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THE RELATIONSHIP BETWEEN CRITICAL CURRENT AND MICROSTRUCTURE OF INTERNAL TIN WIRE

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ABSTRACT

Prior work on internal tin wire has shown that an increase in critical current results when the Nb_3Sn reaction temperatures (650-730°C) are preceded by low temperature diffusion heat treatments that distribute the tin. These heat treatments produce a more uniform tin distribution through the niobium filament array before substantial Nb_3Sn formation has occurred. Heat treatments as long as 19 days have been proposed as the optimal heat treatment for the conductor. However, it is possible to substantially reduce the low temperature heat treatment time while retaining the same high critical current. The success of shortened heat treatments may be interpreted on the basis of the Cu-Sn reaction, diffusion kinetics and the Nb_3Sn growth kinetics.

INTRODUCTION AND SUMMARY

The internal tin process[1,2] is a fabrication technique that makes possible the increase of the fraction of Nb_3Sn in a conductor and thus increases the overall critical current. The process starts with a Cu-Nb-Sn composite that can be deformed to final size without intermediate anneals since no bronze is present and the elemental units co-deform well. In addition, the J_c of the Nb_3Sn produced by this process is high.[3,4,5] Thus the critical current of the internal tin wire is enhanced in that both the quantity and the quality of Nb_3Sn are improved in comparison to bronze-processed wires. A representative conductor of this type, which has a Ta barrier between the Cu-Nb-Sn composite and the Cu stabilizer, is shown in Figure 1.

Many different elaborate heat treatment schedules have been proposed for internal tin wire.[4,5,6,7] The highest critical currents are achieved with multiple heat treatments at different temperatures with total heat treatment times of up to 20 days. Our earlier work[8] indicates that the total heat treatment time may be reduced to about 12 days with no reduction in performance by altering the low temperature tin diffusion steps. The shortened heat treatment still includes the 218 hour heat treatment at 580°C, which has been used in several reaction schemes; however, this step has not been shown to be essential.

The selection of alternate, shortened heat treatments to form the Nb_3Sn phase here is motivated by the improved performance of a bronze-processed conductor that received a low-temperature heat treatment followed by a shorter high-temperature heat treatment.[9] The best heat

treatment for the bronze wire was 700°C/4 days followed by 730°C/2 days. These temperature and time combinations are inappropriate for the internal tin wire since overaging will occur after 4 days at 700°C.[8] Nevertheless, the concept can be applied to the internal tin wire. The low-high scheme is incorporated into most internal tin heat treatment schedules in the form of a treatment at either 580°C or 625°C followed by a treatment at or above 700°C.

The present work was undertaken to simplify and shorten the heat treatment of the internal tin wire without degrading performance and to clarify and quantify the microstructural sources of the observed critical current density. The principal results of the work are that a high overall J_c can be obtained with a short (2-3days), intermediate-temperature (580°C) heat treatment prior to 700°C, or with a 0.1°C/minute temperature ramp from room temperature to 700°C. The high J_c seems to result from the fine equiaxed grained Nb_3Sn region which forms at intermediate temperatures.

EXPERIMENTAL PROCEDURE

Two internal tin wires fabricated by Intermagnetics General Corporation were utilized in this study. One, a 1.73mm (0.068in.) wire, which was used for metallography and x-ray analysis, contained 61 sub-elements with nominal diameter of 0.12mm (0.005in.). The other wire, a 0.27mm (0.0105in.) wire (Fig. 1), was produced with a Ta barrier and Cu stabilizer specifically for critical current measurements. The internal dimensions and filament size of this wire approximated those in the large conductor. The Ta barrier and Cu stabilizer of the sub-element accounted for approximately 15% and 60% of the cross sectional area of the wire respectively.

Heat treatments were done in sealed quartz tubes back-filled with argon such that the pressure would approximate ambient pressure at the highest temperatures (650-700°C). To ensure that no tin was lost, the conductors' ends were sealed by melting until a small alloy bead was created. At least 20mm at each end of the 0.27mm diameter conductor was removed prior to critical current measurements. The heat treatment notation and the corresponding temperature/time combinations are given in Table 1. The effects of two temperature ramps, 1.0 and 0.1°C/minute from room temperature to 700°C, were also investigated.

All the critical current measurements were done in a transverse magnetic field with a potential criterion of 0.1μV/mm or less. At least five samples were measured for heat treatments with a high I_c (700°C/4d, 0.1°C/min.+700°C/1d, IVB+700°C/1d, and I+700°C/1d) while a minimum of three tests were performed on low I_c samples (IVA, IVB, and I). A four point probe arrangement was used with 30mm long samples. The voltage taps were placed 5mm apart in the middle of the sample, equidistant from the current contacts. The overall J_c is calculated from the active core area (i.e. area within the Ta barrier) which is $1.3 \times 10^{-2} mm^2$. The areas were measured from SEM micrographs using a digitizing tablet. To correct for magnification error, the active core areas were scaled by the ratio of the wire area obtained from the micrographs to the wire area determined by a physical measurement of the wire diameter.

