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CRANFIELD

THE EFFECT OF NITROGEN IN α-IRON ON FERROMAGNETIC DOMAIN BOUNDARY MOVEMENT EXAMINED BY A MECHANICAL DAMPING METHOD

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CORRIGENDUM

Page 6, line 6 should read: higher than that obtained by most other workers (4)(2), but was felt to

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Summary

The effect of ageing below 200°C on the hysteresis loss in α-iron containing nitrogen has been investigated. The extent to which nitrogen is removed from solid solution during ageing, and the domain boundary hindrance developed, was assessed by mechanical damping, using a low-frequency inverted torsional pendulum. Nitrogen, both in solution and as the precipitated form Fe_8N, was found to increase the resistance to domain boundary movement, the precipitated form being the more effective in this respect. Most of the damping changes due to ageing are developed in less than 2 hours at 150°C; a further investigation to cover this region would need to be undertaken to establish quantitative relationships.

Contents

1. Introduction

The 'core loss' of power transformers can increase by amounts of up to 10% during service at operating temperatures, which are usually about 60°C and sometimes as high as 80°C or 90°C. Any additional 'core loss' is principally due to higher hysteresis losses, brought about by the increased resistance to domain boundary movements offered by the small amounts of impurity elements precipitated from solid solution at these operating temperatures. The principal impurity elements in high grade silicon irons used as core materials are carbon and nitrogen. While carbon is strictly controlled to a specified maximum of 0.005%, nitrogen receives less attention, and is the most probable cause of the increased losses on ageing.

Ferromagnetic domain boundaries absorb energy in moving through a structure containing non-magnetic inclusions or stress gradients (1), and this effect can be conveniently studied by a damping method, since any applied stress will cause domain boundary movement through magnetostriction. For a few simple and idealized cases, it is possible to calculate the magnitude of the resistance to domain boundary movement due to a specific inclusion or stress field(1). However, the case of a precipitated phase, involving the variables of shape and size of precipitate, and its associated stress field, is too complex to assess theoretically. This investigation will therefore be confined to the increased domain boundary hindrance which results from precipitation of iron nitride (in the form Fe_8N) in α-iron. Since Fe_8N is the precipitate formed from supersaturated solutions of nitrogen both in pure iron and in an iron - 3% silicon alloy, below 220°C(2)(3), the behaviour of pure iron should be similar in this respect to that of a commercial iron - 3% silicon alloy.

As domain boundary movement is opposed by the precipitation of nitride, the magnetic damping resulting from a specified alternating applied stress is reduced. The extent to which precipitation has progressed can also be followed by a damping measurement, since the Snoek peak height due to the presence of nitrogen (the peak occurs at about 1 cycle per second at room temperature) is approximately proportional to the amount of nitrogen in solid solution(4).

2. Experimental method

2.1 Material

The basic material used was a low carbon iron in the form of 1/16" diameter wire. The micrographs (Figures 9 and 12) show a low carbon content, there being no carbide evident either as pearlite, or as a grain boundary constituent. The micro-structure of material 'A' (Figure 9) did, however, show a considerable amount of non-metallic inclusion. After tests on the first two specimens, a change was made to material 'B' (Figure 12) which was of similar carbon content but contained less inclusions.

2.2 Heat treatment

All heat treatment was carried out in a small tube furnace similar to the type T.3 experimental furnace designed by Morganite Electroheat Limited. Some small modifications were necessary to obtain a reasonably uniform hot zone 9″ in length, with a temperature control within ±5°C at 900°C, achieved by means of a Foster proportional controller operating through a 50 B Variac.

A controlled atmosphere within the furnace tube was maintained throughout all heat treatments, oxygen-free hydrogen being used for the annealing cycles, and an argon stream, bubbled through 0.880 ammonia, for nitriding. One end cap of the furnace could be easily removed to allow rapid extraction of the specimen for quenching operations.

All specimens were annealed at 900°C in hydrogen and furnace-cooled to 600°C, prior to either the application of a particular cooling rate to obtain an un-nitrided specimen as a standard, or the nitriding process.

If nitriding was required, the argon-plus-ammonia mixture was passed over the specimen at 600°C for a controlled time before the temperature was raised to 900°C for 4 hours, to allow homogenization by diffusion. Furnace cooling to 600°C then took place, and finally a selected cooling rate from 600°C was applied to retain nitrogen in solid solution.

3. Damping measurements

Two torsion pendulums were used for the damping measurements; these may be described as follows.

3.1 A normal Kê type pendulum

This pendulum was fitted with a furnace, enabling temperature scans to be made over the Snoek peaks for carbon and nitrogen. The temperature over an 8-inch specimen in this assembly could be kept uniform to ±1°C and measured to ±0.25°C at its mid-point by a chromel-alumel thermocouple and vernier potentiometer.

3.2 An inverted pendulum

This instrument, which is illustrated in Figures 1, 2 and 3, was used to follow the nitride precipitation process, and to measure the magnitude of the magnetic damping. This latter quantity is given by the difference between the total damping with and without a saturating field. This field is provided by the coil shown in

Figure 3. The specimen in this pendulum was 5 inches in length, and its temperature was controlled by operating the pendulum in a perspex enclosure containing a heating source of 260 watts. Temperature variation over the specimen length was ±0.5°C and measurement was by means of a chromel-alumel thermocouple and potentiometer with an accuracy of ±0.25°C. Longitudinal stress in the specimen could be reduced to a very low value by counterbalancing the mass of the inertia arm and weights through a phosphor-bronze suspension wire of negligible damping capacity.

To avoid any damping due to the stray magnetic fields produced by the coil, the inertia bar and weights were of non-magnetic material, and a test run using a non-magnetic specimen gave no damping difference between the field-off and field-on conditions.

The damping results from both pendulums were obtained from records made by the continuous tracking of a light spot reflected from a galvanometer mirror on the inertia arm. A graphispot recorder was used as the following device, a typical record being reproduced in Figure 4.

4. Results

4.1 The logarithmic decrements (δ) quoted in the table below were all measured at a maximum surface shear strain amplitude of 1×10^{-4}.

