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An investigation by Auger and laser acoustic microscopy of the bond between layers of consolidated amorphous ribbon (Powercore)

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It has been found possible to consolidate layers of amorphous ribbon to produce a thick strip which does not require annealing, and has similar magnetic properties to those of the much thinner ribbon. The reduced importance of eddy current losses that this implies must be due to the nature of the bond between the layers of ribbon. An extensive investigation of this bond has been carried out using Auger microscopy and laser acoustic microscopy. These show that bonding is localized and involves local plastic flow into ribbon irregularities, and the probable formation of a silicon oxide "glue". These explain the low eddy current losses and the improved pack factor of the strip compared to that of the starting ribbon. The uniformity of both mechanical and magnetic properties is also improved. Successful trials of the strip have been undertaken. This work has been jointly funded by the Electric Power Research Institute (EPRI) and Allied Corp.

1. Introduction

Amorphous metals are presently being manufactured as thin ribbons which have excellent soft magnetic properties. Distribution transformers made from alloys such as Metglas* alloy 2605-S2 have typical core losses 75% lower than conventional materials, such as grain oriented silicon steel. This significant saving has been confirmed on prototype transformers with sizes ranging from 10 to 50 kV A which have been recently constructed by several companies. Appreciable savings are also possible with the much larger 500 kV A plus power transformers. Ribbon thickness constraints imposed by the rapid solidification conditions necessary to produce amorphous ribbon have, however, reduced the economic attractiveness for the larger stacked transformer. In particular, the amorphous ribbon is thinner than the silicon steel presently used, [1×10^{-3} inch (0.02 mm) compared to around 12×10^{-3} inch (0.3 mm)], so existing transformer manufacturing equipment would have to be modified, and then would have to lay down an order of magnitude more material. This would also reduce the amount of material that could be packed into a given space, thus increasing the physical size of the transformer.

These problems have been largely overcome during an EPRI† sponsored research programme on the consolidation of amorphous ribbon. This process allows thick amorphous strip to be made with magnetic properties similar to those of a single annealed amorphous ribbon. For a 5×10^{-3} inch (0.125 mm) strip, typical losses are about 0.22 W kg^{-1} and the exciting power is about 0.4 V A kg^{-1} measured at 1.4 T and 60 Hz. The magnetic properties have been measured by both U core single strip and by Epstein frame techniques. This Powercore‡ strip can be used as a direct substitute for silicon steel; it can not only be supplied to the desired thickness, but also supplied already annealed; in which state, it can be readily handled. Its clear advantage is that, for the same thickness of about 10×10^{-3} inch (0.25 mm), it has losses of less than 0.30 W kg^{-1} at 1.4 T and 60 Hz compared to 0.90 W kg^{-1}, or over, for M4 silicon steel, Fig. 1.

2. Experimental details

For this investigation, layers of Metglas ribbon were consolidated by the application of pressure and temperature to form 1 and 2 inch (25 and 50 mm) wide strip of 2×10^{-3} to 20×10^{-3} inch (0.05 to 0.5 mm) thickness. Metglas alloy 2605-S2, nominal composition $Fe_{78}B_{13}Si_9$ (at %), was used as it is a low cost alloy intended for use in transformer applications. During this investigation strips from many consolidation runs and ribbon batches were investigated; all were observed to have the same basic consolidation features. Processing differences are believed, therefore, not to affect the nature of the bond between the layers of ribbon.

The bond was observed by snapping the strip and then peeling the layers of ribbon apart. As a comparison, as-cast ribbon was also investigated.

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†Electric Power Research Institute (Palo Alto, CA 94303, USA).
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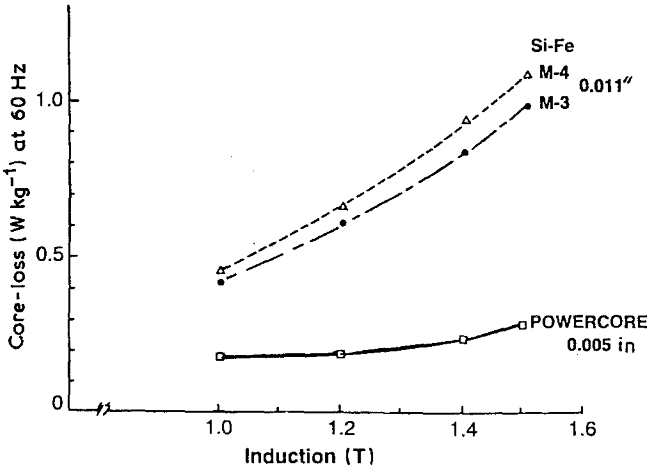


Figure 1 A comparison of the core losses of Powercore strip and silicon steel. Note that Powercore strip with a similar thickness (0.01 inch) to that of the silicon steel has a core loss at 1.4 T of less than 0.30 W kg^{-1}.

Surface analysis was used to study the nature of the bond, in particular, with scanning Auger microscopy (SAM). A SAM-SIMS instrument, model 595 from Physical Electronics (Eden Prairie, MN 55344, USA) was used. The depth dependence of the elemental composition of localized surface areas was analysed by Auger depth profiling. The scanning electron microscope capability of the SAM was employed to obtain images of the interface which could be correlated to the chemical information.

The etch rate of the ion gun (used for Auger depth profiling) was calibrated to a SiO_2 layer of known depth. Both xenon and argon were used as gas sources for the ion gun.

Complementary work was carried out using scanning laser acoustic microscopy (SLAM). This technique transmits acoustic waves through the specimen, and these are observed by laser interferometry. This allows resolution in the micron range and a real time display on a CRT of a picture resembling the object. This picture shows areas of bonding as light areas, and unbonded areas through which the acoustic wave cannot pass as dark areas. This investigation was carried out by Sonoscan (Bensenville IL 60106, USA) personnel in the author's presence. Most of the work was carried out in the amplitude mode at 100 MHz. The amorphocity of the Powercore strip was confirmed by X-rays during the initial work. The magnetic properties were, however, continuously evaluated and these are a more sensitive indicator of the commencement of crystallization.

