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Factors affecting the magnetic properties of consolidated amorphous powder cores

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It has been shown that it is possible to produce high density amorphous metal compacts on a routine basis, that have interesting soft magnetic properties. A study of some of the parameters determining their magnetic properties has been made. As expected, interparticle insulation and compact thickness affect the a.c. properties. Surprisingly, the magnetic properties were found to be relatively independent of compact density, in the range 73 to 98% of theoretical density. Low or zero magnetostriction alloys gave superior properties. If more than ~ 10% interparticle insulation is employed then the alloy composition is of less consequence. It is shown that a wide range of properties may be obtained.

1. Introduction

Metallic glasses have attracted considerable interest in recent years [1–3], and are finding several commercial applications. The magnetic Metglas† alloys, in particular, are of interest for transformer laminations because of their good soft magnetic properties.

A limitation of these materials has been that one of their dimensions must be small, in order to achieve the high cooling rates required to reach the amorphous state. A typical maximum thickness is around 50 μm. Consolidation techniques to produce bulk amorphous parts are, therefore, of obvious interest. Conventional powder metallurgy and related consolidation techniques cannot, be employed, as the metallic glasses of interest crystallize when exposed to temperatures of about 500° C. In recent years, techniques such as shock wave compaction [4, 5] of mechanically comminuted amorphous ribbon have allowed the production of three-dimensional amorphous solids. This opens up the possibility of using amorphous compacts in a variety of soft magnetic devices, for instance, in high speed motors, pole pieces, chokes and small transformer cores. Such compacts should have the attractive magnetic properties of the amorphous alloys plus the ability to be formed into shapes that cannot be obtained from the ribbon, thus extending the use of these alloys.

The first attempts to capitalize on combining the advantages of amorphous alloys and powder metallurgy involved the use of a plastic binder, which resulted in poor magnetic properties. This technique has problems that might be anticipated; the annealing temperatures required for the metallic glass represent relatively high exposure temperatures for the binder. If the binder does sustain these temperatures, residual stresses tend to be produced on the powder with resultant deterioration of magnetic properties. Deterioration of magnetic properties with stress is well documented [1–3].

The initial successes with consolidating amorphous alloys involved mechanically soft alloys; for instance, shock wave consolidation of $Pd_{78}Cu_6Si_{16}$ was achieved [4], as was warm pressing of $Cu_{60}Zr_{40}$ [6]. The more interesting magnetic alloys are much harder, with a Vickers hardness of around 1000.

†Metglas is the registered Trademark of Allied Corporation for amorphous alloys of metals and brazing metals.

TABLE I Typical properties for the annealed amorphous ribbon of the principal alloys investigated [13]

		Metglas alloy 2605-S2	Metglas alloy 2826-MB	Metglas alloy 2705-M
Density ($g\,cm^{-3}$)		7.18	8.02	7.89
Curie temperature (° C)		415	353	350
Saturation induction (kG)		15.6	8.8	7.2
Coercivity (Oe)		0.03	0.015	0.01
Resistivity ($\mu\Omega$ cm)		130	160	130
Saturation magnetostriction		27×10^{-6}	12×10^{-6}	$< 1 \times 10^{-6}$
Maximum permeablility, d.c.		590 000	750 000	–
Impedance permeability	(60 Hz)	50 000	130 000	–
at $B_{max} = 0.1$ T	(10 kHz)	7 000	9 000	22 000
	(50 kHz)	3 000	3 500	15 000

Some alloys of magnetic interest have been consolidated by warm pressing and extrusion [7], although, in the latter case, crystallization occurred. They have also been consolidated by dynamic shock wave compaction, via the impact of a high speed gas propelled punch [5, 8, 9] and via explosives [10]. A variation is explosive consolidation to produce a tube of material, the explosive being detonated at the tube exterior and an absorber being placed in the tube interior.

One might expect that compacts of metallic glass would have good magnetic properties, because the amorphous ribbons have good properties and because the powder, as produced by comminution of the ribbon, has a platelet-like shape. Spherical or irregular shaped crystalline iron powder, produced by atomization, has its magnetic properties improved if it is rolled to a platelet-like shape prior to compaction [11]. Although production of amorphous compacts has been reported, little or no detailed data have been given previously on their magnetic properties. This is probably because of the poor properties obtained; it being reported, for instance, that the magnetic properties of consolidated parts are severely degraded owing to the introduction of high stresses and shear bands which cannot be annealed out. The degree of this magnetic property degradation diminishes as the magnetostriction of the alloy approaches zero [12] sensitivity appears to be a problem with amorphous compacts. Annealing of the compact is, therefore, important, even more so than with ribbon, as there are more variables involved in producing the compact, stresses are introduced into the powder during comminution of the ribbon to powder and during consolidation.

The aim of the present work is to show that high density amorphous compacts may be produced on a routine basis, and that good magnetic properties may be obtained. The sensitivity of the magnetic properties to several variables is shown.

2. Experimental details

The primary alloys investigated were $Fe_{78}B_{13}Si_9$*, Metglas alloy 2605-S2 and $Fe_{40}Ni_{38}Mo_4B_{18}$*, Metglas 2826-MB. Some work was also carried out on $Co_{69}Fe_{4.1}Ni_{1.4}Mo_{1.5}B_{12}Si_{12}$*, Metglas alloy 2705-M. The first alloy is designed to be used in large quantities in ribbon form in transformer laminations. The second and third are specialized and more expensive alloys having, in ribbon form, good magnetic properties at higher frequencies [13]. In particular, alloy 2826-MB has low magnetostriction and alloy 2705-M has near zero magnetostriction. Some of the properties of these alloys are given in Table I.