Specimen	Treatment	Testing Frequency cy/sec.	Testing temperature °C	Unmagnetised δ	Saturated δ	Magnetic δ
A 1a	Furnace cooled, un-nitrided	0.7	19.5	0.0174	0.0136	0.0138
A 2a	Water quenched, nitrided for 15 mins.	0.7	19.5	0.0623	0.0597	0.0026
A 2b	Aged for 600 hrs. at 120°C	0.7	19.5	0.0068	0.0068	
A 2c	Aged for 400 hrs. at 120°C	0.7	19.5	0.0064	0.0064	
B 1a	Furnace cooled, un-nitrided	0.7	19.5	0.0171	0.0124	
B 2a	Air cooled, nitrided for 5 minutes.	0.7	19.5	0.0575	0.0188	0.0387
B 2b	Aged for 70 hrs. at 95°C	0.7	19.5	0.0146	0.0128	0.0018

Specimen	Treatment	Testing frequency cy/sec.	Testing temperature °C.	Unmagnetised δ	Saturated δ	Magnetic δ
B 6a	Water quenched, un-nitrided	1.1	23.5	0.0384	0.0028	0.0356
B 6b	Aged 2 hrs. at 150°C.	1.1	23.5	0.0242	0.0024	0.0214
B 6c	Aged 5 hrs. at 150°C.	1.1	23.5	0.0246	0.0024	0.0222
B 6d	Aged 10 hrs. at 150°C.	1.1	23.5	0.0246	0.0049	0.0197
B 6e	Aged 20 hrs. at 150°C.	1.1	23.5	0.0208	0.0026	0.0182
B 6f	Aged 50 hrs. at 150°C.	1.1	23.5	0.0201	0.0028	0.0173
B 6g	Aged 100 hrs. at 150°C.	1.1	23.5	0.0199	0.0030	0.0169
B 7a	Water quenched, nitrided 5 mins.	0.7	23.5	0.0384	0.0334	0.0050
B 7b	Aged 2 hrs. at 150°C.	0.7	23.5	0.0124	0.0119	0.0005
B 7c	Aged 5 hrs. at 150°C.	0.7	23.5	0.0097	0.0092	0.0005
B 7d	Aged 10 hrs. at 150°C.	0.7	23.5	0.0102	0.0100	0.0002
B 7e	Aged 20 hrs. at 150°C.	0.7	23.5	0.0091	0.0088	0.0003
B 7f	Aged 50 hrs. at 150°C.	0.7	23.5	0.0087	0.0083	0.0004
B 7g	Aged 100 hrs. at 150°C.	0.7	23.5	0.0091	0.0091	0.0000

4.2 The results of the temperature scan over the Snoek peaks associated with nitrogen and carbon, for the three specimens B3, B4 and B5, are shown in Figure 5. Specimen B3 was un-nitrided and quenched in water from 600°C, while specimens B4 and B5 were nitrided for 5 minutes and homogenized as previously described, before being quenched in water from 600°C. The strain amplitude applied to these specimens was 10^{-5}, the magnetic damping contribution being negligible in these specimens at this strain amplitude.

4.3 The variation of damping with strain amplitude for specimen A1 in the absence of a magnetic field is shown in Figure 6.

5. Discussion

The magnetic component of damping for specimen A1 is shown, in Figure 6, to be proportional to amplitude over the range examined, being completely suppressed by an axial field of 300 Oe. This result is in agreement with those of previous workers.(5)(6) To enable comparative measurements of magnetic damping to be made between specimens, it is necessary to quote damping values at a specific strain amplitude; an arbitrary strain of 10^{-4} was selected for this purpose.

The results from specimen A2a show a very high as-quenched damping, as a consequence of nitriding, most of which is not magnetic in origin. This damping diminishes very rapidly on ageing, and is accompanied by precipitation, as shown in the photomicrographs Figures 10 and 11. The ageing process also completely eliminates the small amount of magnetic damping present in the as-quenched condition. The massive nitride present in the microstructure, Figures 10 and 11, shows that a 15 minute nitriding treatment is excessive, since nitrogen contents are required up to the solid solubility limit. As a result of the tests on material 'A' a change to material 'B' was made. A comparison between specimens B1a and A1a, each of which had the same heat treatment, shows the two materials to be almost identical. 'B' has a lower non-metallic inclusion content, but exhibits a slightly higher damping due to carbon and nitrogen.

Specimen B2 was nitrided for 5 minutes only, to avoid the appearance of massive nitride, and was then cooled in still air, as an alternative to a water quench, in order to obtain lower internal stresses. The effect of internal stress on domains and their movement is considerable, both the domain size and the arrangement being modified. The magnetostrictive energy of the domains minimises the internal stresses, and the external applied stresses will change the domain configurations only when they exceed the internal stresses. As a result of the lower cooling rate of specimen B2a, a higher proportion of the as-cooled damping was of magnetic origin, but a much smaller amount of nitrogen was retained in solution, giving a lower damping in the magnetically saturated condition. However, ageing at a lower temperature for a shorter time than A2 still developed sufficient domain boundary hindrance to suppress most of the magnetic damping.

The results from specimens B3, B4 and B5, shown in Figure 5, were obtained to assess the efficiency of the nitriding process by establishing the nitrogen peak position, in respect of temperature and frequency, and also the peak height resulting from a specific nitriding time. The results show that in the case of specimen B3, the basic material used contains some interstitial carbon, giving a peak at about 45°C and 1.8 cycles per second. This is in agreement with other published work(4)(7)(8).

Specimens B4 and B5 show a higher maximum Snoek damping, and a shift towards a lower temperature due to an increase in interstitial nitrogen. The nitrogen peak obtained, on subtracting the results for B3 from those for B4 and B5, shows a maximum at about 30°C for a frequency of 1.8 cycles per second. This result is 1° to 1.5°C higher than that obtained by most other works(4)(2), but was felt to be sufficiently close to justify fixing the nitrogen peak at 23.5° for a frequency of 1 cycle/second. It was decided to adopt this temperature and frequency for all subsequent damping measurements. Magnetic damping is almost independent of temperature and frequency over the testing range(5), and by operating on the nitrogen peak frequency and temperature, the precipitation of nitrogen can be accurately followed since nitrogen peak damping, as a function of nitrogen in solid solutions, has been established by Köster and other workers. Nitrogen peak damping as a function of nitrogen in solution, according to Köster(4), is shown in Figure 7.

The results obtained from specimens B6 and B7 are plotted in Figure 8, and clearly show that in both the nitrided and the un-nitrided specimens, the major part of the changes in Snoek damping and magnetic damping are complete in 2 hours at 150°C. In the case of the nitrided specimen this is not in agreement with Fast and Verrijp(2) who obtained results indicating that for specimens containing 0.0267% nitrogen, or less, in solution, ageing changes are not complete until at least 10 hours at 150°C (specimen B7 as-quenched contained 0.012% nitrogen). However, the earlier work of Wert(10) shows the removal of nitrogen from solid solution to be virtually complete in under 100 minutes at 150°C. Unfortunately, due to lack of data within the first 2 hours of ageing, it is not possible to give any quantitative estimate of the precipitation rate over this period.