3. Results

3.1. As-cast ribbon

The virgin Metglas alloy 2605-S2 ribbon was examined by SAM and found to have the usual morphology. The shiny side had a smooth wavy appearance, and the dull side had the usual valley formation, Fig. 2, formed during casting when air is trapped between the casting wheel and the ribbon. The thin oxide layer, typical of as-cast ribbon, was found to be present. Its depth was measured as around 5.0 nm on the dull (wheel) side, and around 7.0 nm on the shiny (air) side.

3.2. Powercore strip

3.2.1. Auger microscopy

Examination of the bond between the ribbon by SAM showed no areas of metallic fracture as is, for instance, found in a well bonded powder metal compact. No evidence of ductile dimpling or tearing, as would be associated with a good metal to metal bond, was found on any of the many debonded laminate surfaces examined, even as an isolated example. Instead, the laminate fracture surface was "smooth" and similar to the as-cast ribbon morphology. The valley formation characteristic of the dull side of the as-cast ribbon was found to be present in all of the laminated samples

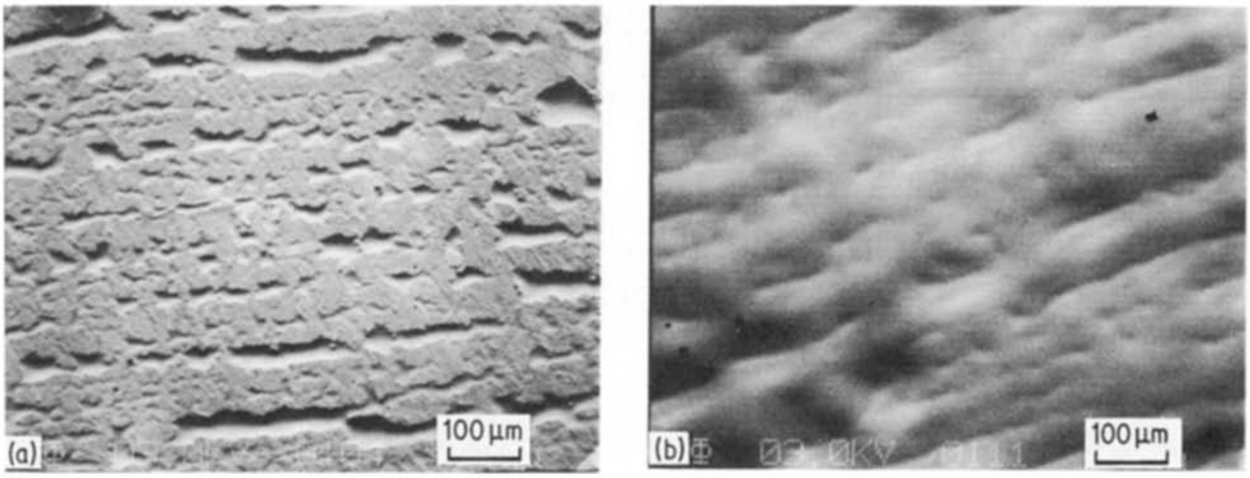


Figure 2 (a) and (b) scanning electron micrographs of typical surfaces of the as-cast amorphous ribbon.