The powders used were produced by pulverizing amorphous ribbon cast on a rotating chill block. This technique can produce powder of any desired particle size, with high cleanliness. Both ribbon and subsequent powder production have been demonstrated on a large scale over a period of years at Metglas Products and are commercially available.

The powders used were either − 14 to + 80 mesh (− 1400 to + 180 μm) or − 140 mesh [− 100 μm]. These are given the respective designations grade I and II.

It was found possible to consolidate these powders by a variety of techniques; for instance, isostatically pressing in evacuated cans, vacuum hot pressing or cold pressing [14]. In all cases, compact density could be increased by increasing the pressure. During the work, relatively large (> 80 mm diameter), high density parts were produced. All the magnetic studies were conducted on toroidal samples of varying thickness, with an

*Nominal at %.

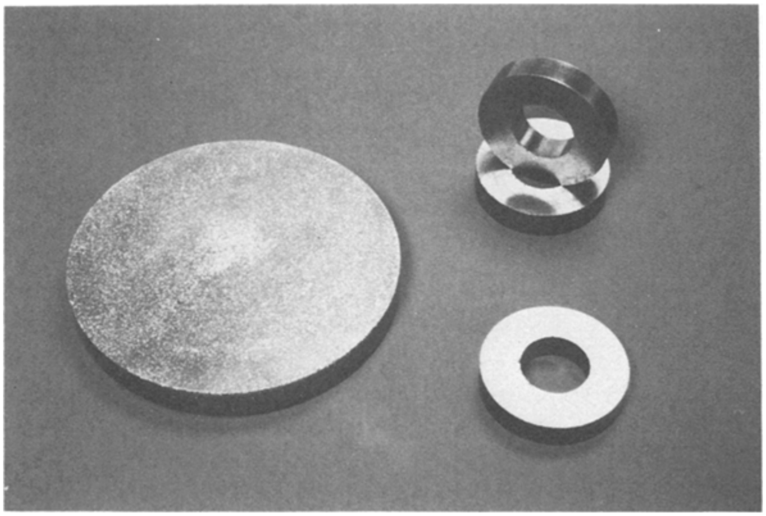


Figure 1 High density amorphous parts made from Metglas alloy 2605-S2 powder, showing an 80 mm diameter disc and typical 29 mm diameter test toroids.

external diameter of 28 mm and an internal diameter of 13 mm (Fig. 1). In the course of this work, several hundred such toroids were produced and tested. In this work all the parts are quasistatic compacts.

Initially, the amorphous nature of the annealed compacts was checked by X-ray diffraction. This was employed basically as a speedy quality control tool, repeat values of over 3% crystallinity warranted modification to the procedure. The magnetic properties themselves are a more sensitive measure, and much more useful in determining optimum conditions.

The compacts were annealed in the same general manner as the ribbon [1–3]. A magnetic field was applied in the circumferential direction by wrapping turns of copper wire around the toroid and passing a direct current through the wire, care being taken to avoid local heating effects. Annealing occurred under an inert atmosphere, in this case, nitrogen, and the annealing temperature was around the Curie temperature of the alloy (Table I). The magnetic field and inert atmosphere were maintained during cooling.

2.1. Testing procedures

The a.c. magnetic properties, such as core losses, exciting power and permeability were measured using the "Sine flux" or "Sine B" test described in IEEE Standard No. 106. This procedure was similar to that used for ribbon wound into toroids, a description of this is given in the booklet "Metglas Electromagnetic Alloys" [13]. The data were taken at $B_{max} = 0.1$ Tesla at varying frequencies. The toroids were wound with 10 turns in the primary coil and 10 turns in the secondary coil. The properties are calculated from standard formulae for toroidal samples.

A "Bridge" technique was also employed on some cores to evaluate the same properties. Results similar to those from the "Sin B" measurement were obtained on these cores. Therefore, the data reported are only from the "Sin B" measurement.

3. Results and discussion

3.1. Properties of loose powder

A set of experiments (Table II) was carried out on loose powder poured into a Plexiglas toroidal shaped mould. The data show that the properties

TABLE II a.c. properties of loose Metglas alloy 2605-S2 powder in a plexiglas toroidal mould

Particle size (mesh)	10 kHz				1 MHz			
	As-produced		Annealed		As-produced		Annealed	
	L (nH)	μ	L (nH)	μ	L (nH)	μ	L (nH)	μ
− 16 + 35	43.7	19.9	–	–	41.7	19.0	–	–
− 35 + 80	41.6	19.2	33.0	15.4	40.4	18.7	32.4	15.1
− 80 + 140	34.2	15.7	38.6	17.7	33.4	15.3	38.1	17.5
− 140	23.4	11.2	35.0	16.3	22.8	10.9	34.4	16.0
− 200	26.4	12.7	31.0	14.6	25.7	12.4	30.5	14.4

