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Structure and properties of melt-spun iron–silicon alloy filaments having single crystalline structure

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Melt-spun iron–silicon (Fe–Si) alloy filaments were obtained by a rapid solidification method using an in-rotating-water spinning apparatus. The silicon content in the alloy composition was selected to be about 6.5% by weight, which is expected to have no magnetostriction and to show the lowest coercive force in the magnetic field. Heat-treated filaments were also investigated in terms of the structure and the mechanical and magnetic properties. It was found that the cross-sectional areas and structures of as-spun filaments were affected by processing conditions, such as spinneret diameter, throughput, etc. In the case of the filaments with diameters less than about 90 μm, well-grown structures having primary dendrite arms oriented towards the direction of the filament length, can be observed in place of the polycrystalline structure. By means of high-temperature heat treatment, the filaments with the above structure were found to show single-crystalline structure without any clear boundary. The single-crystalline filaments were found to have good ductility to bending through 180° and excellent magnetic properties such as an almost perfect rectangular loop, low coercive force, and high saturation magnetic flux density in the d.c. magnetization curve.

1. Introduction

As is well known, it is difficult to obtain an iron–silicon (Fe–Si) alloy material having high permeability characteristics by the cold rolling method for a silicon content exceeding 4% by weight. The rapid solidification process, therefore, is generally applied to obtain ribbon-shaped Fe–Si alloys with higher silicon contents. It is reported that Fe–Si alloy shows a very good soft magnetic property, having the lowest coercive force for a silicon content around 6.5%, owing to the heat-treatment procedure [1]. Ohnaka *et al.* [2, 3] have conducted extensive works on wire processing of various kinds of metal alloys using in-rotating-water spinning method. However, few reports on Fe–Si alloy filaments of round cross-section with 6.5% Si composition have been published [4, 5]. It is expected that filament materials could be applied to wider end uses by making best use of their one dimensional properties.

In this paper, structure and properties of Fe–Si alloy filaments having a silicon content of around 6.5% are reported. In particular, the magnetic properties of single-crystalline structure filaments are compared with filaments having other structures.

2. Experimental procedure

The metal alloys used for the experiments were prepared from electrolytic iron (99.98 wt % purity) and semiconductor silicon (99.999 wt % purity) by melting in alumina crucibles (95 wt % Al_2O_3, 3 wt % SiO_2 and others) in an argon atmosphere. The actual compositions were measured by chemical analysis and they were found to be almost the same as the nominal compositions.

Rapidly quenched filaments were obtained by using an in-rotating-water spinning apparatus, shown in Fig. 1. In this spinning process, a molten metal jet, melted by a high-frequency induction furnace, is ejected through a ceramic nozzle into a water layer formed by centrifugal force on the inner surface of a rotating drum. The speed of the rotating water surface was about 10 m sec^{-1}, and the temperature of the water was 288 K. The diameters of the experimentally obtained filaments were 60 to 130 μm. Filaments were cleaned with methyl alcohol and dried in a vacuum. Heat treatment of the as-spun filaments was carried out in a vacuum-sealed quartz tube at 1473 K for 2 h.

Structural observations were made for the filament samples which were obtained by 3% to 5% nital etching and the micro etch-pits method [6], measured by optical microscopy and scanning electron microscopy (SEM). The tensile strength of the as-spun and annealed samples was measured using an Instron-type tensile testing machine, and the hardness was measured with a micro-Vickers hardness tester. The measurement of electrical resistivity was done by standard four-probe method. Magnetic properties were measured on linear filaments 70 mm long using a d.c.–*BH* curve tracer, Type 3257 Yokogawa.

