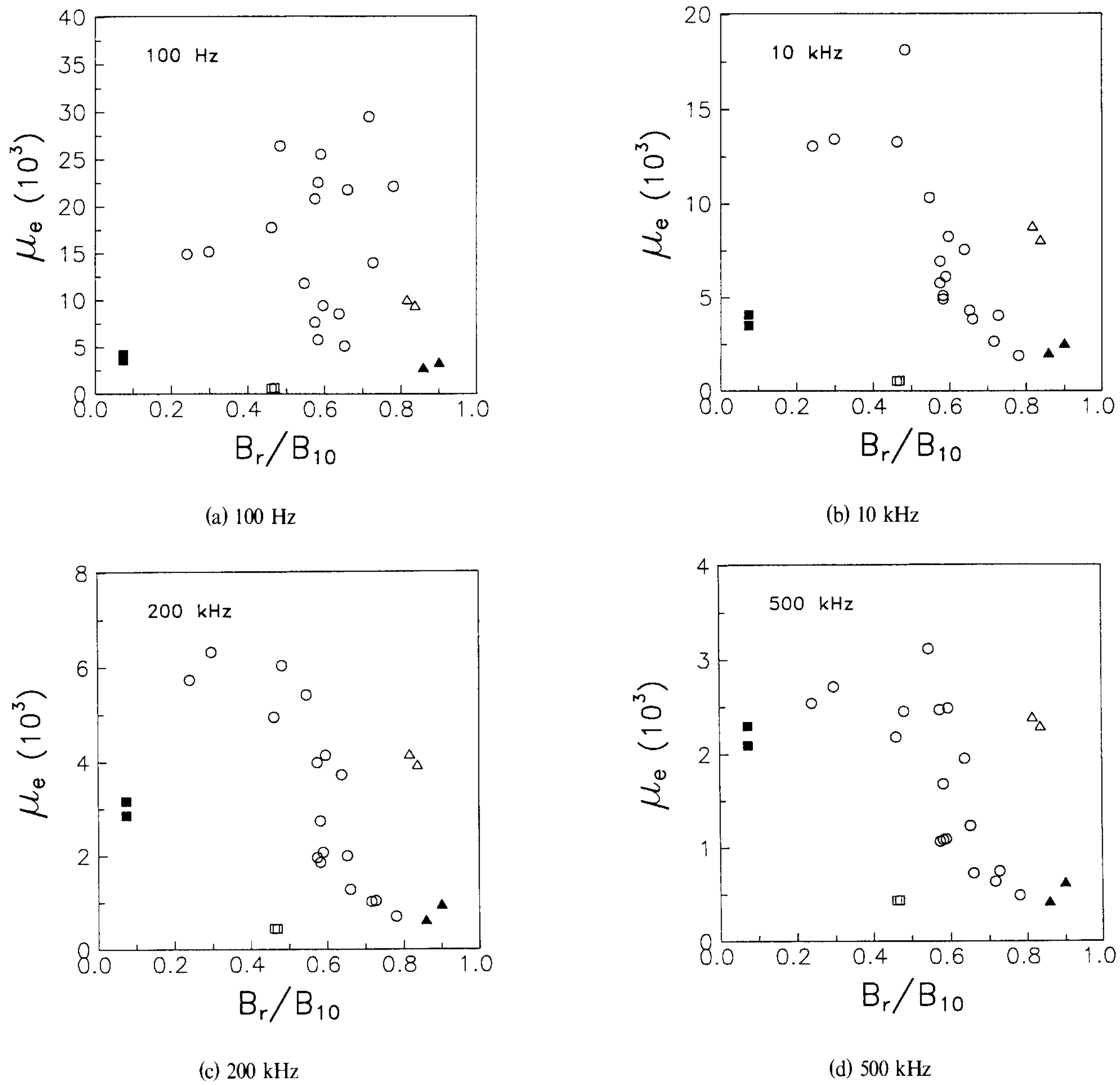


Fig. 4 The value of effective permeability of coated or uncoated Metglas 2605S3A as a function of frequency.
The samples were annealed at 435℃ for 2 hr.