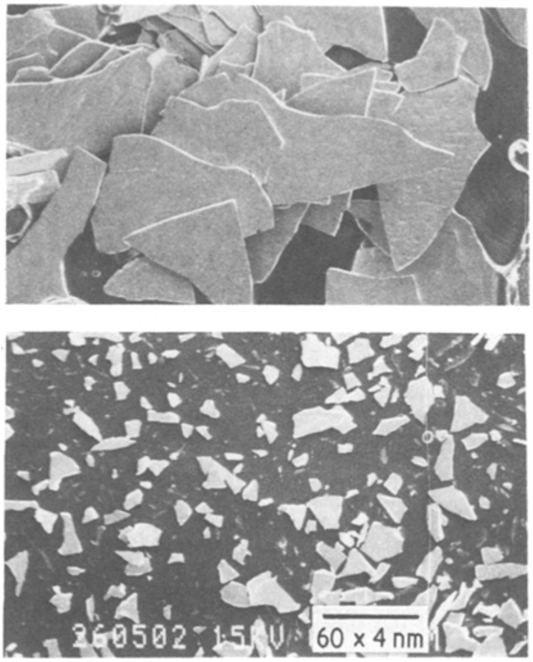


Figure 2 Scanning electron micrographs of the two commercially available grades of powder that were investigated. Note, the characteristic and desirable platelet/flake shape. Both powders are photographed at the same magnification.

are improved slightly by annealing the powder prior to placing it in the toroid. This anneal simply relaxes the elastic stresses. Field annealing the powder in the toroid would have been more effective; this is more easily carried out with a compact.

3.2. Effect of particle size

Both grades of powder have the characteristic platelet/flake shape resulting from being comminuted from thin ribbon (Fig. 2). This shape has been found advantageous in obtaining good magnetic properties for crystalline iron [11] and should also be advantageous for amorphous powders. It is emphasized that, because of the fabrication route, the particles, regardless of size, are amorphous.

The effect of particle size on permeability is illustrated in Fig. 3 where toroids from the two grades of powder are compared. In the case of crystalline iron, fine powder is undesirable [15–17]; this trend is seen to be also the case for amorphous alloys, though a larger difference in properties is seen. The poorer properties of fine powder are attributed to the difficulties associated with proper orientation of the powder particle and the larger demagnetization field. In this case, smaller particles may also have more elastic stresses remaining from the pulverization of the ribbon than large particles.

Subsequent data are for the larger particle size, grade I powder, which shows better properties.

3.3. Annealing

As stated earlier, the toroids were annealed under a magnetic field in a manner similar to that used for ribbon wound into a toroid [1–3, 13]. The annealing temperature–time cycles is an important variable, with the optimum conditions depending on the alloy composition and the compaction process. Fig. 4 illustrates the improvements in properties attainable by annealing the toroids.

3.4. Effect of the density of the compact

Density is an important parameter in determining the magnetic properties of crystalline powder cores [15–18]. To determine the effect of density

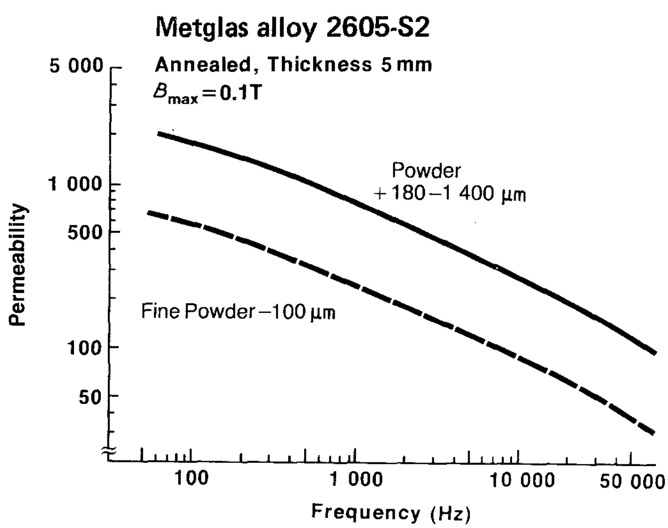


Figure 3 Illustration of the difference in a.c. properties between toroids 5 mm thick made from grade I (+ 180 − 1400 μm) and grade II (− 100 μm) powder.

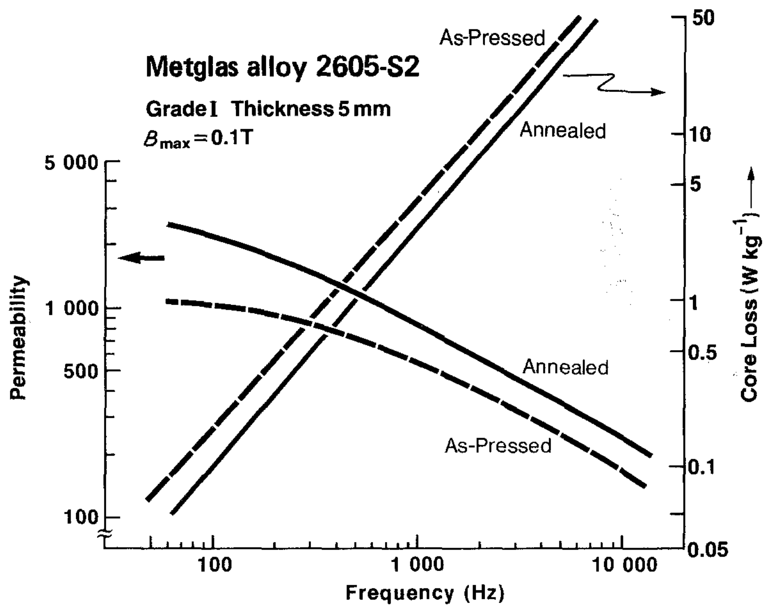


Figure 4 A comparison of as-pressed and annealed toroids. The same toroid was tested in the as-pressed and annealed condition.

on amorphous powder cores, toroids were produced to near theoretical density (TD) having little or no visibility porosity, as illustrated in Fig. 5. The properties across the compact were uniform and no frictional effects were observed.