3. Results and discussion

Fig. 2 shows optical micrographs of longitudinal

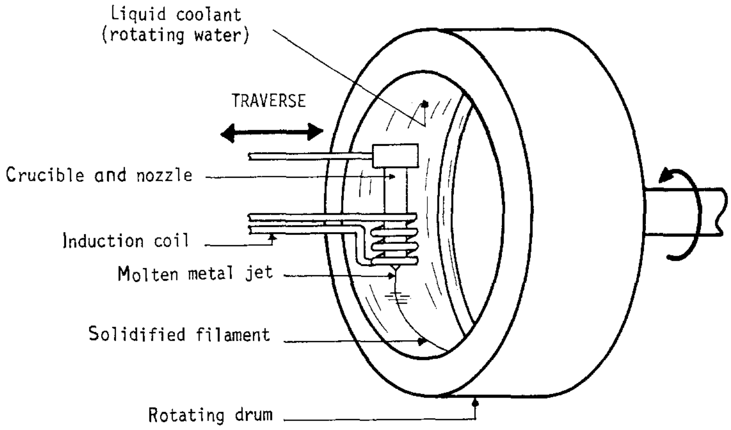


Figure 1 Schematic diagram of the in-rotating-water spinning apparatus for rapid solidification of Fe–Si alloy filaments.

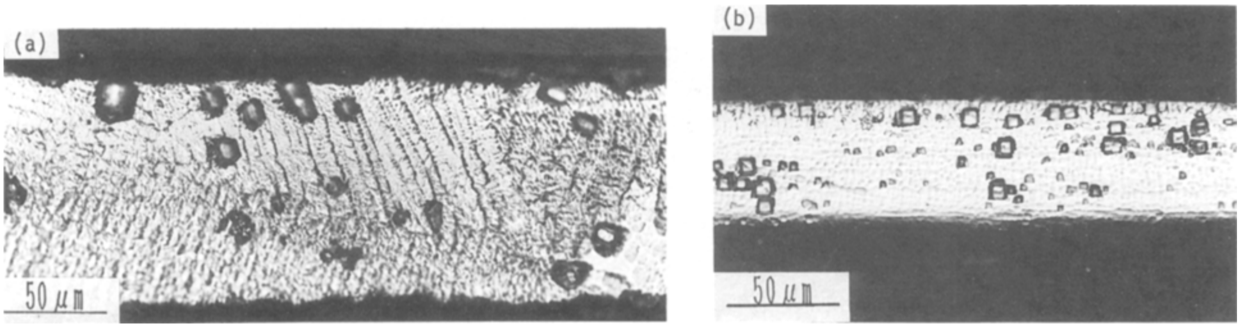


Figure 2 Optical micrographs of longitudinal cross-sections with etch pits in Fe–6.47 wt % Si as-spun filaments. Filament diameter: (a) 130 μm, (b) 70 μm.

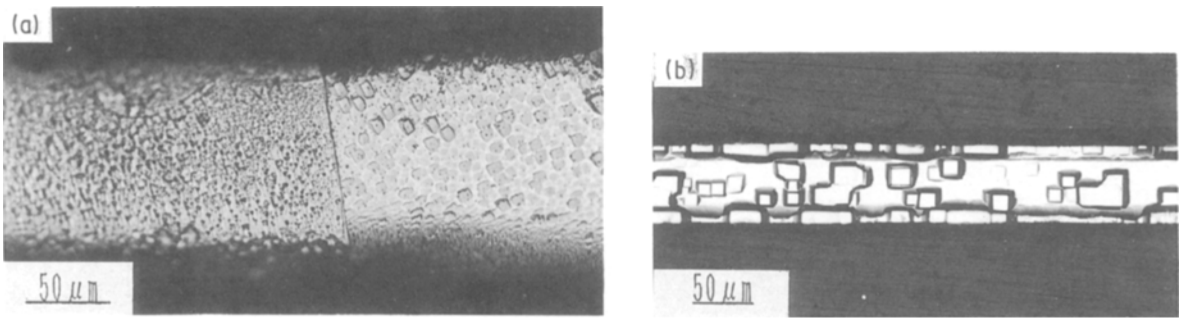


Figure 3 Optical micrographs of longitudinal cross-sections with etch pits in Fe–6. 47 wt % Si annealed filaments. Filament diameter: (a) 130 μm, (b) 70 μm.