TEM observations were performed on longitudinal sections of the 1.73mm conductor. The samples were thinned mechanically to about 0.025mm and then ion milled.

RESULTS AND DISCUSSION

Many different heat treatments have been proposed as the optimum for internal tin wires. However, there is a limited amount of work relating conductor performance to the reaction temperatures and times. Microstructural characterization of the conductor led to the development of heat treatments that included only a short low temperature step.[8] In fact, long heat treatments at 580°C may be detrimental to the magnetization properties of the conductor by promoting filament bonding (i.e. bridging). Figures 2 and 3 illustrate the positive and negative aspects of a long 580°C treatment. Figure 2 is a high magnification micrograph of an individual filament that has been etched to reveal the Nb_3Sn. At the filament periphery large grains of 200 to 300nm in size are visible. Some filaments have been observed to grow together when adjacent filaments have large grain extrusions in close proximity. These large grains can also be seen in the upper portion of the TEM micrograph in Figure 3. The same micrograph also illustrates a beneficial aspect of the 580°C treatment, which is the fine equiaxed grains (40-45nm) produced at this intermediate temperature.

Prior work has shown that a multi-phase bronze microstructure is produced after 2 days at 580°C.[8] The microstructure consisted of the α phase (Cu-Sn solid solution) and decomposed γ phase ($\alpha + \delta$). The bronze adjacent to the Nb filaments is predominantly α but contains isolated regions of decomposed γ. Energy dispersive x-ray spectroscopy indicates that the Sn distribution throughout the α phase is relatively uniform. The analyses show that the tin concentration in the bronze on either side of the Nb filament array is ~7.5at.%Sn. Recent observations show that at this point in the heat treatment the Nb filaments are already about 40-50% converted to Nb_3Sn. The extent of the filament reaction and the good tin distribution suggested that this was the appropriate time in the heat treatment schedule to increase the temperature. This led to heat treatment schedules IVA and IVB.

Alternative heat treatments which employ a slow temperature ramp to 700°C were designed to take advantage of the variation with temperature of the Nb_3Sn formation rate and the diffusivity of tin in the Cu-Sn system. Nb_3Sn does not readily form until a temperature of ~500°C yet available data on Sn diffusion in Cu[10] suggest that diffusion of Sn is relatively fast at this temperature. Therefore, it was thought that a slow ramp would produce a redistribution of tin at intermediate temperatures and provide a "low-high" temperature heat treatment for the Nb_3Sn.

The overall critical current density at 10T and 4.2K at various stages in the heat treatments described above is shown in Figure 4. The data in the lower portion of the graph are the critical current densities of wires that are partially reacted using schedules IVA, IVB, and I. The data in the upper portion of the graph are the critical currents of the wires after an additional heat treatment at 700°C. Data from reference 8 for raising the temperature directly to 700°C is included for comparison. All the samples that received an intermediate temperature (580-625°C) heat

treatment for more than 2 to 3 days by either a temperature ramp or constant temperature had a higher critical current density than those taken directly to the high reaction temperature. The heat treatment times for samples IVA, IVB, and I include the 2 day treatment at 380°C. Figure 5 shows the J_c variation with field for the three best heat treatments. There is no significant difference between these heat treatments at fields in the range 10 to 14T.

The high critical current densities measured here agree favorably with those measured by Suenaga et al. on long barrel samples of a similar wire.[11] Although different testing techniques were used, the overall critical currents shown in Fig. 5 for heat treatment I+700°C/1 day agree to within 15% of sample A in Fig. 3 of reference 11. There are at least two possible reasons for the higher J_c values given in this study (see also comments at end of text). One is the the less sensitive voltage criterion, 0.1 μV/mm as opposed to 0.01 μV/mm. However, the large n value of the conductor at 10T (35-40) has the consequence that this difference in technique can cause at most a 5% change in the critical current. The second reason is the different wire dimensions of the two conductors. Each wire must receive a different heat treatment. Minor differences in the resulting final microstructure could cause differences in J_c.

Our testing procedure, which uses very short samples, provides the accuracy and speed needed to map out a sizeable heat treatment matrix for a conductor. Although the barrel technique may be more precise and reflect the current densities expected in operating magnets more accurately, it is not as well suited to a general survey of heat treatments.