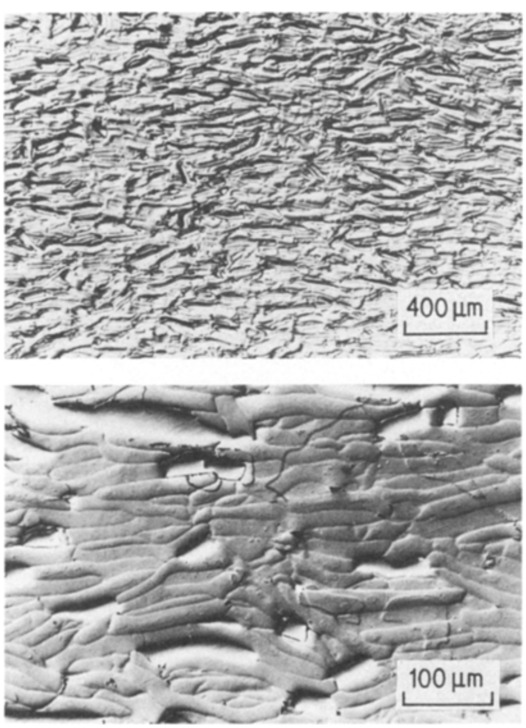


Figure 5 Micrographs of high density amorphous compacts of Metglas alloy 2605-S2 powder. Note the near absence of porosity, as expected for these 99% density compacts.

Surprisingly, our work found no consistent effect on the a.c. magnetic properties due to density over the range of densities investigated (73% to 98% TD). This is illustrated in Fig. 6 for toroids of similar size with 80 to 97% TD made from the same grade powder and subject to the same annealing cycle. This is probably because the benefits of increasing the amount of material within a given volume are cancelled by the increased stress on the particles and by the greater interparticle contact which increases eddy current losses.

Althought the a.c. properties appear insensitive to density, the d.c. properties are affected by it. The induction level at 100 Oe field, B_{100}, and the coercivity, H_c, both increase with density (Fig. 7).

The attainment of extremely high densities appears, therefore, unnecessary from the magnetic viewpoint. The need to provide for interparticle insulation will be described later. This insulation would automatically preclude attainment of theoretical densities. A benefit from high densities, is that the mechanical strength of the compacts increases rapidly as the density increases.

3.5. Effect of compact thickness

Thick bulk parts for a.c. applications have poor properties because of high eddy current losses. For such applications thin stampings are usually stacked together to form a bulk part. These laminations increase the elctrical resistance so that eddy current losses are low. For these applications the

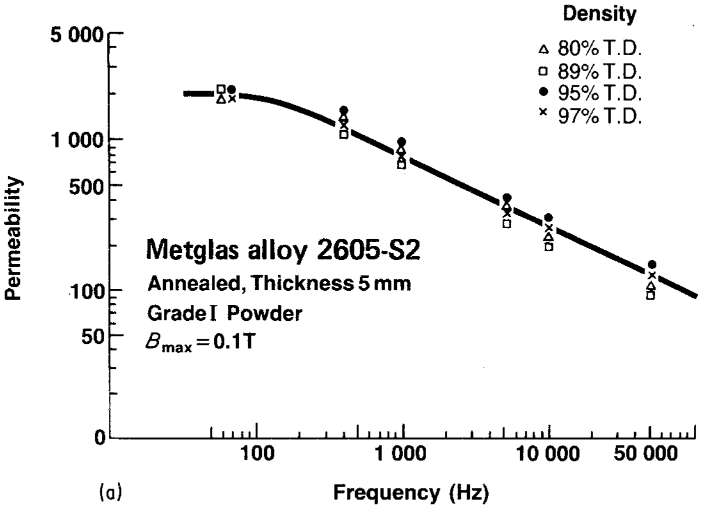
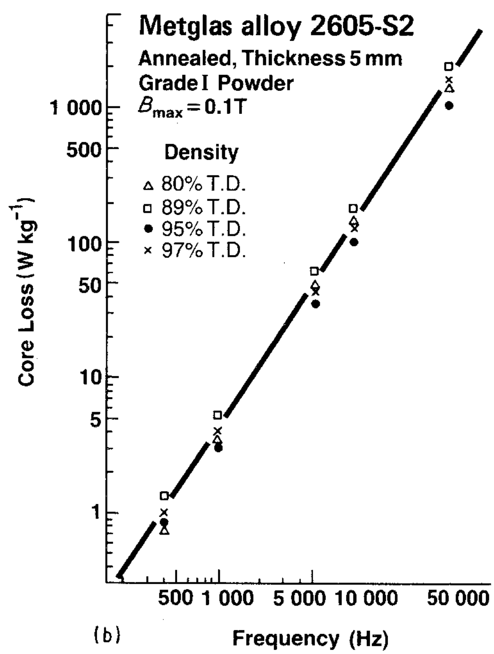


Figure 6 (a) and (b) The effects of density on the a.c. properties.



properties of both Metglas ribbon and iron silicon sheet depend upon their thickness [19]. For PM crystalline iron it is known that a way of reducing these eddy current losses is to make thin compaction [15, 16]. Fig. 8 illustrates the effect of thickness on the amorphous compacts. As expected, the thicker the compact, the poorer are the a.c. properties.