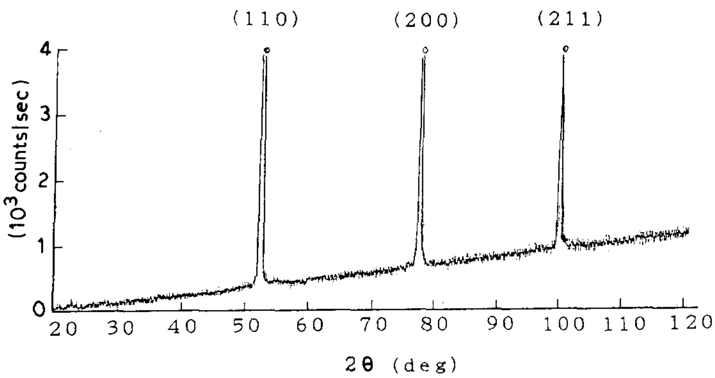


Figure 4 X-ray diffraction pattern of annealed Fe–6.47 wt % Si filament with a filament diameter of 70 μm.

cross-sections with etch pits in Fe–6.47 wt % Si as-spun filaments having diameters of 70 and 130 μm. The 130 μm diameter filaments have polycrystalline structure with crystal grains of about 100 to 600 μm in length along the direction of the filament axis. In the case of the 70 μm diameter filaments, primary dendrite arms growing along the filament direction can be observed without any clear grain boundary, which is confirmed by nital-etching observation. This may be caused by the balance of the crystal growth rate and

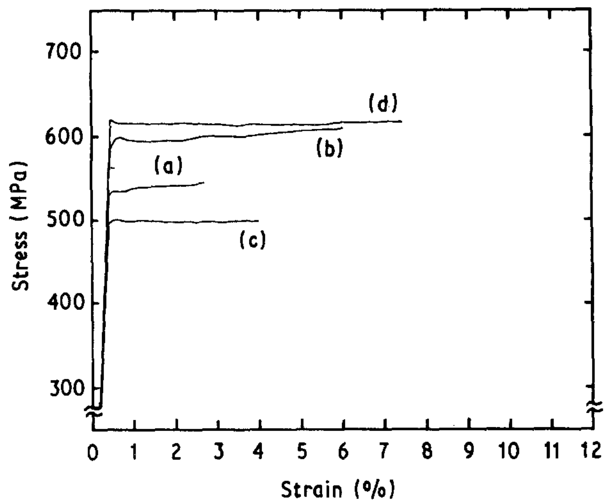


Figure 5 Stress–strain curves of as-spun Fe–6.47 wt % Si filaments and annealed filaments. As-spun filament diameter: (a) 130 μm, (b) 70 μm. Annealed filament diameter: (c) 130 μm, (d) 70 μm.

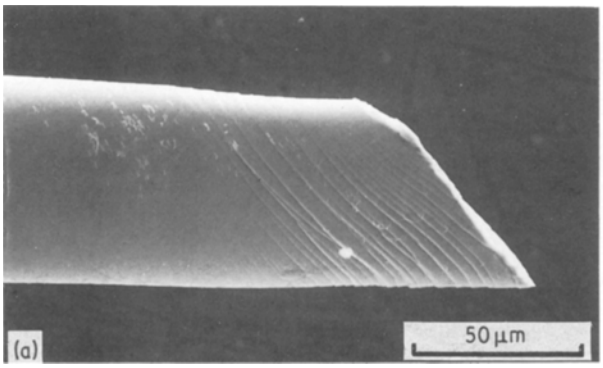
the jet speed of the molten metal, resulting in an extreme anisotropy of crystal growth.

The longitudinal cross-sections with etch pits in the annealed filament samples of Fig. 2, are shown in Fig. 3. A bamboo structure, similar to that reported by Wray [7] for an Fe–3 wt % Si alloy specimen, can be observed for the 130 μm diameter heat-treated filaments. The crystals with nodes at about 7 mm distances, are oriented to the direction of the filament axis [8, 9]. The single-crystalline structure, on the other hand, can be observed for 70 μm diameter annealed filaments. The length of the single-crystalline structure along the fibre direction might be more than 150 mm, which was confirmed by our microscopic observations.