The results show that the precipitated constituent Fe_8N is able to offer resistance to domain boundary movement, and will therefore increase the power loss on the reversal of magnetisation. Nitrogen in solution, if coupled with a high cooling rate, also resists boundary movement, and a lower cooling rate, (e.g. that applied to the air-cooled specimen B2a), gives greater boundary freedom. An optimum cooling rate to retain nitrogen in solution, consistent with a low internal stress, will therefore give maximum domain boundary freedom, or maximum magnetic damping at a pre-set strain amplitude.

Specimen B6a, the un-nitrided and quenched specimen, suffers an extremely rapid initial Snoek damping change due to ageing, but continues to show a slow fall up to 100 hours due to carbide precipitation. In addition, the as-quenched magnetic damping is higher than that of furnace-cooled specimens A1a and B1a, lending support to the conclusion that a critical cooling rate exists for minimum interference with domain boundary movement.

It is unfortunate that an examination of the microstructures reveals a considerable grain size difference between the nitrided and un-nitrided specimens. This complication has arisen due to the homogenisation treatment at 900°C after nitriding. On cooling in the furnace prior to quenching from 600°C, the temperature of maximum interstitial solubility, the nucleation rate is increased due to the nitrogen present, giving a smaller ferrite grain size, with a higher background damping, and a lower magnetic damping due to the grain boundaries interfering with domain boundary movement.

6. Conlusions

The damping measurements confirm that there is an effect on domain boundary movement due to the precipitation of iron nitride from a supersaturated solid solution of nitrogen in α-iron. The following points are of particular interest:

(1) Nitrogen in solution, coupled with a high cooling rate (e.g. by water quenching), suppresses domain boundary movement. Precipitation of Fe_8N also resists boundary movement, the precipitated nitride offering a greater resistance than the same amount of nitrogen in solid solution.

(2) The removal of excess nitrogen from supersaturated solution is complete, or nearly so, in 2 - 5 hours at 150°C.

(3) The nitriding process used is satisfactory in that 5 minutes treatment is sufficient to give nitrogen contents approaching the solid solubility limit at 600°C. The refinement of grain size due to subsequent homogenizing may possibly be avoided by allowing a longer time at a lower temperature, so avoiding any phase change.

(4) The carbon content of the basic material used was sufficiently high to cause interference with the results by carbide precipitation, and it would therefore be preferable to use a base material of lower carbon content.

The conclusions above indicate the need for a more comprehensive study, particular attention being paid to the earlier stages of process, and to the removal of complicating effects arising from the presence of carbon in the basic material, and grain size differences.

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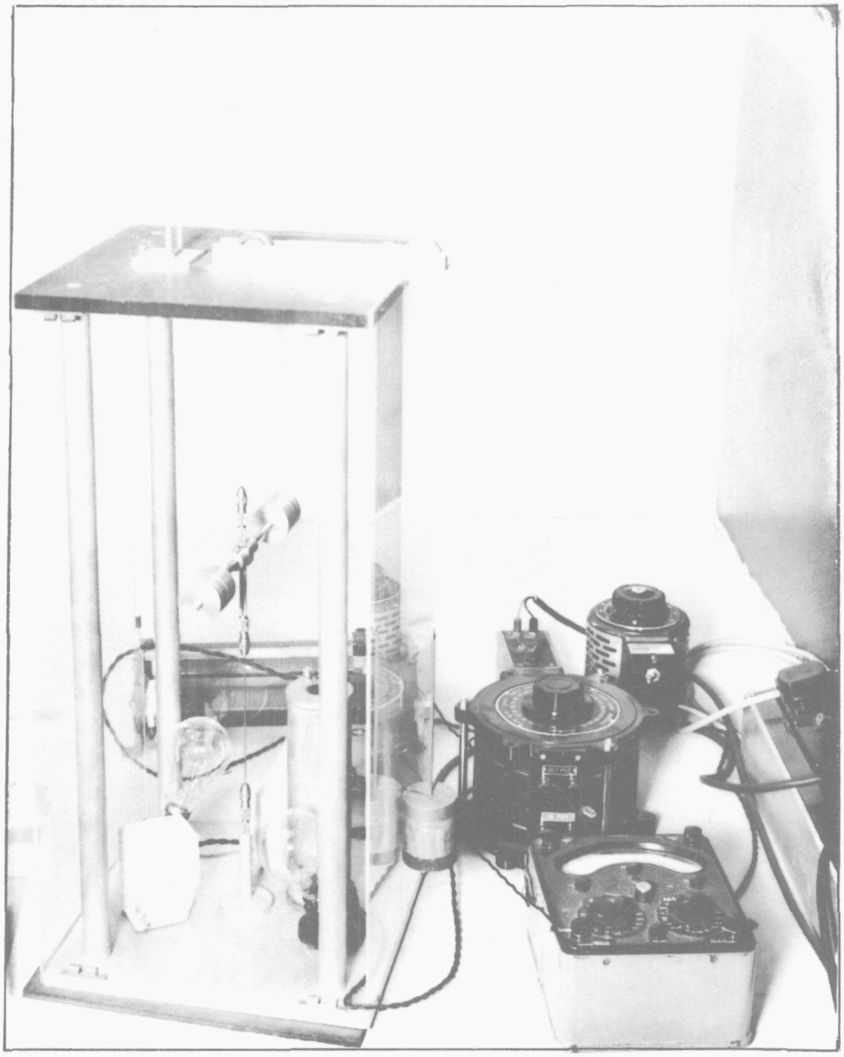