3.6. Effect of interparticle insulation

For a given thickness of compact the a.c. properties may be improved further by reducing interparticle contact so as to increase the electrical resistance. This is achieved by coating each particle with an insulator as shown in Fig. 9. This reduces the losses, especially for the thick toroids Fig. 10. It has been found that MgO and SiO_2 are effective insulators [14]; both decrease the core losses at high frequencies where the eddy current losses are most important (Fig. 11). Additions of over 10 wt% insulator result in a compact with a permeability which is relatively insensitive to frequency (Fig. 11a), although at the expense of poorer d.c. and low frequency properties.

3.7. Effect of alloy compositions

The importance of the alloy composition is seen in the d.c. properties. These were tested by standard

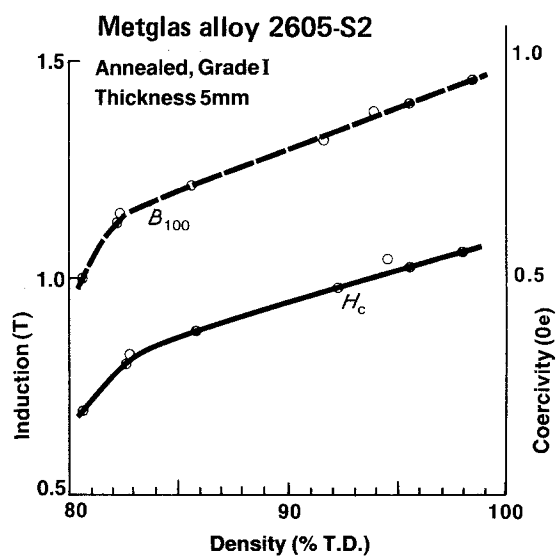


Figure 7 Variation of d.c. properties with density. Both the induction measured at 100 Oe and the coercivity increase with density. For comparison, B_s for the ribbon is 1.56 T (Table I).

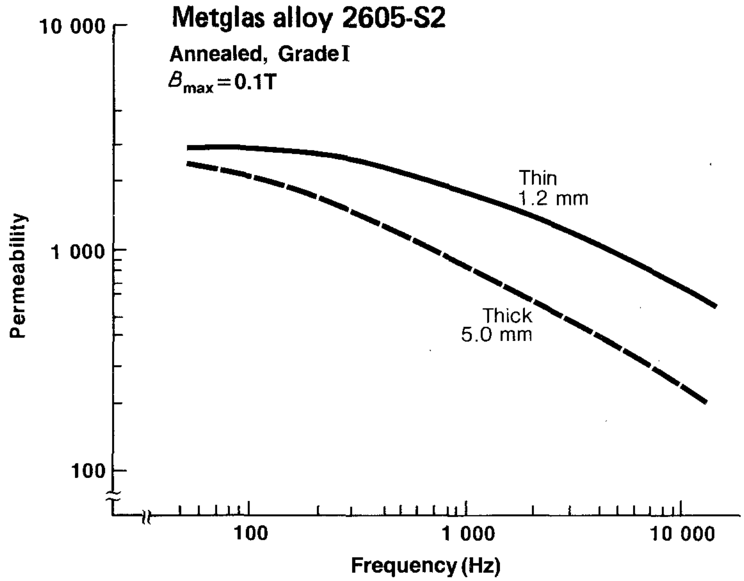


Figure 8 The effect of compact thickness on the a.c. properties.

techniques. The d.c. hysteresis loop of annealed toroids (Fig. 12) shows the difference between alloy 2605-S2 and alloy 2826-MB. The effect of adding insulation is also shown. It appears that the insulation has no appreciable effect on the coercivity (Table III). As expected, the low magnetostrictive alloy 2826-MB has lower values of coercivity.

Similarly, at low levels of insulation the low magnetostriction alloy gives superior a.c. magnetic properties. Table IV, compares alloy 2605-S2 with a zero magnetostriction alloy 2705-M. The compacts in Table IV were not annealed and were produced by a slightly different pressing condition than those in Figs. 10 and 11.

The effect of a high degree of insulation is to reduce the importance of the alloy composition. For example, a series of Metglas alloy 2605-S2 toroids with 15% insulation had average losses at 20 kHz and 0.1 T of 18.0 $W kg^{-1}$ and an average permeability of 352; these are very comparable to the average values obtained under the same conditions for Metglas alloy 2826-MB with 15% inulation, i.e. 16.2 $W kg^{-1}$ and 336. Therefore, for high frequency applications it is possible to use the lower cost Metglas alloy 2605-S2 rather than the alloys which give the best high frequency properties in ribbon form.

4. Comparison with other materials

It is difficult to compare the present data with previous work on amorphous compacts, because of the scarcity of published data. However, it does appear that the present properties are at least an order of magnitude superior. For example, the best published data [10] gives for annealed toroids a coercivity of around 5.5 Oe which compares to our values of around 0.4 Oe for the same alloy (alloy 2605-S2). This improvement is believed to be a result of superior consolidation and annealing techniques.