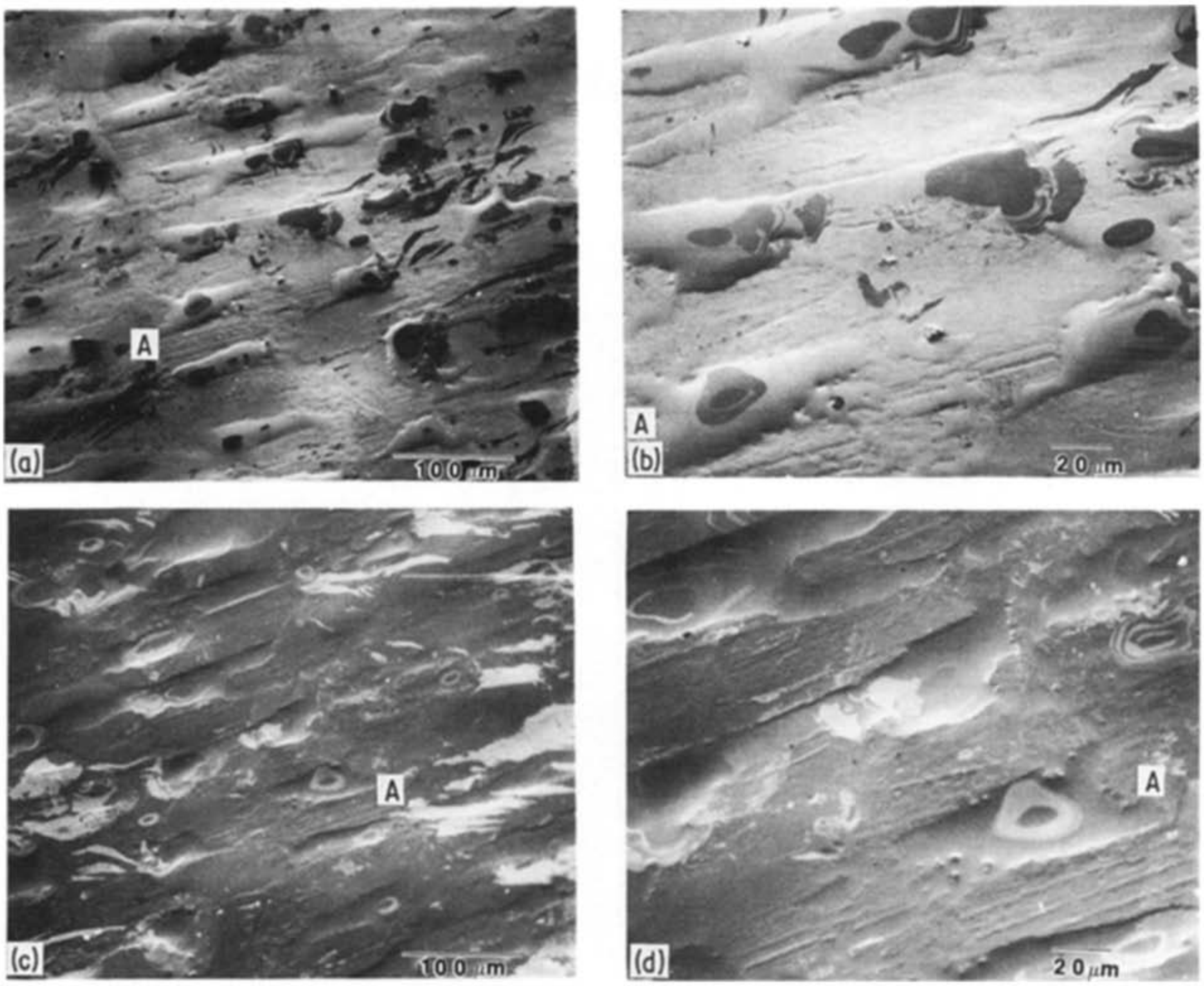


Figure 3 Matching micrographs from the two faces of a delaminated amorphous strip. The shiny side (a) and (b) has been moulded to take the shape of the dull side (c) and (d).

investigated. During fabrication, it was naturally convenient to place the ribbons on top of each other, dull side to shiny. This allowed the ribbons to fit together easily due to their slight curvature. The micrographs show that during consolidation the flat shiny side of the as-cast ribbon was moulded to take on the shape of the valleys in the dull side of the as-cast ribbon, Fig. 3. This is probably a result of less flow being required to form the hills, compared to removing the valleys.

The bonded surfaces were also characterized by light contrast areas, Fig. 3. These light areas were found to be consistently richer in oxidized silicon than the background. This is clearly seen in the silicon and iron Auger maps of Fig. 4. The observed shift in the kinetic energy of the LMM Si Auger line is consistent with that observed for SiO_2 [1]. The background areas are mainly an iron oxide of undetermined stoichiometry. Auger depth profile analysis showed that the SiO_2 concentration is a surface phenomenon disappearing as one enters the ribbon. This is illustrated

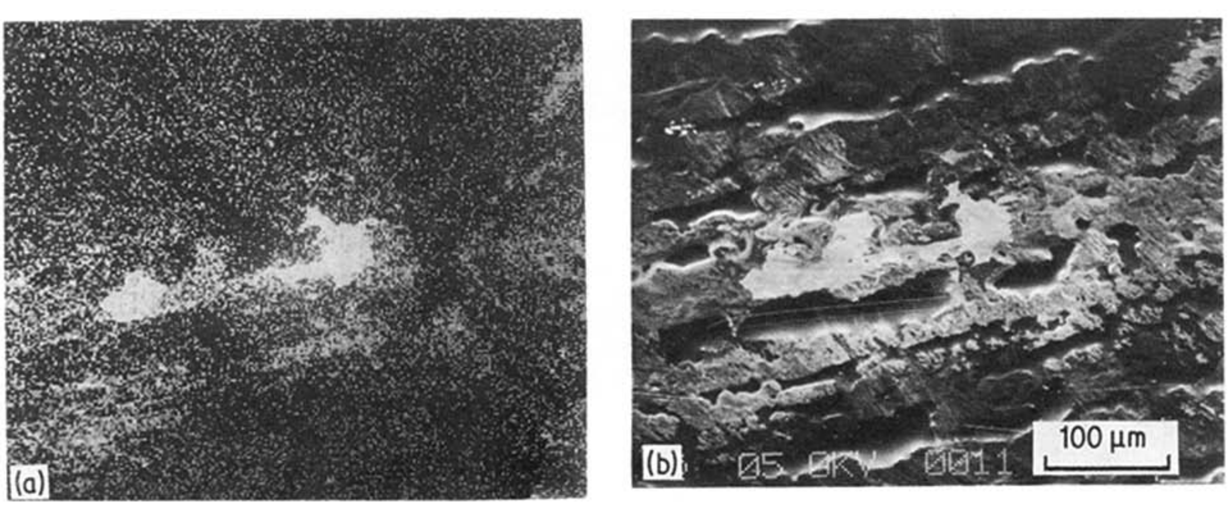
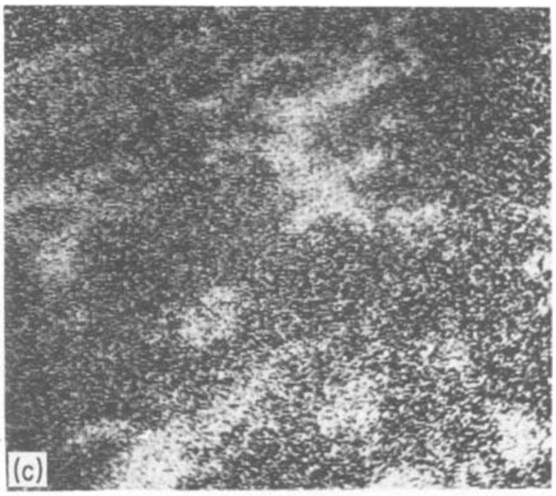
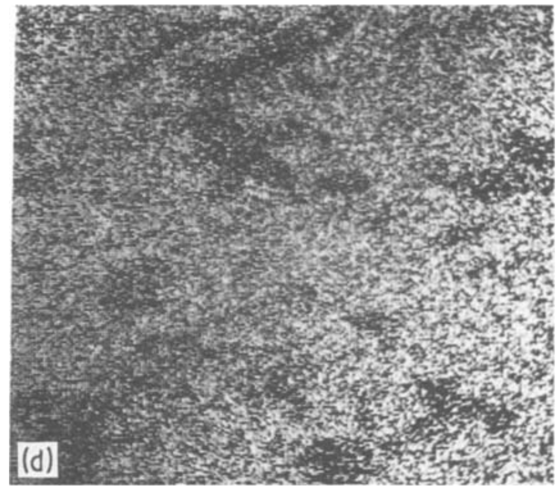


Figure 4 (a) An Auger map of (b) showing that the light areas have a high silicon concentration. Auger maps showing the (c) iron and (d) silicon distribution of (e).