FIG. 1. GENERAL VIEW OF THE INVERTED PENDULUM

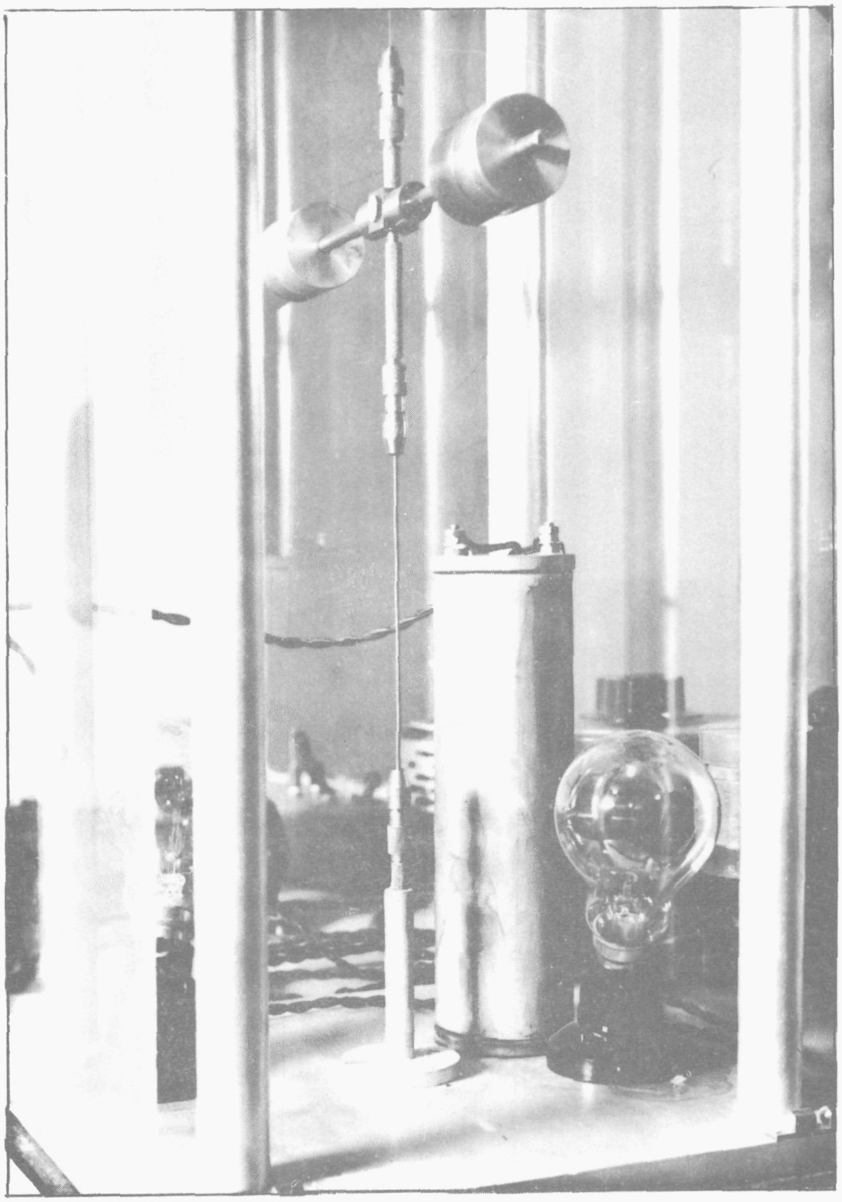

FIG. 2. DETAIL OF SPECIMEN, GRIPS, AND INERTIA ARM

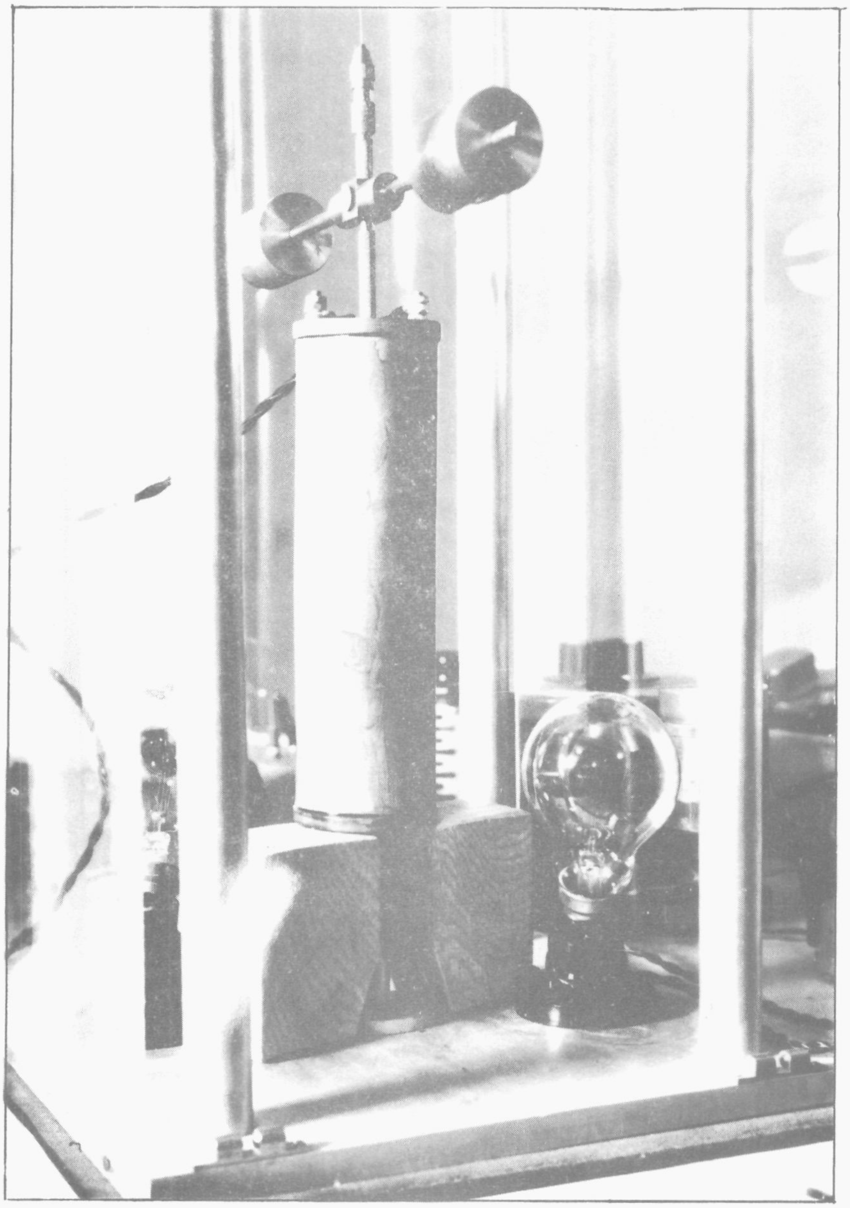

FIG. 3. MAGNETISING COIL IN POSITION