TABLE III Some typical d.c. properties of annealed toroids made from grade I powder. Alloys with high and low magnetostriction are compared both with and without insulation

Material Metglas alloy	Thickness (mm)	Density ($g cm^{-3}$)	H_c (Oe)	B_r (kG)	B_5 (kG)	B_{100} (kG)
2605-S2	5.5	6.9	0.4	2.4	6.0	13.5
2605-S2	2.6	6.49	0.7	2.3	5.4	12.8
2605-S2 + 6% Insulation	2.2	4.75	0.6	0.8	2.5	9.2
2605-S2 + 15% Insulation	2.92	4.68	0.40	0.30	2.3	8.6
2826-MB	2.3	6.96	0.14	0.85	5.2	7.3
2826-MB + 15% Insulation	3.2	4.93	0.18	0.05	0.80	4.8

TABLE IV Some a.c. properties of a positive magnetostriction alloy, Metglas 2605-S2, and a zero magnetostriction alloy, Metglas alloy 2705-M. The toroids were tested as pressed, i.e. without annealing

Metglas alloy	Insulation	Density ($g\,cm^{-3}$)	Thickness (mm)	d.c. properties		a.c. 60 Hz		a.c. 1 kHz		a.c. 10 kHz	
				H_c (Oe)	B_r (kG)	μ	losses ($W\,kg^{-1}$)	μ	losses ($W\,kg^{-1}$)	μ	losses ($W\,kg^{-1}$)
2605-S2	None	6.9	5.5	0.75	2.3	1200	0.075	600	7	150	250
2605-S2	4%	5.9	5.5	0.73	1.3	1000	0.025	900	2	280	100
2705-M	None	7.1	3.1	0.17	2.0	5000	0.02	1500	1.5	350	60
2705-M	4%	6.5	3.1	0.28	1.0	2000	0.01	1600	1.0	400	50

In contrast, comparison with existing soft crystalline magnetic materials is complicated by the wealth of data available on the many different materials, each of which is available in several grades from numerous manufacturers. Therefore, comparison has been restricted to PM iron and soft ferrites. A range of typical properties for these widely available materials is shown in Fig. 13 compared to the range of properties obtained for the amorphous toroids. The present properties lie between those of PM iron and soft ferrites. Compared to PM iron a much higher permeability and lower core losses are obtained; and, the amorphous cores achieved much higher induction levels (1.3 to 1.5 T) (Fig. 13) than the ferrites (0.3 to 0.5 T).

5. Conclusion

It has been shown that powders made from Metglas ribbon may be routinely consolidated to amorphous bulk parts which have good magnetic properties, expecially if an interparticle insulation is used. The effect of some parameters, such as particle size and compact thickness follow trends found in conventional PM crystalline material. The density of the compacts is seen to have little effect on their a.c. properties; more work is in progress, investigating this surprising observation.

The magnetic properties of the compacts are shown to improve as the magnetostriction of the alloy is reduced. However, at high degrees of insulation (⩾ 10%), the observed magnetic properties, though good, become independent of alloy composition. Amorphous alloys such as Metglas alloy 2605-S2 are available in relatively large quantities, while consolidation, especially to medium densities, has been found to be possible by techniques amenable to mass production. Amorphous compacts could, therefore, be used for some of the existing applications for powder metal soft magnets [17].

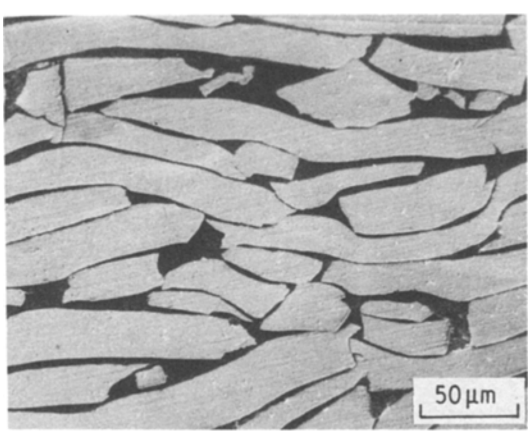


Figure 9 Micrograph showing interparticle insulation (dark areas) in a high density amorphous compact of Metglas alloy 2605-S2.

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The assistance of many colleagues at Allied Corporation is gratefully acknowledged. Thanks are

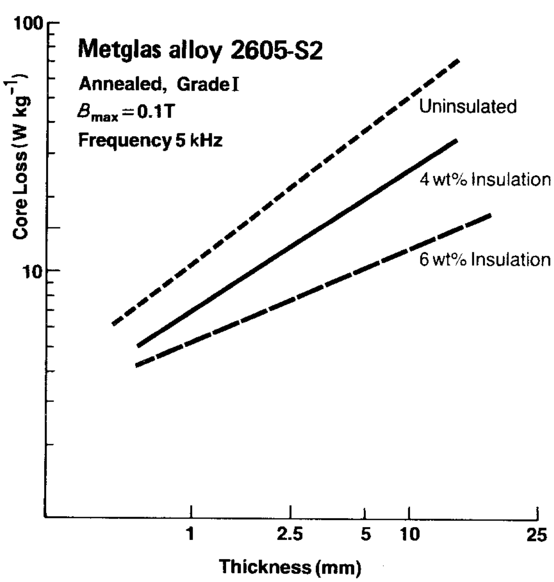

Figure 10 The effect of interparticle insulation on core losses for toroids of various thicknesses.