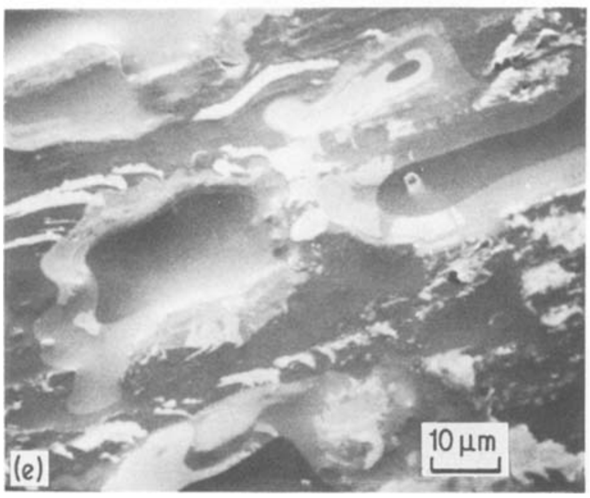


Figure 4 Continued.

in Fig. 5, where the SiO_2 distribution is shown, and in a graphical depth profile in Fig. 6. Such analysis showed that the majority of the surface had a high silicon concentration. A few cases of increased boron concentration were also observed, but this was not usual.

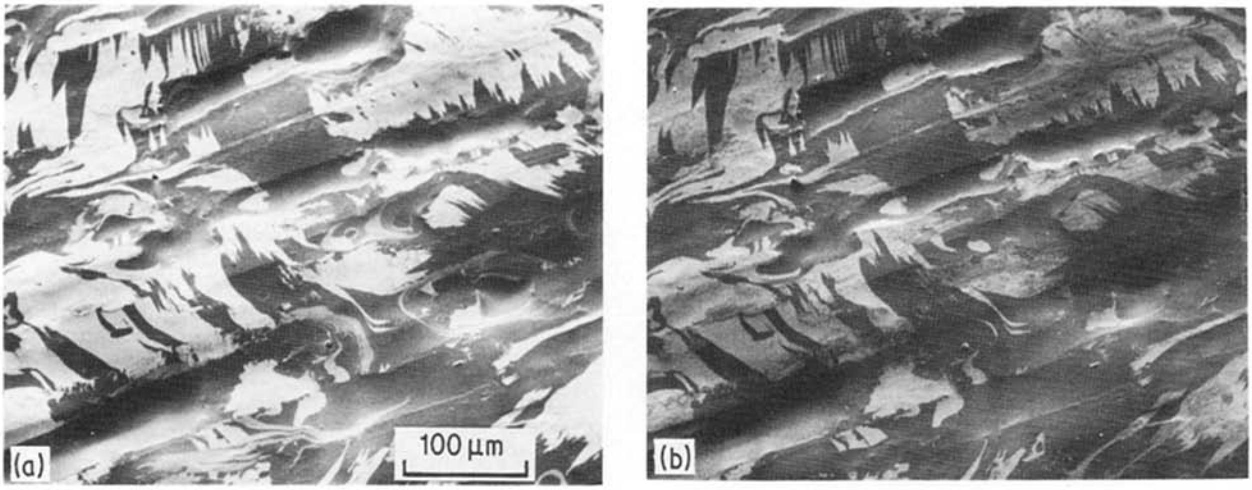
The depth of the oxide layer, and, in some cases, of the silicon concentration were measured by depth profiles of the type shown in Fig. 6. The results, Table I, show that the depth of the oxide layer is increased by the consolidation process. A single ribbon subjected to the consolidation process had little change in surface shape, but did experience a similar increase in oxide thickness as the bonded ribbons, Table I. This was due to the heat treatment imparted during consolidation.

One strip was examined in several areas on the shiny and dull surfaces of the same laminate. It indicated that the dull side may have a thinner oxide layer. It also showed that the increase in silicon concentration was present for most of the ribbon surface; only the black regions having the silicon concentration of the as-cast ribbon.

Careful examination of both faces of a bond allowed the observations of "mirror images" i.e. both sides of the bond. In certain cases, light (silicon-rich) areas on one face correspond to similar light areas on the other face, Fig. 7. In other cases, the light areas correspond to dark areas, Fig. 3, suggesting that a high silicon concentration layer may have peeled off one face, exposing the nominal alloy composition plus some oxidation, and giving, on the other surface, a higher silicon concentration. This peeling of the silicon layer is also suggested by the ragged edges of the light/dark interfaces that were often observed, as in Fig. 5. At low magnification (× 200) it appears that the light to light and light to dark areas are associated with the hills and valleys previously referred to. This is the case in Fig. 3. At high magnifications (× 1000) a series of circles and smooth curves can often be seen, these are typified by Fig. 8 where a light (high silicon concentration) circle is observed with a dark (nominal

TABLE I The depth of the increased surface concentration of silicon and the oxide

Side and location		As-cast ribbon		Single ribbon subject to consolidation process		Consolidated strip	
		Oxide (nm)	Silicon (nm)	Oxide (nm)	Silicon (nm)	Oxide (nm)	Silicon (nm)
Shiny air side							
Normal		70	none	90	60	130	30
Moulded	light hill	no such area	no such area	130	–	150	20
Moulded	dark hill	no such area	no such area	–	none	60	none
Dull wheel side							
Normal		50	none	no such area		no such area	
Valley	light area	no such area	no such area	60	60	50	20
	dark area	no such area	no such area	none	none	65	none
Ridge	light area	no such area	no such area	> 150	–	30	10
	dark area	no such area	no such area	130	80	70	none