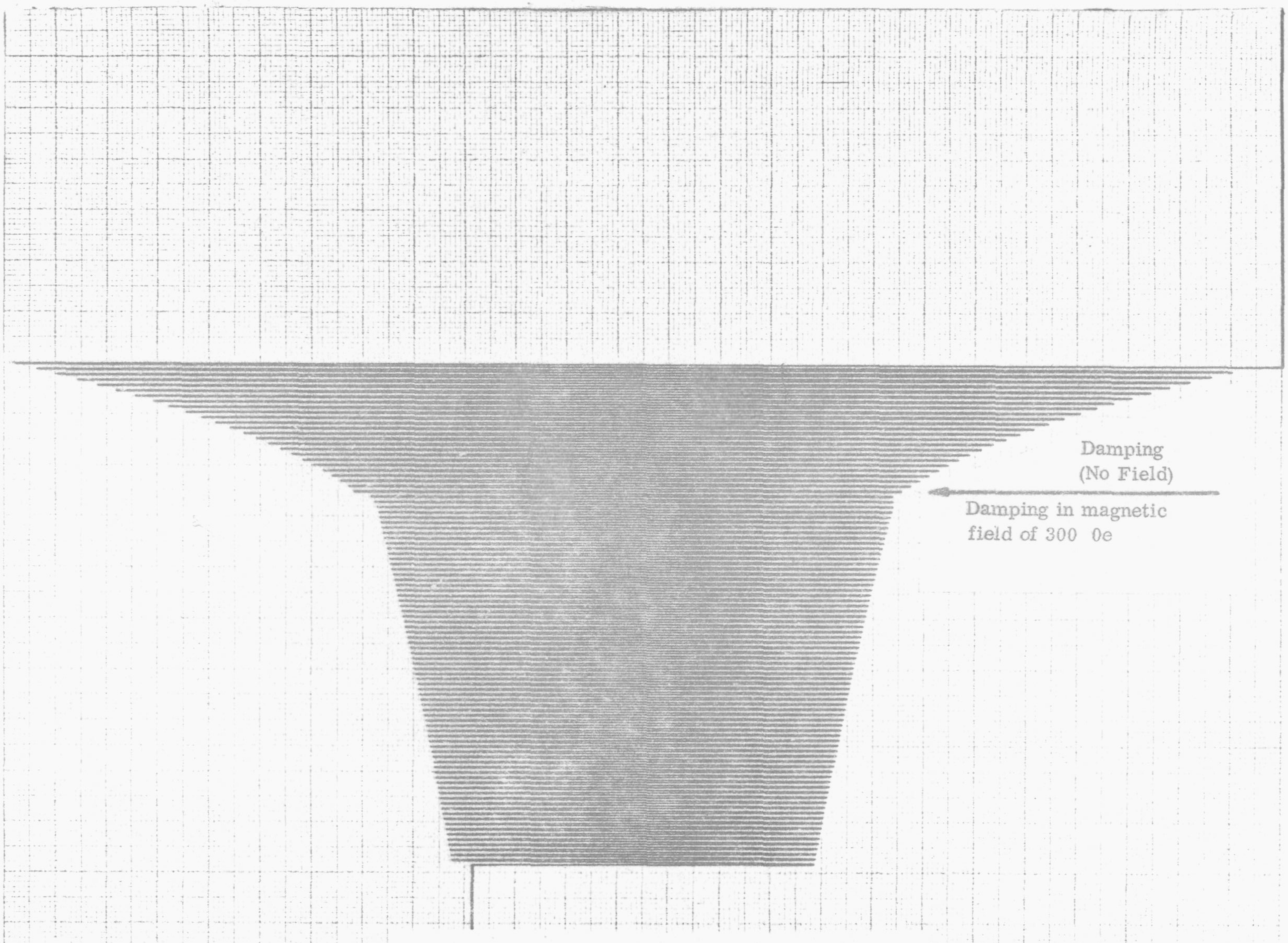

FIG. 4. TYPICAL GRAPHISPOT RECORD

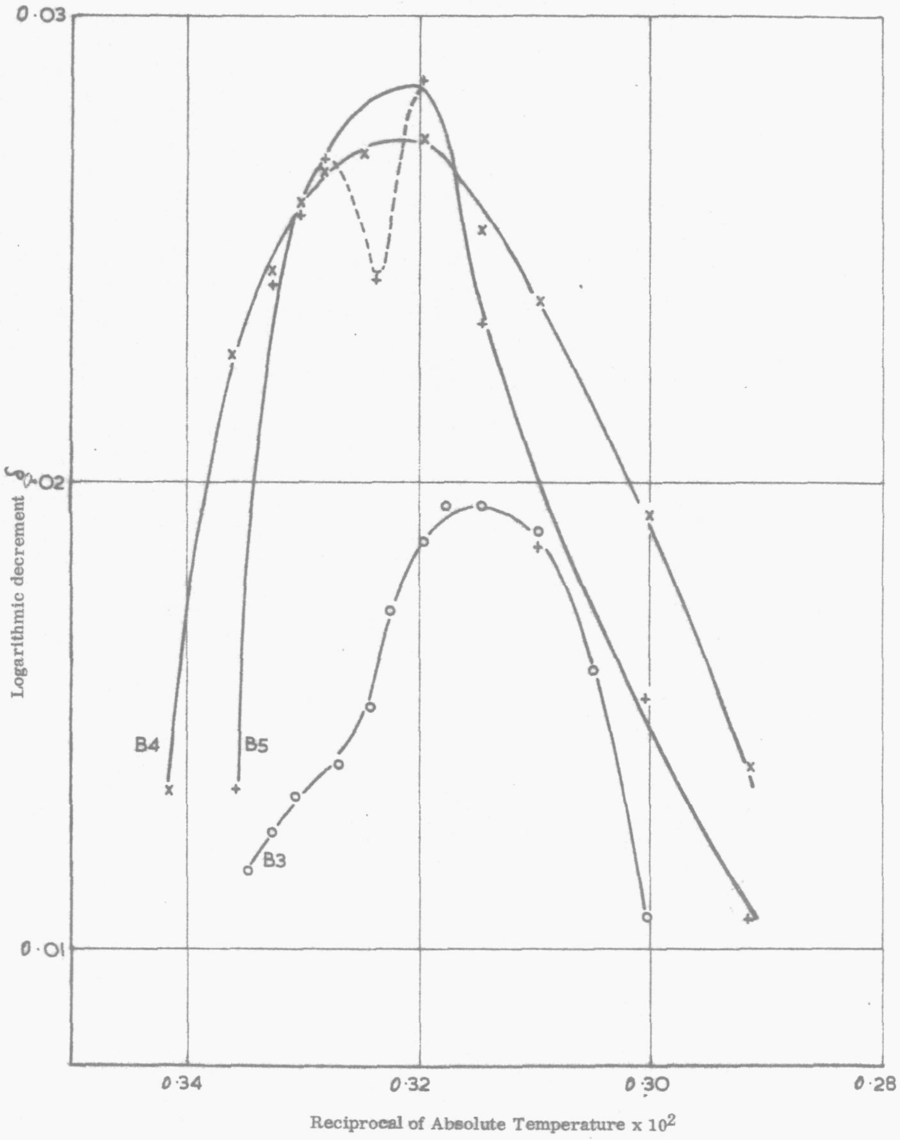

FIG. 5. DAMPING v.s. TEMPERATURE PLATE FOR SPECIMENS B.3, B.4 AND B.5