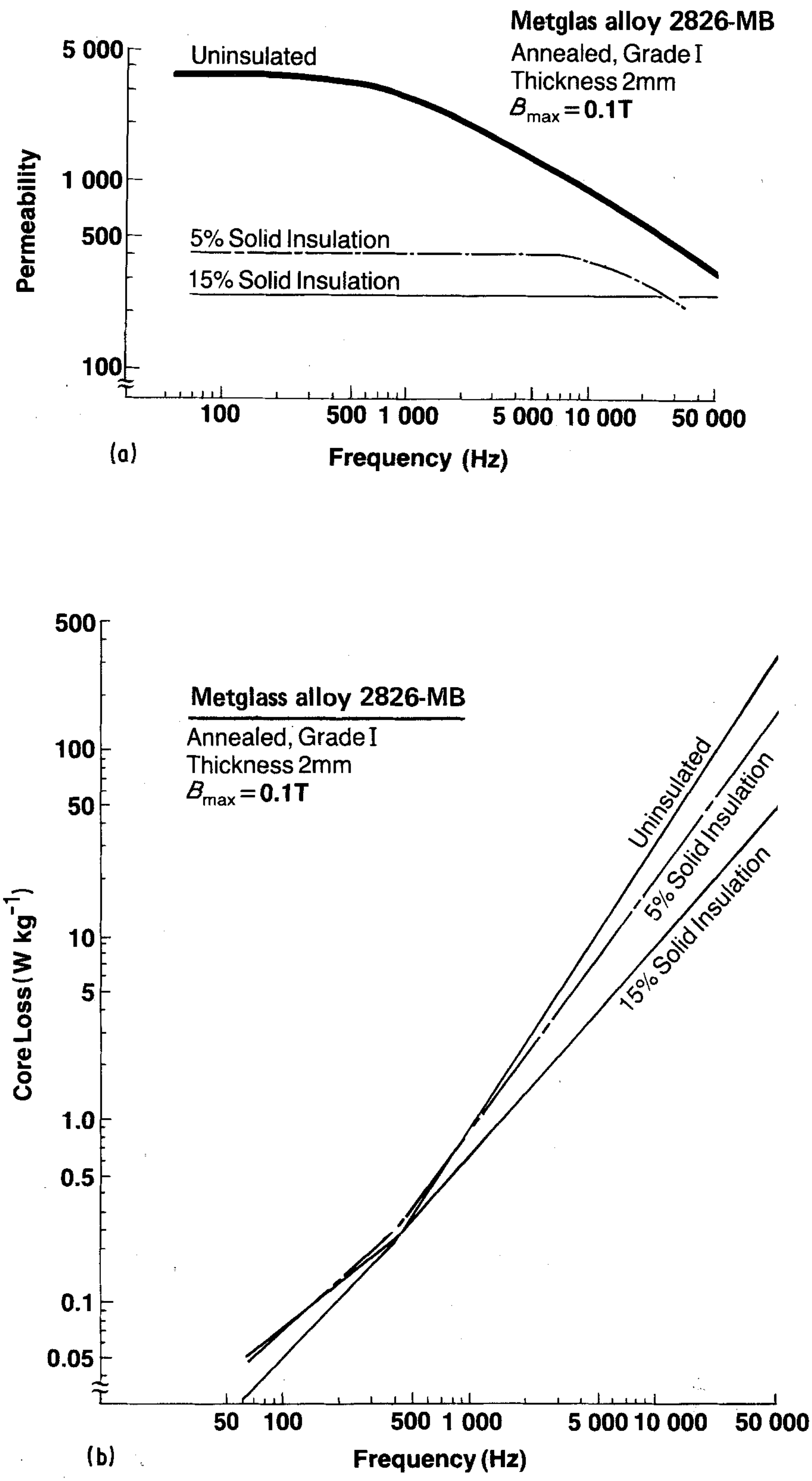

Figure 11 (a) and (b) As the insulation additions increase the permeability becomes relatively insensitive to frequency. Data are for the low magnetostriction alloy Metglas alloy 2826-MB, but similar results are obtained for Metglas alloy 2605-S2.