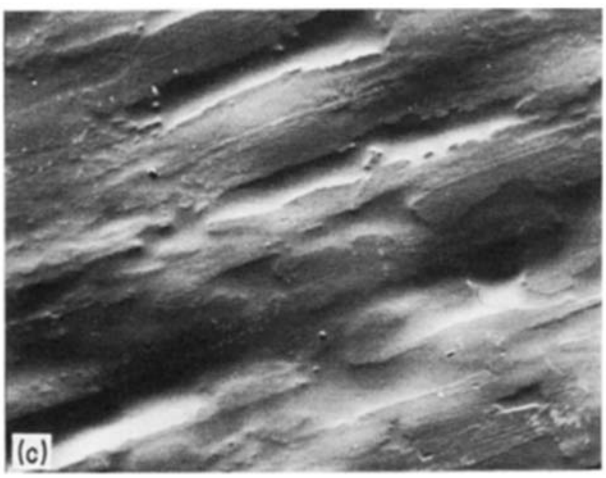


Figure 5 Illustrating the surface nature of the silicon concentration. Micrographs taken of (a) the delaminated strip, (b) when half the oxide layer had been etched away, and (c) when all the oxide layer has been completely etched away.

composition) spot at its centre. The mirror image of this is a dark (nominal composition) ring, as previously found, but at the centre is a dark (nominal composition) spot. These dark spots are either areas of no bonding, or indicate an iron or iron oxide bond. At least some of the iron in the dark spot is oxidized, but the nature of the oxide was not determined. Both types of light and dark areas with light to dark and dark to dark correspondence were commonly found. It was

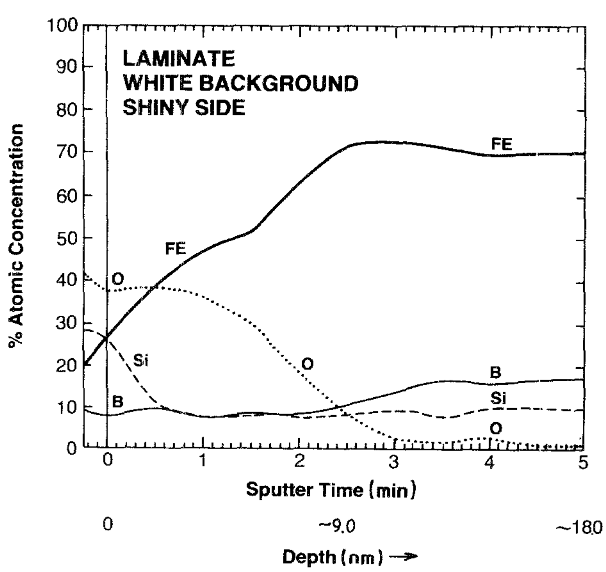


Figure 6 Depth profile for silicon, boron, iron and the depth of oxidation.

not possible, however, to determine that all had mirror images.

This data suggests to the authors two or three bonding mechanisms, which may be separate or complementary. Firstly, the possibility offered by the hill formation is that the ribbons are moulded together and a mechanical interlocking mechanism is occurring as in "Lego" building blocks. Secondly, the silicon enrichment and the correspondence of very high silicon and nominal alloy composition (light or dark) areas leads to a glue mechanism with silicon oxide being the glue, or perhaps, in some cases, a borosilicate or boron oxide. The difference in surface concentration of the silicon and boron was unexpected, especially as, in the literature, both are reported to have similar diffusion coefficients in a Fe–B–Si metallic glass [2]. The thermodynamics actually favours the formation of B_2O_3 over SiO_2 [3]. However, it has been observed in previous work by the authors on as-cast Metglas alloy 2605-S2 that a surface depletion of boron relative to silicon is common. The third mechanism is of an iron or iron oxide bond. There is a sufficient number of dark to dark areas to support an iron rich oxide bond.

3.2.2. Scanning laser acoustic microscopy

An investigation using SLAM was undertaken in order to further determine the nature of the bond. SLAM shows up areas of bonding, through which the acoustic waves can travel, as white areas. This is illustrated in Fig. 9 in which part of a delaminated ribbon may be seen as a dark triangular area: the rest of the micrograph showing a series of white spots and lines which are the bonded area. The bond is not uniform across the surface of the ribbon. This, of course, will be beneficial with regard to magnetic properties, as it will help to reduce eddy current losses.

Further information may be obtained from the micrographs. The valleys of the dull side of an as-cast ribbon, which were described previously in the moulding process resulting in the interlocking mode, may be