3.2 Relationship between the effective permeability and the remanence ratio

With the results presented in the previous section, the relationship between ac and dc magnetic properties can now be investigated. Some of the results for the correlation between the effective permeability and the remanence ratio are displayed in Figs. 5 (a)-(d) at the frequencies of 0.1, 10, 200 and 500 kHz, respectively, although the results were obtained at many frequencies ranging from 0.1 to 1000 kHz. The results for the samples used to examine the effects of annealing time and post-anneal cooling method are indicated by unfilled circles. The field-annealed samples in the transverse and length directions are indicated by filled squares and filled triangles, respectively and the coated samples are indicated by unfilled triangles. The as-cast samples are indicated by unfilled squares. The very low remanence ratio and the very high remanence ratio observed for the field annealed samples (in the transverse and length directions, respectively) are due to the formation of induced anisotropies in the direction of applied field and these results are well in agreement with those reported previously [7, 8]. The high remanence ratio of the coated samples is considered to be due to the development of tensile stress in the length direction [9] which aligns the domains in the same direction by magnetoelastic interactions.

No clear correlation between the permeability and the remanence ratio is seen at 100 Hz. A correlation begins to emergy as the frequency is increased to 1 kHz, if the datum point of as-cast sample is not taken into account. The permeability increases with the remanence ratio, exhibits a maximum at the remanence ratio of 0.5 and then decreases with the further increase of the remanence ratio. This correlation behavior is simi-



(a) 100 Hz

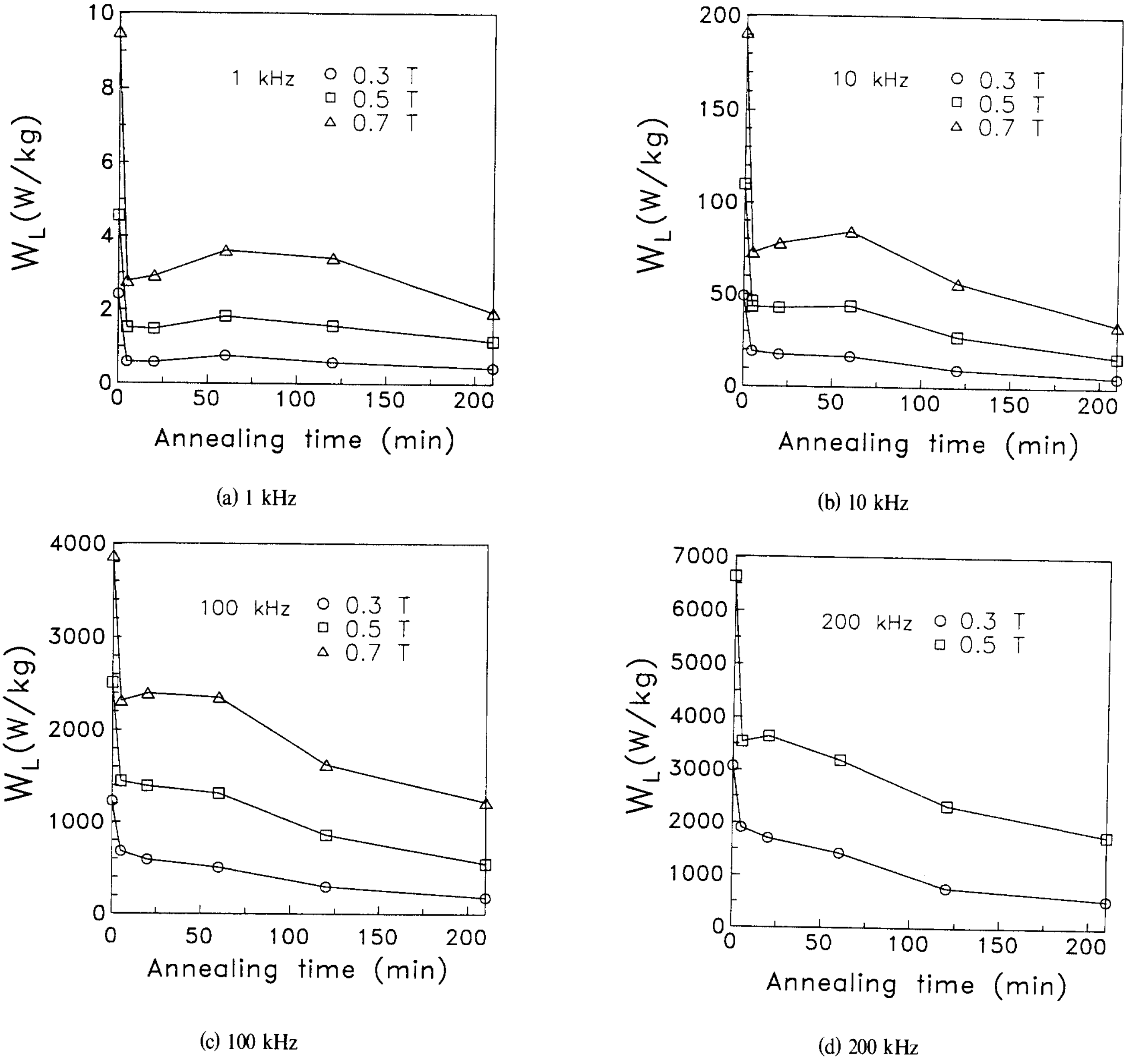
(b) 10 kHz

(c) 200 kHz

(d) 500 kHz

Fig. 5 The relationship between effective permeability and remanence ratio for Metglas 2605S3A. (a) 100 Hz, (b) 10 kHz, (c) 200 kHz and (d) 500 kHz.