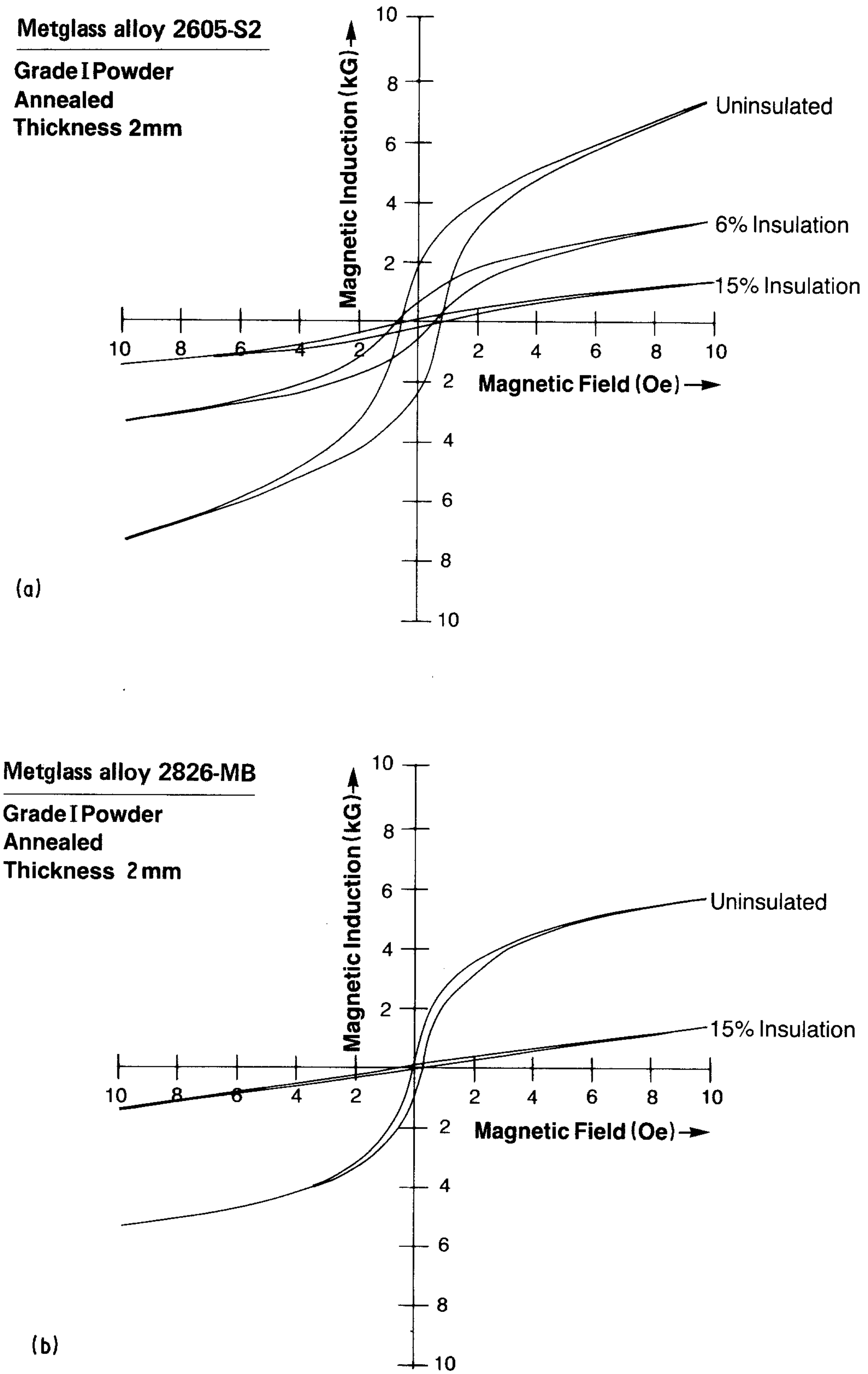

Figure 12 The d.c. hysteresis loops for annealed Metglas alloy 2605-S2 (a) and Metglas alloy 2826-MB (b), for insulated and uninsulated particles.

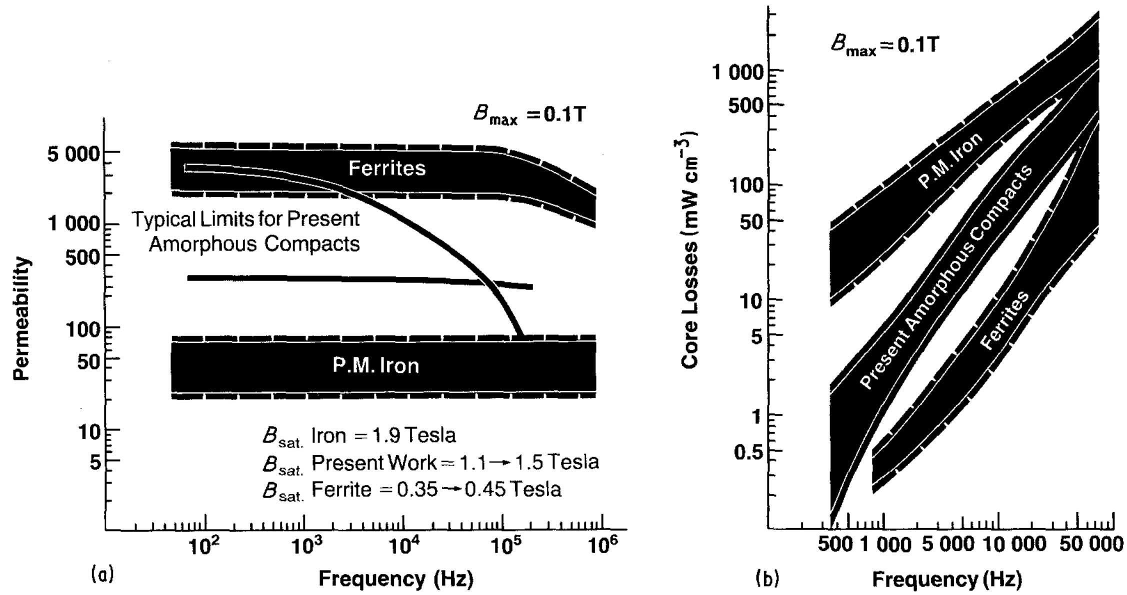

Figure 13 (a) and (b) A comparison of typical values of core loss and permeability for the amorphous toroids of our study with a typical range of values obtained with soft magnets made from P.M. iron and ferrites. The amorphous compacts have a saturation induction level three times higher than that of the ferrites.

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