The high critical currents obtained here after slow ramps to temperature make the internal tin conductor a promising choice for large wind-and-react magnets. It may not be feasible to heat treat a large magnet uniformly in discrete temperature steps because of the time required for the temperature to stabilize throughout the magnet. By contrast, a slow ramp is a convenient treatment for a large magnet. The ramping treatment is effective since it combines time at low temperature, which distributes the tin and promotes formation of fine grained Nb_3Sn, and time at high temperature, which improves the stoichiometry of the superconductor.

CONCLUSIONS

Several shortened heat treatments produce a high overall J_c at 10T. The shortest of these reduces the total heat treatment time from approximately 19 days to about 6 days. To achieve the highest possible critical current a low-high temperature sequence is required. Some time at an intermediate temperature is necessary to obtain the fine equiaxed grain structure required for high critical currents. A temperature ramp is a practical and effective low-high heat treatment scheme.

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COMMENTS

A potential source of critical current density variation is strain in the Nb_3Sn. A compressive strain in the Nb_3Sn will reduce the critical current density. The origin of the compressive strain is the thermal contraction differences of the wire components. Altering the amount of any component (Ta, Nb_3Sn, and Cu-Sn) will change the strain in the Nb_3Sn. Since the conductor used in this study has a larger Ta diffusion barrier than that of a typical commercial wire (~15% vs. ~5%) less compressive strain is expected, and thus a higher J_c. This difference can account for about a 10-15% increase in J_c at 10T in the conductor used in this study over a commercial wire. However, the conductor referred to in reference 11 also has a diffusion barrier that accounts for about 15% of the wire cross section. Therefore, this implies that it and the wire in this study have similar compressive strains, thus, making the amount of Ta unable to account for the difference in the critical current density.

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Table 1. Heat Treatment Notation

IVA	IVB	I
380°C/2d + 580°C/3d	380°C/2d + 580°C/2d + 625°C/1d	380°C/2d + 580°C/9d

FIGURE CAPTIONS

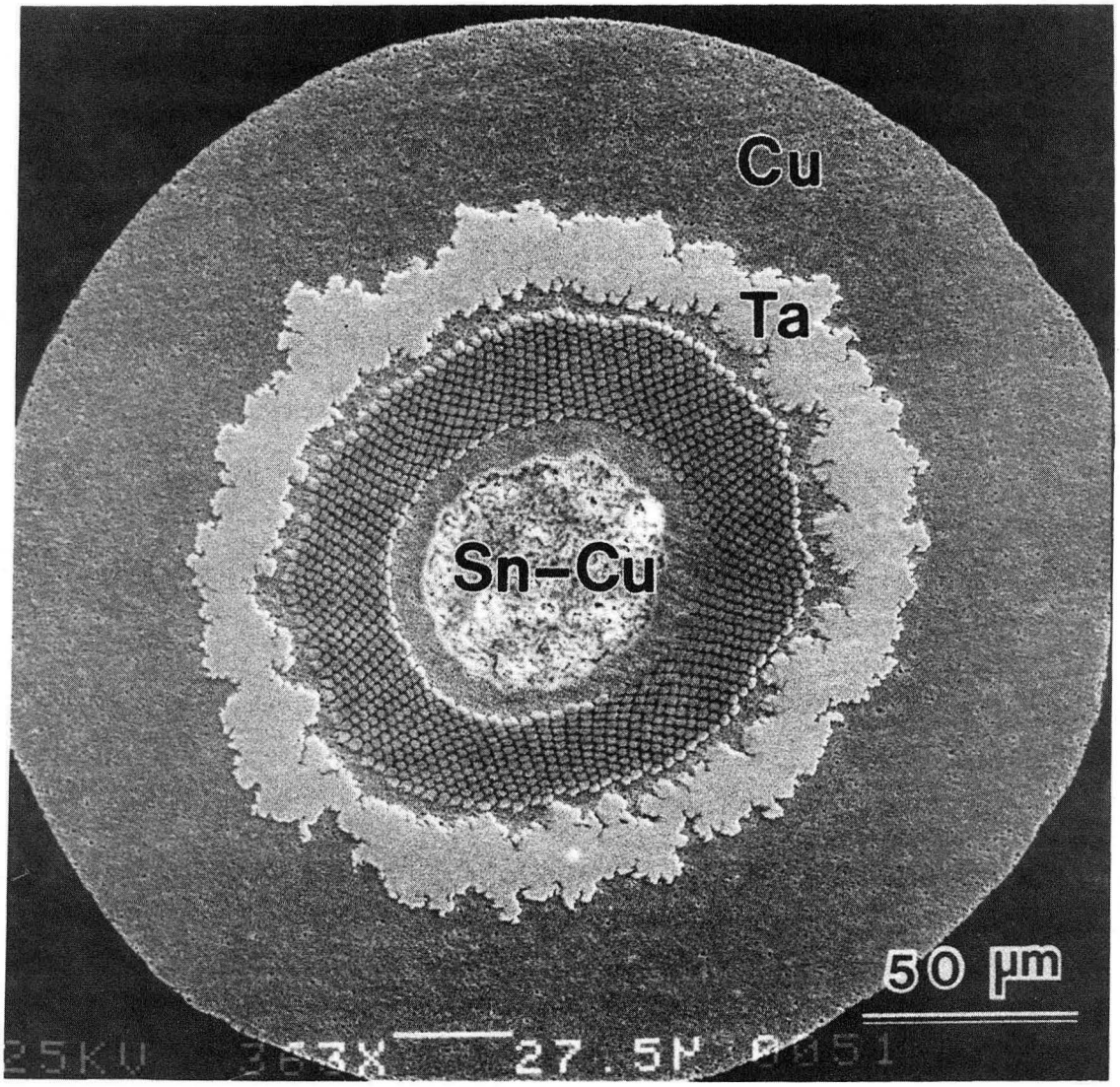
Fig. 1. SEM micrograph of an internal tin wire.