lar to that observed previously for a Co-based amorphous alloy for which the maximum permeability was obtained at the remanence ratio of 0.5-0.6 [5]. It is noted here that the remanence ratio is a measure of the distribution of the magnetization direction of domains [10]. For ribbon type samples with 180o domains, the value of remanence ratio is 0.5 if domains are distributed randomly, and the values are 0 and 1 if domains are aligned completely in the transverse and length directions, respectively. This correlation behavior is maintained up to 200 kHz. It is noted, however, that the value of effective permeability for samples with a small remanence ratio decreases more slowly with increasing frequency than that for samples with a large remanence ratio does. For example, the permeability of the field-annealed sample in the transverse direction is very low at low frequencies but the relative magnitude increases with the frequency. Finally, at 1 MHz, the field-annealed sample in the transverse direction exhibits high permeability. Resultantly, at very high frequency of 500 kHz, the correlation behavior changes; a new correlation emerges with a negative correlation coefficient, viz., the permeability decreases with the remanence ratio. One exception is the coated sample which exhibits very high values of permeability and remanence ratio. The negative correlation coefficient observed in the present work indicates that the effective permeability is high when more domains are distributed in the transverse direction. A similar behavior was already observed for the Co-based amorphous alloy. It may be interpreted from the correlation results that the dominant magnetization mechanism at fre-



(a) 1 kHz (b) 10 kHz (c) 100 kHz (d) 200 kHz

Fig. 6 The value of core loss of Metglas 2605S3A as a function of annealing time. The annealing temperature is 435℃. (a) 1 kHz, (b) 10 kHz, (c) 100 kHz (d) 200 kHz

quencies higher than 500 kHz is spin rotation. This interpretation is supported by the fact that dynamic magnetization by spin rotation occurs easily when directions of applied field and domain are perpendicular to each other [4].

3.3 Core Loss

In Figs. 6 (a)-(d) are shown the results for core loss as a function of annealing time for the measured frequencies of 1, 10, 100 and 200 kHz, respectively. At each frequency, the loss results are shown for the flux densities of 0.3, 0.5 and 0.7 T. The core loss of as-cast samples is very high but is reduced significantly by annealing for 5 min at all the frequencies and flux densities measured in this work. The reduction in core loss increases with the measured flux density; for example, at 1 kHz and 0.3 T, the core loss of as-cast samples is 240 W/kg and that of the sample annealed for 5 min is 60 W/kg, while, at the same frequency of 1 kHz and 0.7 T, the loss of as-cast sample is 940 W/kg and that of the sample annealed for 5 min is 270 W/kg. The annealing time dependence of core loss for the samples annealed for 20 min and longer differs depending on measured flux density and frequency. At high flux densities (0.7 T and 0.5 T in the case of low frequencies), core loss initially increases slightly and then decreases gradually with annealing time. This tendency is more pronounced at lower frequencies and higher flux densities. At low flux densities (0.3 T and 0.5 T in the case of high frequencies), core loss decreases monotonically with annealing time, the degree of the decrease being larger at higher frequencies. At the frequencies and flux densities investigated in this work, the sample annealed for 210 min exhibits the lowest loss.