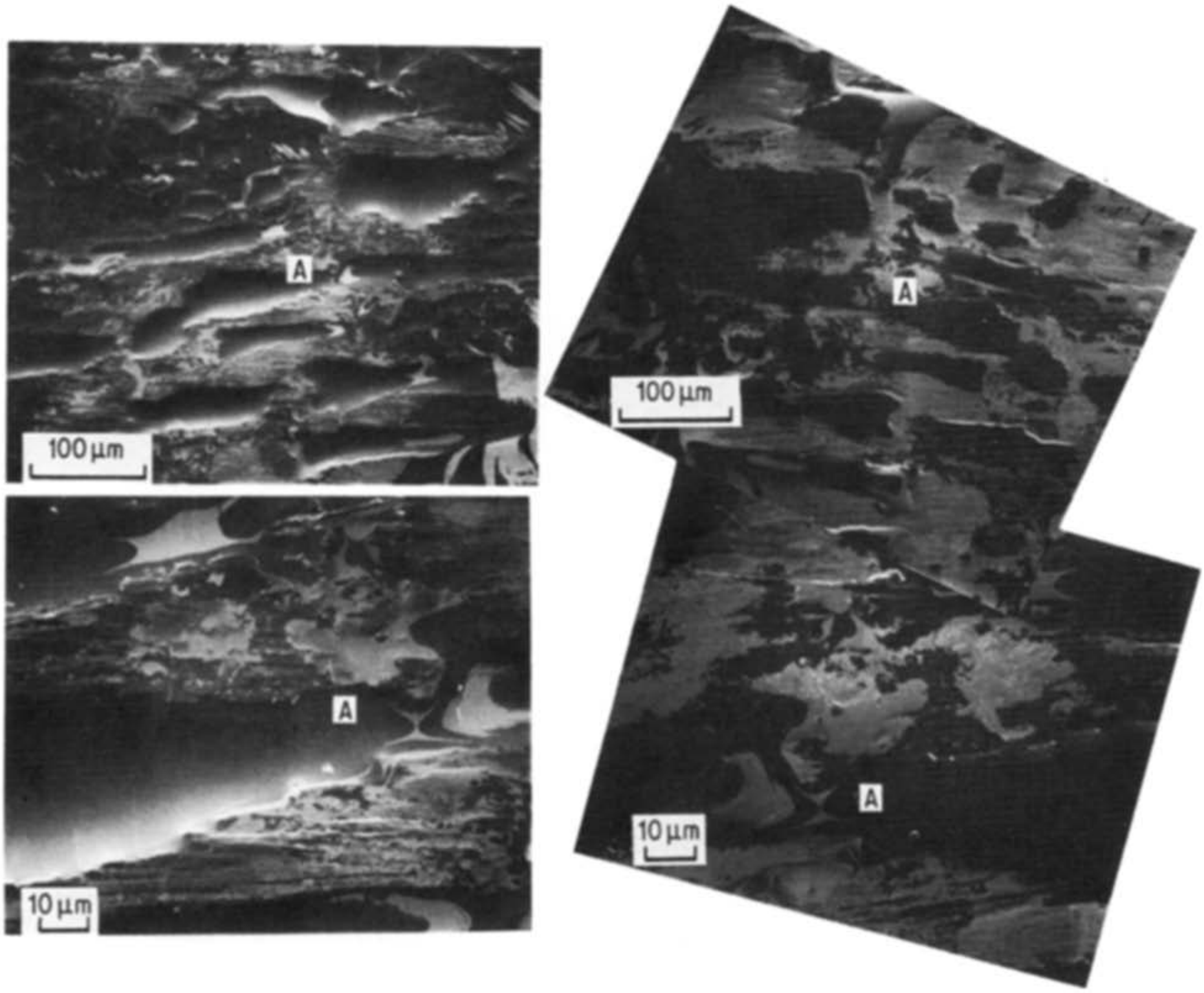


Figure 7 A mirror image of a delaminated strip. One point of correspondence is indicated, by the letter A.

the white lines visible in most of the micrographs. Indeed in Fig. 10, they appear to represent the only bonding mechanism. As the number of layers of ribbon increase, so the micrographs become more difficult to interpret. In addition to the superimposition of the different bonding at each layer, there is also a loss of energy at each interface, this is illustrated in Fig. 11. A typical 5-layer strip of the type commercially available is shown in Fig. 12. It shows the good bonding of the strip, which on a macro level is very uniform, especially when produced under the standard conditions. On a micro scale, even for 5-layer strip, the bond has a non-uniform speckled appearance. To the right of centre, in Fig. 12, can be seen a small region of white lines of the type believed to be associated with moulding. Presumably, there has been a chance alignment of the valleys on the dull side of the ribbons. However, with an acoustic microscope several possible reasons exist for the formation of white lines. They may, therefore, be due to the mode of observation rather than to the specimen. This is not the present case, as observations of a single ribbon delaminated from a bonded strip, Fig. 13 also show up these lines, which, in this case, are definitely due to a mode converted wave from the surface interfering with the bulk wave travelling through the specimen. Mode conversion occurs at sites of surface discontinuities, which may, or may not, be smaller than those normally resolvable by the acoustic microscope. The lines in Fig. 13 are therefore due to the surface texture of the ribbon. From the lines shape, size and uniformity, this texture must be the valley formation on the dull side of the ribbon. For a delaminated ribbon, valleys should be present on one side, and hills on the other. The presence of these lines in micrographs of bonded 2-, 3- and 5-layer strip with the same brightness as areas known to be bonded, indicates that the white lines are areas of bonding thus supporting the interlocking model.

It has also been possible to distinguish broad bands or waves of bonding, Fig. 14. These are believed to be associated with the wavy structure observed on the shiny side of as-cast ribbon. It may be assumed that the high points of these "waves" are the areas of maximum pressure during consolidation and are therefore the first places to bond. Careful observations allow these preferential areas of bonding to be found on most of the micrographs: Fig. 9, for instance, has two such vertical lines, or waves. Originating from these geometric factors are larger areas of bonding which show up as white spots, Fig. 15. These match in size the white to dark areas observed by SAM better than the dark to dark areas. This would suggest that high silicon (light) areas are the regions of bonding.