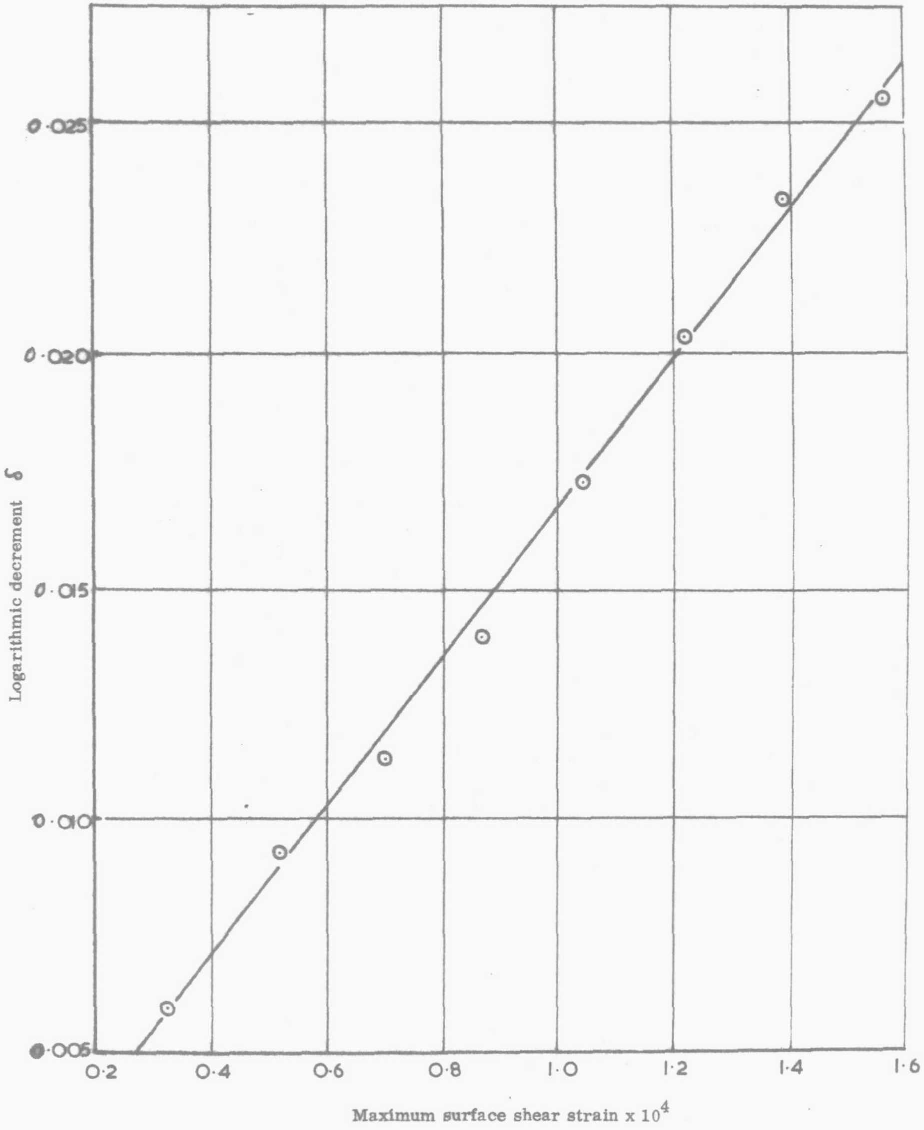

FIG. 6. DAMPING v.s. STRAIN AMPLITUDE FOR SPECIMEN A.1 (UNMAGNETISED)

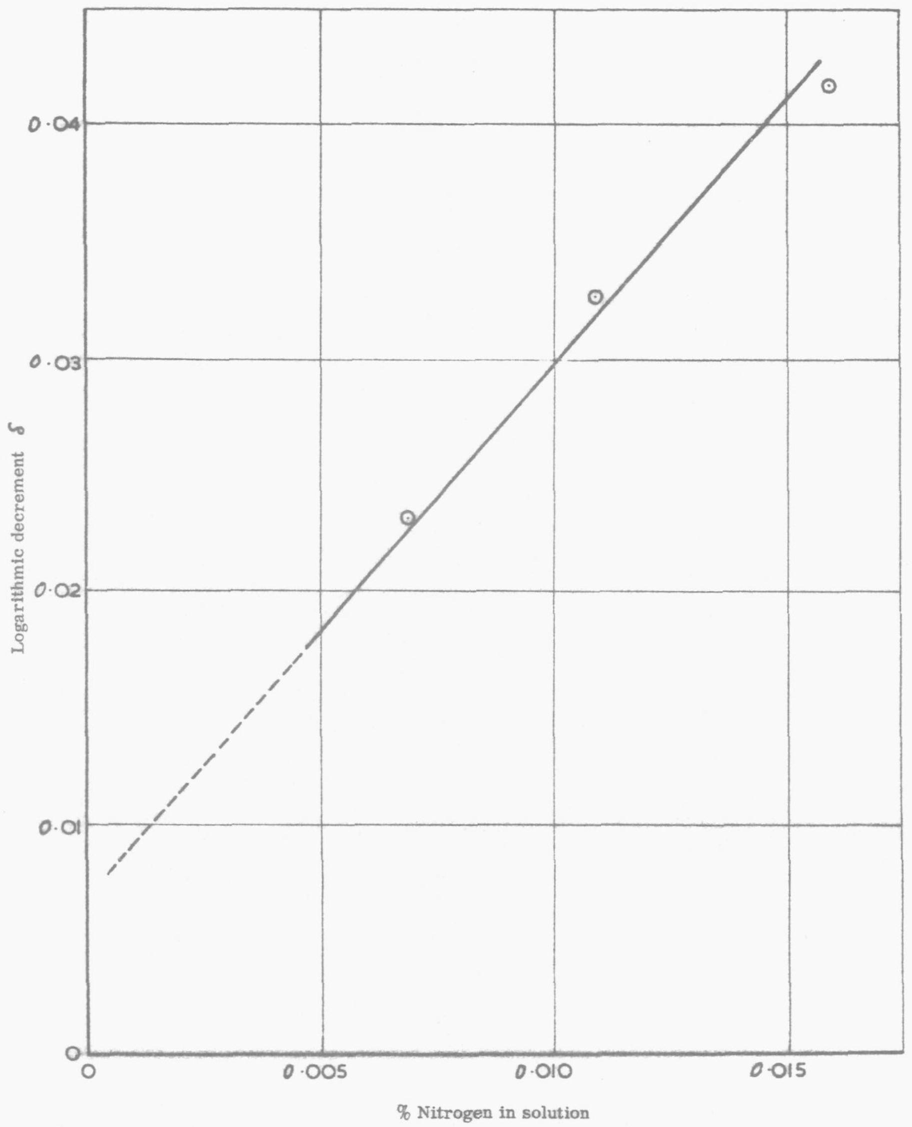

FIG. 7. NITROGEN PEAK DAMPING v.s. NITROGEN IN SOLUTION IN α-IRON (KÖSTER 1961)