The dependence of annealed filament structure on filament diameter may be explained in terms of the original structures of the as-spun filaments. Further investigations will be needed to clarify the mechanism of structure development. X-ray diffraction results revealed the existence of α (b′c′c) single phase having crystal planes (1 1 0), (2 0 0) and (2 1 1). However, the (1 1 1) plane which originated with ordering, is not observed in Fig. 4.

As is evident from the stress–strain curves shown in Fig. 5, the elongation of the 70 μm diameter filaments is larger than that of other filaments, showing more ductile properties. Fig. 6 shows typical SEM views of fracture edges after tensile testing. The angle between the fracture surface and the filament axis is about 50°, and many slip bands near the fracture edge can be observed along the fracture surface direction. Such slipping deformation may be similar to the theoretical aspect reported for cylindrical single-crystal materials [10]. Table I shows the micro-Vickers hardness number, electrical resistivity and Curie temperatures of the Fe–Si alloy filaments. As is seen from this table, a 70 μm diameter heat-treated filament has about twice the hardness of stainless steel, but has a lower hardness than sendust. The electrical resistivity of Fe–6.47 wt % Si alloy filaments is similar to that of sendust. The Curie temperature of the 70 μm diameter annealed filament was measured as about 973 K in agreement with the well-known phase diagram.

Fig. 7 shows the knotted state of the single-crystal structure filament, presenting evidence of ductility in spite of its higher silicon content. On the other hand,



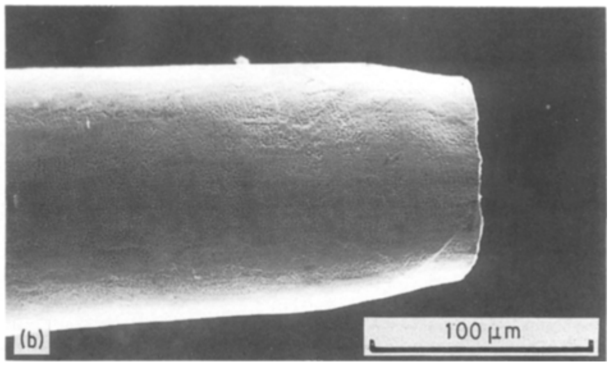


Figure 6 Scanning electron micrographs of fracture edges after tensile testing for (a) single crystalline 70 μm diameter and (b) polycrystal 130 μm diameter, filaments.

TABLE I Vickers hardness, electrical resistivity and Curie temperatures of Fe–6.47 wt % Si alloy filaments

Sample	H_v (DPN) loaded 50 g	ρ (μΩ cm) at room temperature	T_c (K)
As-spun, 130 μm diameter	392	92	–
As-spun, 70 μm diameter	400	80	–
Annealed, 130 μm diameter	401	–	–
Annealed, 70 μm diameter	430	73	973
Fe–3 wt% Si anisotropic plate	–	45 ∼ 48	–
sendust	500	80	773
Stainless steel (SUS-304)	> 190	–	–

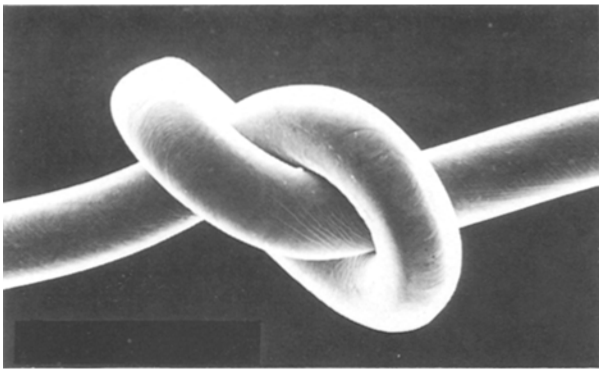


Figure 7 The knotted state of the annealed Fe–6.47 wt % Si alloy filament of 70 μm diameter, having single-crystalline structure.

the 130 μm diameter filaments have no such ductility. It might be thought that the ductile property may be attributed to the macroscopic structure of the filaments as well as their smaller cross-sectional area.