The loss of cores subjected to ac magnetic field may consist of hysteresis loss and eddy current loss. Again, the core loss due to eddy currents may be divided into classical eddy current loss and the anomalous loss [11, 12]. The classical eddy current loss is due to macroscopic eddy currents induced from homogeneous magnetization and is independent of domain structure. On the other hand, the anomalous loss is due to microscopic eddy currents, which are induced from the dynamic movement of domain walls and are proportional to the wall velocity. Since amorphous alloy ribbons have high resistivity and the thickness is small, the classical eddy current loss is much smaller than the anomalous loss [12-14]. The total loss of amorphous alloy ribbons may therefore be understood by considering hysteresis loss and the anomalous loss. Hysteresis loss and the anomalous loss are proportional to the frequency and the frequency squared, respectively, so that the former will dominantly contribute to the total loss at low frequencies but the latter contribution will be dominant at high frequencies. The large loss of as-cast sample is considered to be due to the large contribution by hysteresis loss, which is supported by the large value of coercivity observed in as-cast sample [3]. The reason for the steep increase in the loss with the flux density for as-cast sample may be related to the difficulty of magnetizing the sample, viz., a large field is required to obtain a given flux density. The two loss components contributing to total loss should be taken into account in the case of annealed samples. From the hysteresis loops given in ref. [3] and the coercivity results where H_c is lowest at the annealing of 5 min and then increases with annealing time [3], the hysteresis loss will be lowest at 5 min's annealing and then will increase with annealing time. On the other hand, the anomalous loss is expected to decrease with annealing time, since domains are considered to be simple and large at 5 min's annealing [3, 13] but they are refined due to clusters with chemical short range order and/or precipitates at longer annealing. The fact that the total loss does not vary substantially with annealing time at the frequencies of 1 and 10 kHz may be due to similar magnitudes of the two loss components at these frequencies. The broad maximum in the loss versus annealing time plots which occurs at 60 min's annealing and is observed at these low frequencies and high flux densities may be related to the steep increase in coercivity [3] and hence a similar increase in hysteresis loss. The loss decreases with annealing time at high frequencies and the trend is more pronounced at higher frequencies. This may be because the contribution by anomalous loss is important at high frequencies and the anomalous loss is low at long annealing time.

4. Conclusion

The magnetic properties of the iron-based Metglas 2605S3A amorphous alloy ribbons have been investigated at various conditions. The behavior in the frequency dependence of the effective permeability differs significantly with annealing time when the samples are annealed at 435℃. Good high frequency permeability characteristics are observed for the ribbons annealed for 120 min and longer. Only a slight difference in the magnetic properties is observed depending on the post-anneal cooling methods. The permeability is decreased by magnetic annealing but slightly increased by MgO coating on the ribbon surface. It has been observed from the results for the frequency dependence of relationship between the effective permeability and the remanence ratio that, in the frequency range from 1 to 200 kHz, the permeability is maximum at the remanence ratio of 0.4-0.5 and, at frequencies over 500 kHz, the correlation with negative coefficients emerges. This may indicate that the dominant magnetization mechanism changes from domain wall motion to spin rotation at about 500 kHz. The core loss in most cases decreases with annealing time, the degree of the loss reduction increasing with the measured frequency. The present loss results indicate that the total loss may consist of the hysteresis loss and anomalous eddy current loss. The two loss terms are of similar magnitudes at low frequencies but, at high frequencies, the latter contribution is dominant.

References

[1] A. Hernando and M. Vazquez, in: Rapidly Solidified Alloys, Ed. H. H. Liebermann (Marcel Dekker, New York, 1993) chap. 17.

[2] R. Alben, J. J. Becker and M. C. Chi, J. Appl. Phys. 49 (1978) 1653.

[3] Y. S. Choi, D. H. Kim, S. H. Lim, T. H. Noh and I. K. Kang, J. Korean Magnetics Soc. 5 (1995) 478.

[4] H. Fujimori, H. Morita, M. Yamamoto and J. Zhang, IEEE Trans. Magn. 22 (1986) 1101.

[5] S. H. Lim, Y. S. Choi, T. H. Noh and I. K. Kang, J. Appl. Phys. 75 (1994) 6937.

[6] M. Mitera, H. Fujimori and T. Masumoto, in: Proc. 4th Int. Conf. on Rapidly Quenched Metals Vol. Ⅱ, Eds. T. Masumoto and K. Suzuki (The Japan Inst. of Metals, 1982) p. 1035.

[7] B. S. Berry and W. C. Pritchett, Phys. Rev. Lett. 34 (1975) 1022.

[8] F. E. Luborsky and J. L. Walter, IEEE Trans. Magn. 13 (1977) 953.

[9] D. R. Thornburg and W. M. Swift, IEEE Trans. Magn. 15 (1979) 1592.

[10] S. Chikazumi, Physics of Magnetism (John Wiley & Sons, New York 1964) chap. 12.

[11] R. H. Pry and C. P. Bean, J. Appl. Phys. 29 (1958) 532.

[12] H. Pfutzner, P. Schonhuber, B. Erbil, G. Harasko and T. Klinger, IEEE Trans. Magn. 27 (1991) 3426.

[13] R. F. Krause and F. E. Werner, IEEE Trans. Magn. 17 (1981) 2686.

[14] V. R. V. Ramanan, J. Mater. Engr. 13 (1991) 119.