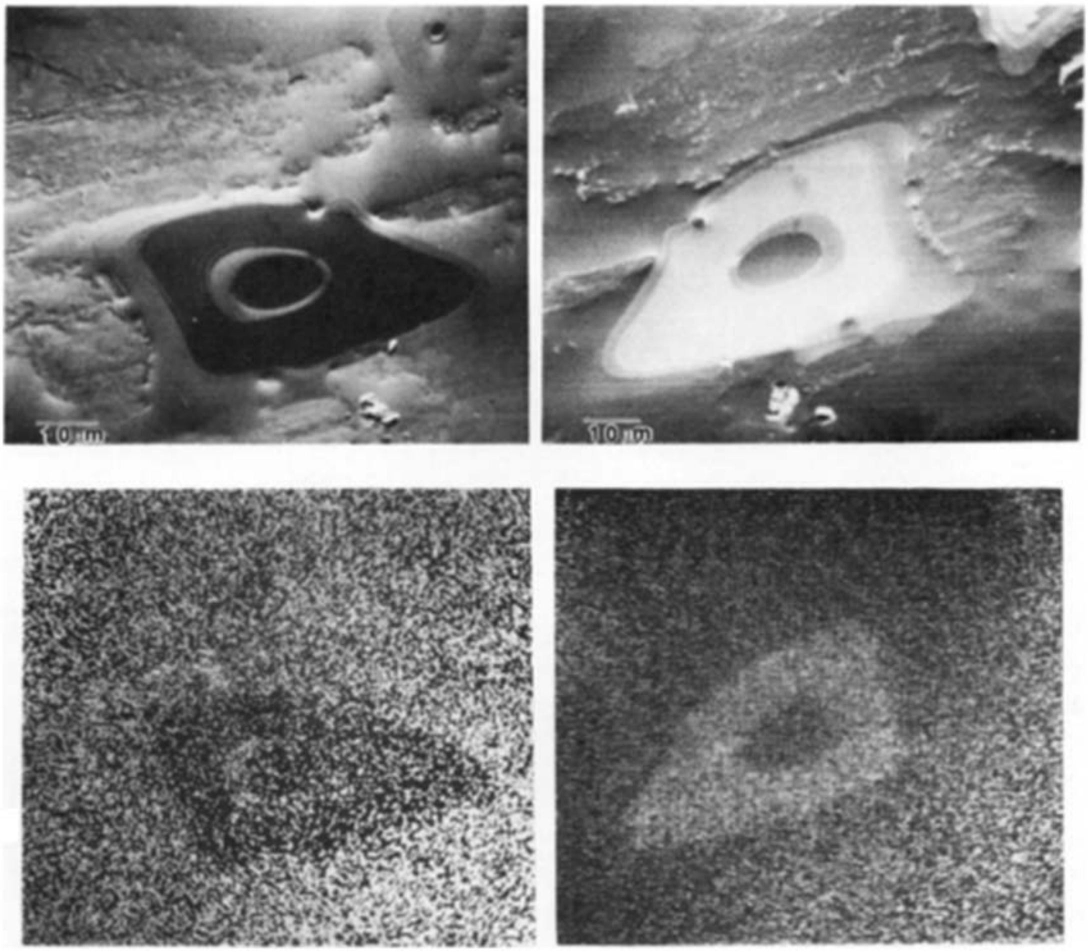


Figure 8 A mirror image of two faces of a delaminated strip showing light to dark correspondence associated with high silicon concentration. At the centre is a dark to dark correspondence that may represent an iron rich bond.

4. Discussion

Both SLAM and SAM show that the bond is localized in discontinuous 10 to 200 μm regions. From the SLAM investigation in particular, it appears that bonding first occurs at high points on the ribbon which are the points of maximum pressure, and spreads out from these areas by plastic deformation that moulds material into the valleys on the dull side of the ribbon. That this produces an interlocking bond, is confirmed by the SLAM micrographs where white, bond lines continue into unbonded areas, but not vice versa. In addition, SAM shows that a silicate glue type bond may also be occurring and the SLAM micrographs do confirm bonded areas of the same size as the areas of SiO_2 enriched areas seen by SAM.

Plastic deformation of the irregularities in the ribbons surface is an essential part of the proposed bonding process. That such deformation occurs is confirmed by the improved packing factor of the strip compared to the ribbon. The strip has a pack factor consistently over 90%, compared to values of 70 to 85% for the ribbon used.

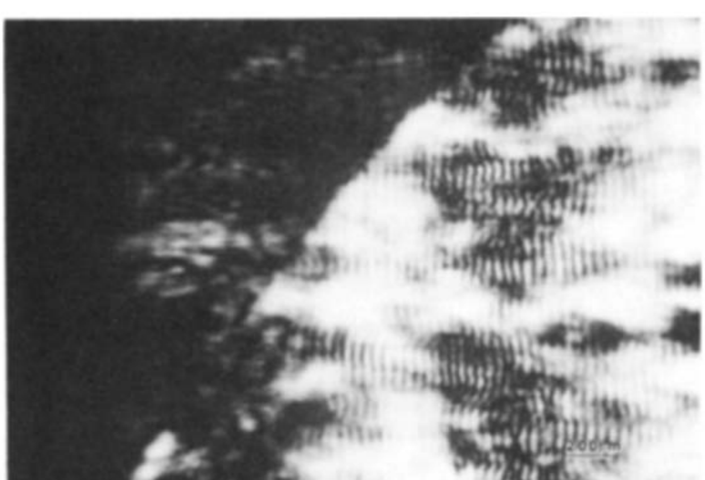


Figure 9 A scanning laser acoustic micrograph. The top delaminated layer shows up as a dark region on the left hand side. Bonded areas are apparent as white regions.

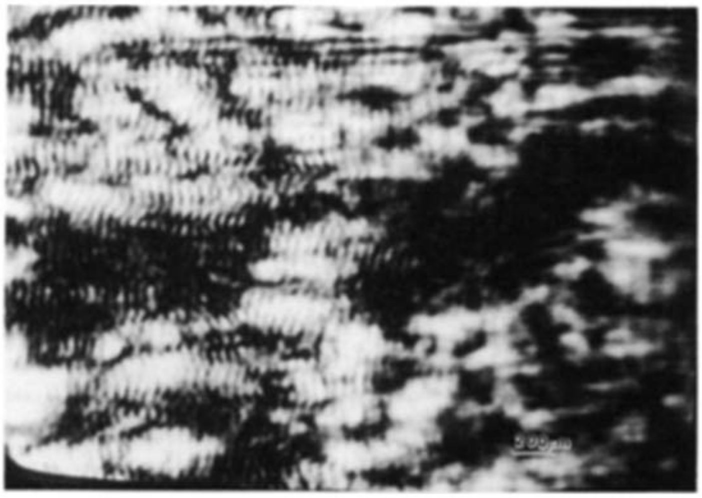


Figure 10 Micrograph of a strip debonded to leave two layers. The white lines of bonding appear to correspond to the valleys on the dull side of the ribbon. Confirming that moulding is occurring.