Fig. 8 shows the direct current magnetization curves (d.c.–*BH* curves) for four kinds of Fe–Si alloy filaments having different structures. The *BH* curves (a) and (b) were obtained by using as-spun filaments with diameters of 130 and 70 μm, respectively. Heat treatments of each filament give the (c) and (d) *BH* curves.

TABLE II Magnetic characteristics of Fe–6.47 wt % Si alloy filaments from the d.c.–*BH* curves

Sample	H_c (A m^{-1})	B_8* (T)	B_r/B_8
As-spun, 130 μm diameter	56.0	1.20	0.58
As-spun, 70 μm diameter	32.0	1.85	0.72
Annealed, 130 μm diameter	16.8	1.17	0.86
Annealed, 70 μm diameter	14.4	1.85	0.99

* B_8 = magnetic flux density at 800 A m^{-1}.

As discussed previously, the structures of (a), (b), (c) and (d) are polycrystal, unidirectional dendritic, bamboo and single-crystalline, respectively. Table II gives a list of coercive forces and squareness ratios for the *BH* curves measured for each sample. As is evident from Fig. 8 and Table II, the coercive force decreases and the squareness ratio increases on heat treatment. This behaviour might be caused by structural changes into well-grown crystal morphologies. The most important information obtained is that the single-crystalline Fe–Si alloy filament shows a squareness ratio of about unity and a rectangular shaped *BH* curve. To clarify the difference in magnetic property between single-crystalline and polycrystal filaments, *BH* curves corresponding to Figs 8d and a are shown