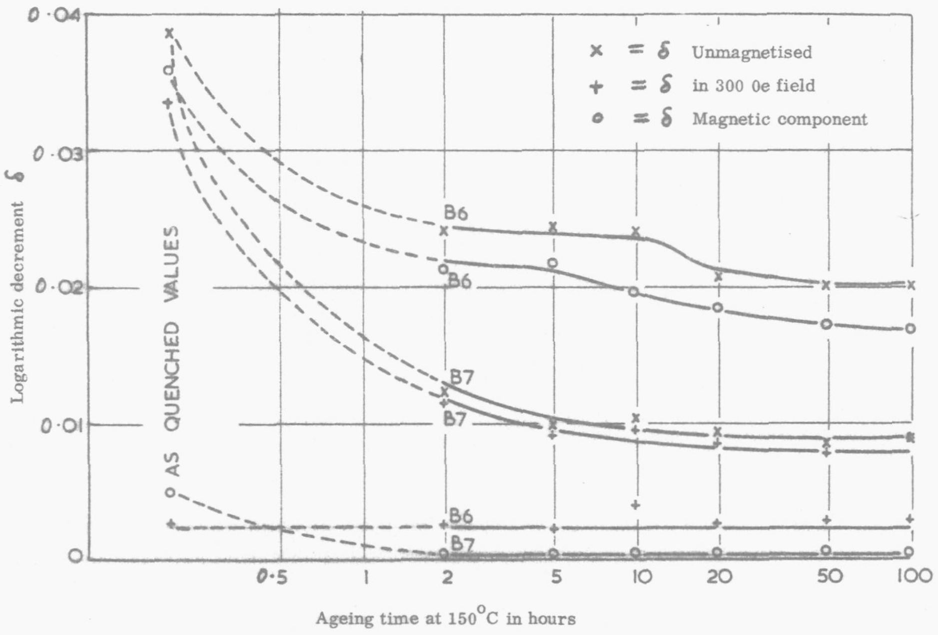

FIG. 8. DAMPING v.s. AGEING TIME FOR SPECIMENS B.6 AND B.7

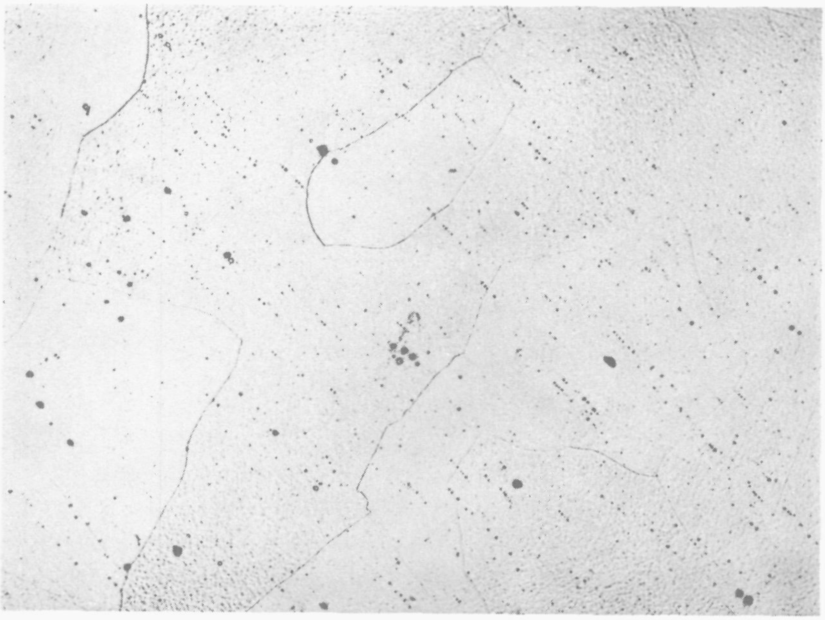

FIG. 9. SPECIMEN A.1(a), MAG. x 200

FIG. 10. SPECIMEN A.2(c), MAG. x 200

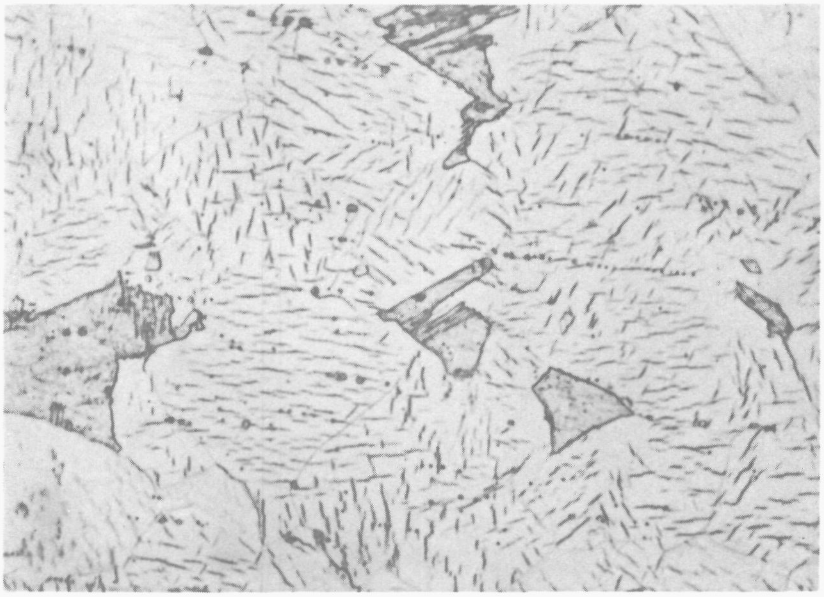

FIG. 11. SPECIMEN A. 2(c), MAG. x 1000

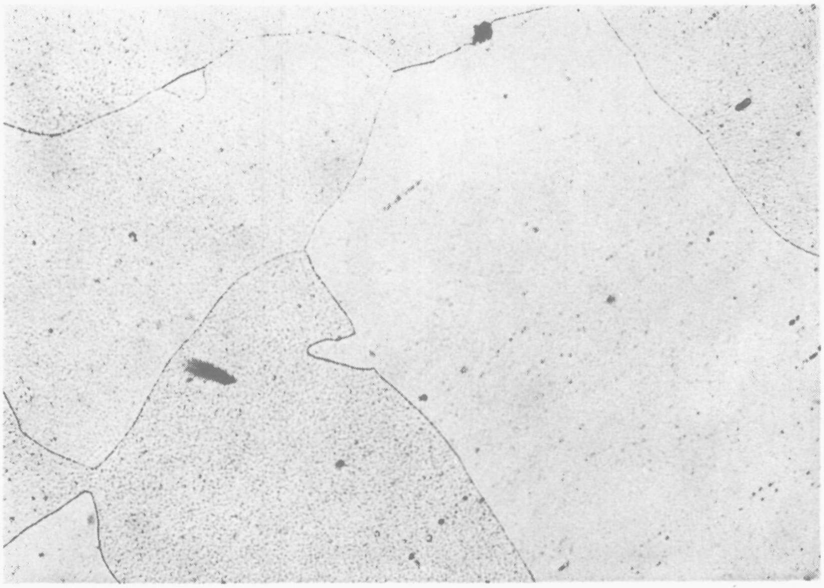