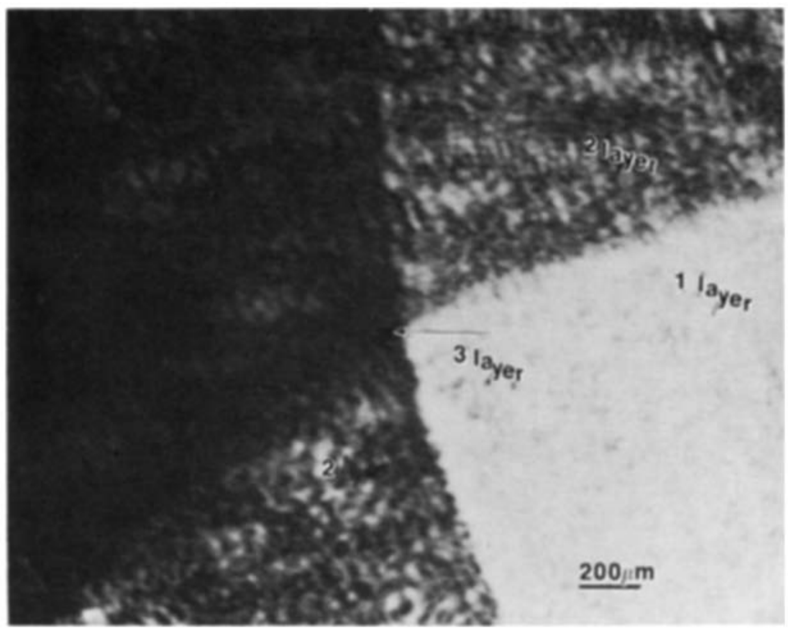


Figure 11 Illustrating the energy lost by the acoustic wave at each interface. The strip has been debonded to leave 1-, 2- and 3-layer thick strip.

Figure 12 A representative micrograph of a 5-layer thick strip of the type commercially available.

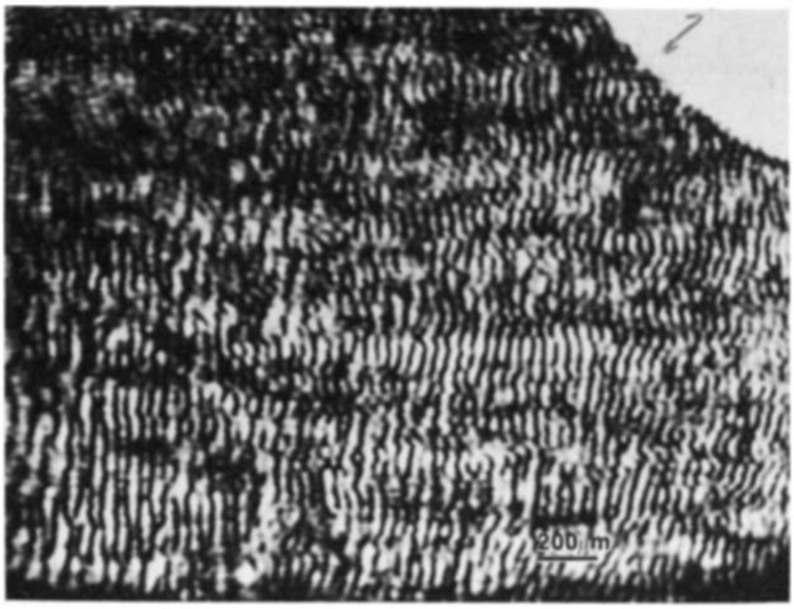


Figure 13 An acoustic micrograph of a single ribbon delaminated from a bonded strip.

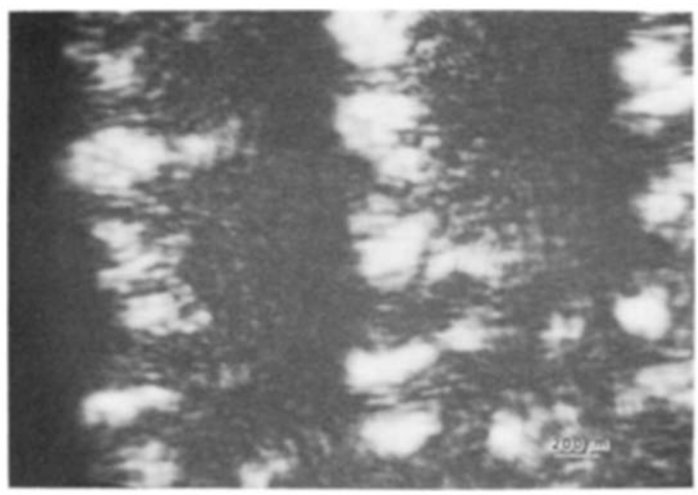


Figure 14 Micrograph of a poorly bonded strip delaminated to two layers. The broad bands of bonding correspond to the higher crests of the waves on the shiny side of the as-cast ribbon.

Figure 15 Micrograph of a well bonded strip delaminated to two layers. The large white bonded areas indicate by their size that bonding is associated with areas of high silicon concentration.

5. Conclusions

By the use of two complementary techniques, information has been obtained which allows a consistent model to be proposed for the bonding process during the consolidation of an amorphous ribbon. The localized nature of the bond, plus the insulating nature of the bonding oxide can be readily understood to give low eddy current losses. The bond apparently originates at high points on the ribbon's surface and spreads as the plastic deformation of the ribbon increases. This is consistent with the improved pack factor of the strip compared to the ribbon.

Further investigations are underway on this useful strip, with the intention of determining further its properties and the nature of the bond, while at the same time, bringing it into large scale production.

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References

1. L. E. DAVIS *et al.*, "Handbook of Auger Electron Spectroscopy", 2nd edition, (Physical Electronics Division, Eden Prairie, USA, 1978) p. 53.
2. F. E. LUBORSKY, *J. Appl. Phys.* **54** (10) (1983) 5732.
3. "CRC Handbook Chemistry and Physics" edited by R. Weast, 55th edition (CRC Press Boca Raton, Florida, 1974).

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