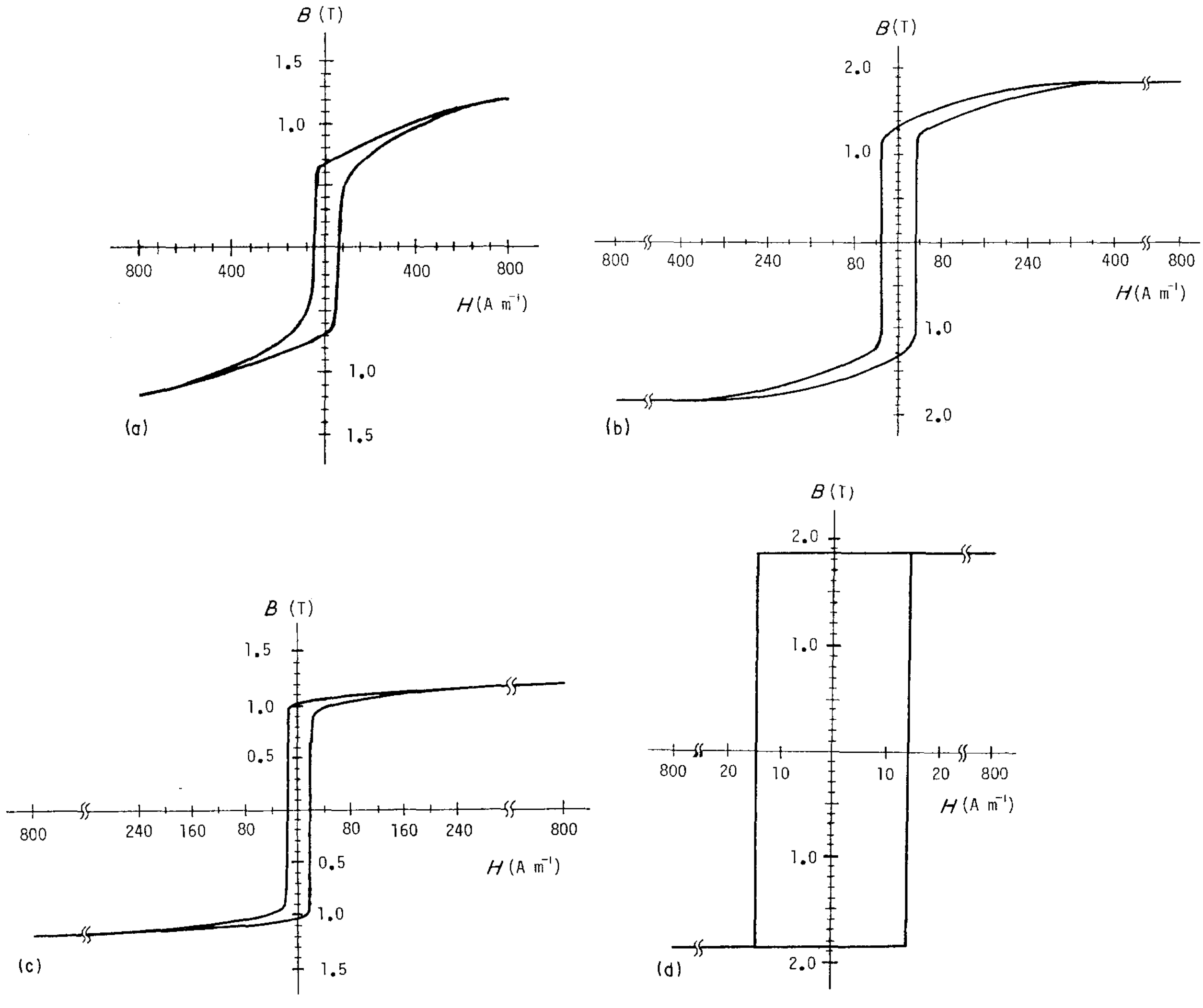


Figure 8 Direct current magnetization curves for as-spun and annealed filaments having different structures. As-spun filament diameter: (a) 130 μm, (b) 70 μm. Annealed filament diameter: (c) 130 μm, (d) 70 μm.

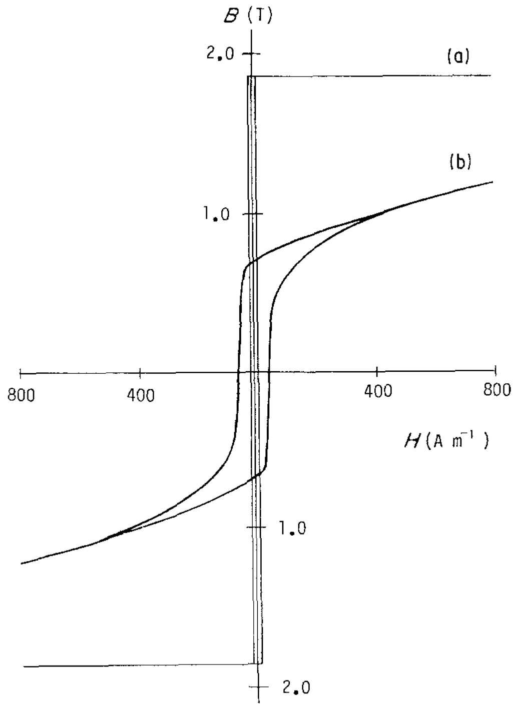


Figure 9 Direct current magnetization curves for the filaments of polycrystal structure and single crystal structure (b) as-spun filament of 130 μm diameter having polycrystal structure, (a) annealed filament of 70 μm diameter having single-crystal structure.

in Figs 9a and b, respectively. The remarkable characteristics of single-crystalline filaments can be understood from this figure.

At present, further investigations of the single-crystal structure filaments are being conducted in more detail.

4. Conclusions

Rapidly quenched Fe–Si alloy metal filaments having a silicon content of about 6.5% by weight were investigated in terms of structure and mechanical and magnetic properties. It was found that the as-spun filament of 70 μm diameter has primary dendrite arms directed along the length of the filament, and on annealing, the single-crystalline structure appears. The heat-treated Fe–Si filament has a good ductility in spite of its high silicon content. This filament also has excellent magnetic characteristics, such as a squareness ratio of about unity and rectangular shaped direct current magnetization curves.

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