FIG. 12. SPECIMEN B. 6(a), MAG. x 200

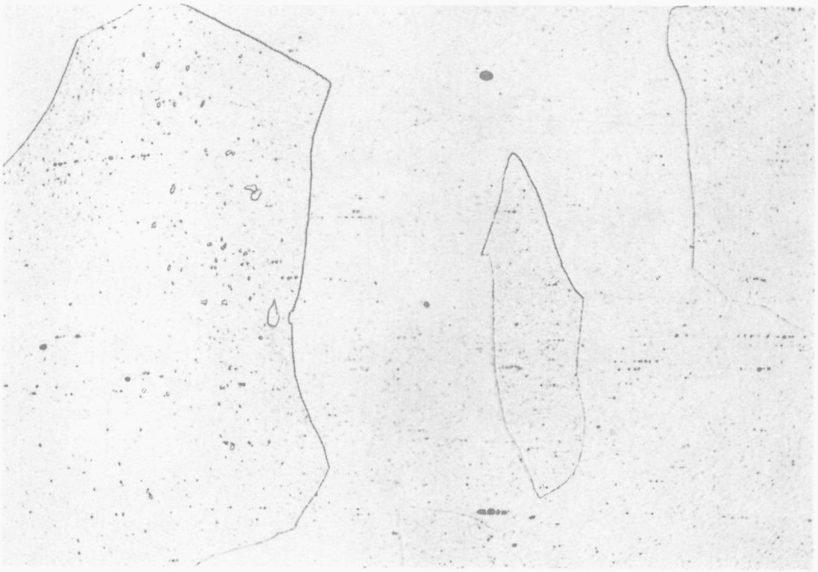

FIG. 13. SPECIMEN B.6(g), MAG. x 200

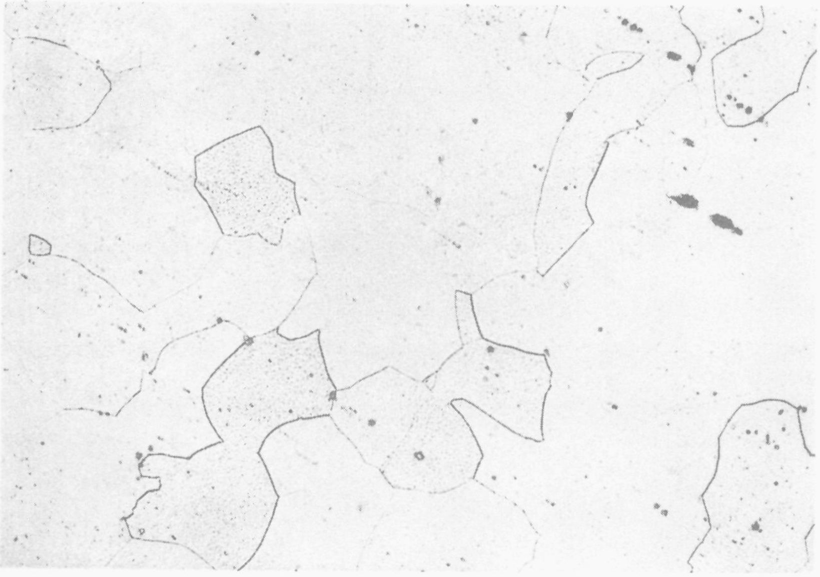

FIG. 14. SPECIMEN B.7(a), MAG. x 200

FIG. 15. SPECIMEN B. 7(g), MAG. x 200

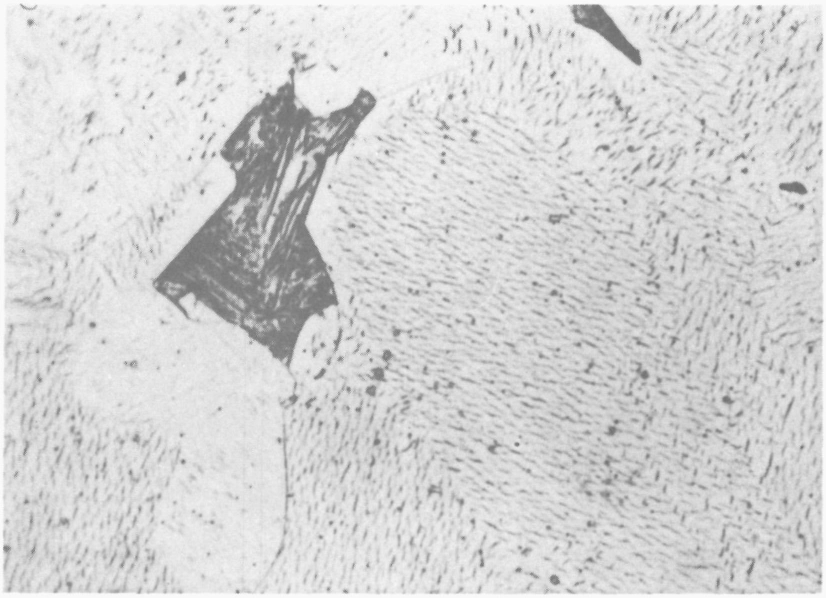

FIG. 16. SPECIMEN B. 7(g), MAG. x 1000