Fig. 2. SEM micrograph of a reacted filament that has received heat treatment I. The sample has been etched to reveal the Nb_3Sn. Note the large grains at the filament periphery.

Fig. 3. TEM micrograph from a sample with heat treatment I. Note the large grains (200-300nm) at the bronze interface and the small grained (40-45nm) region.

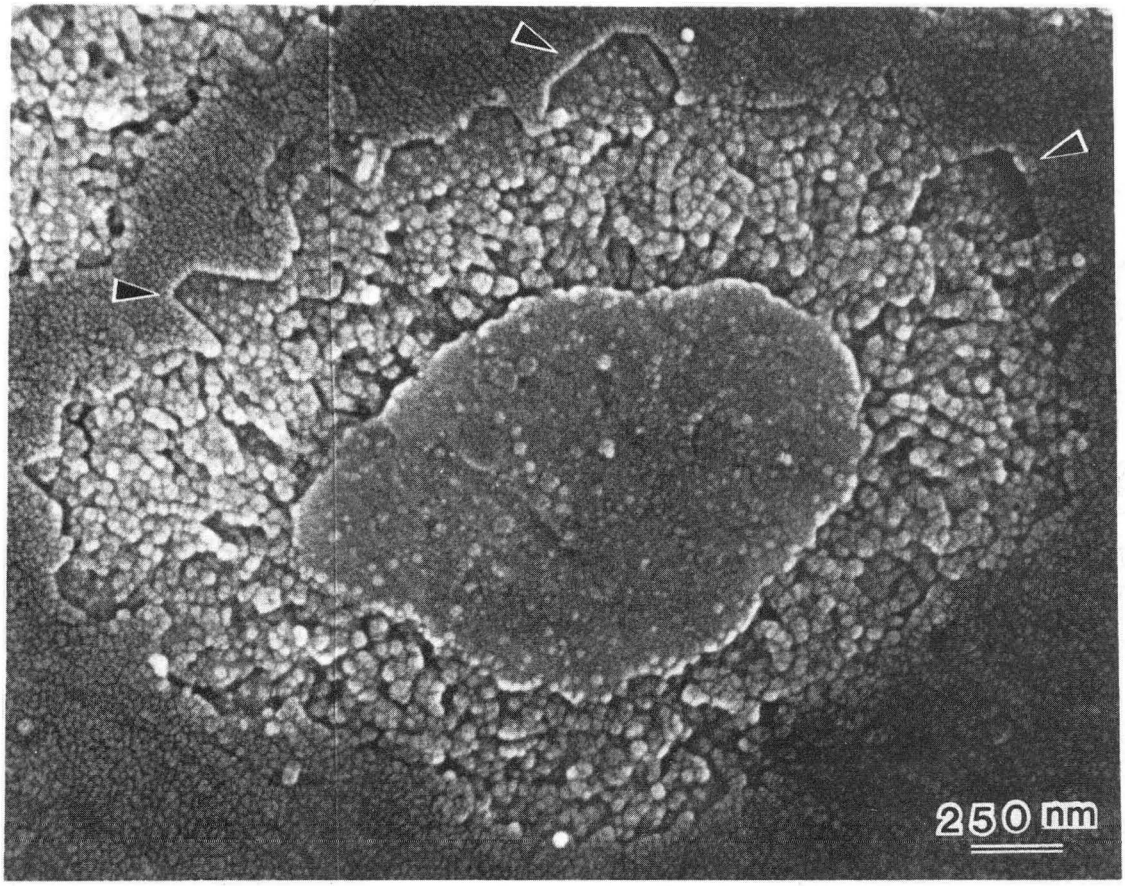
Fig. 4. The overall critical current density (10T, 4.2K) vs. the total heat treatment time the wire received.

Fig. 5. The overall critical current density (4.2K) vs. applied field.



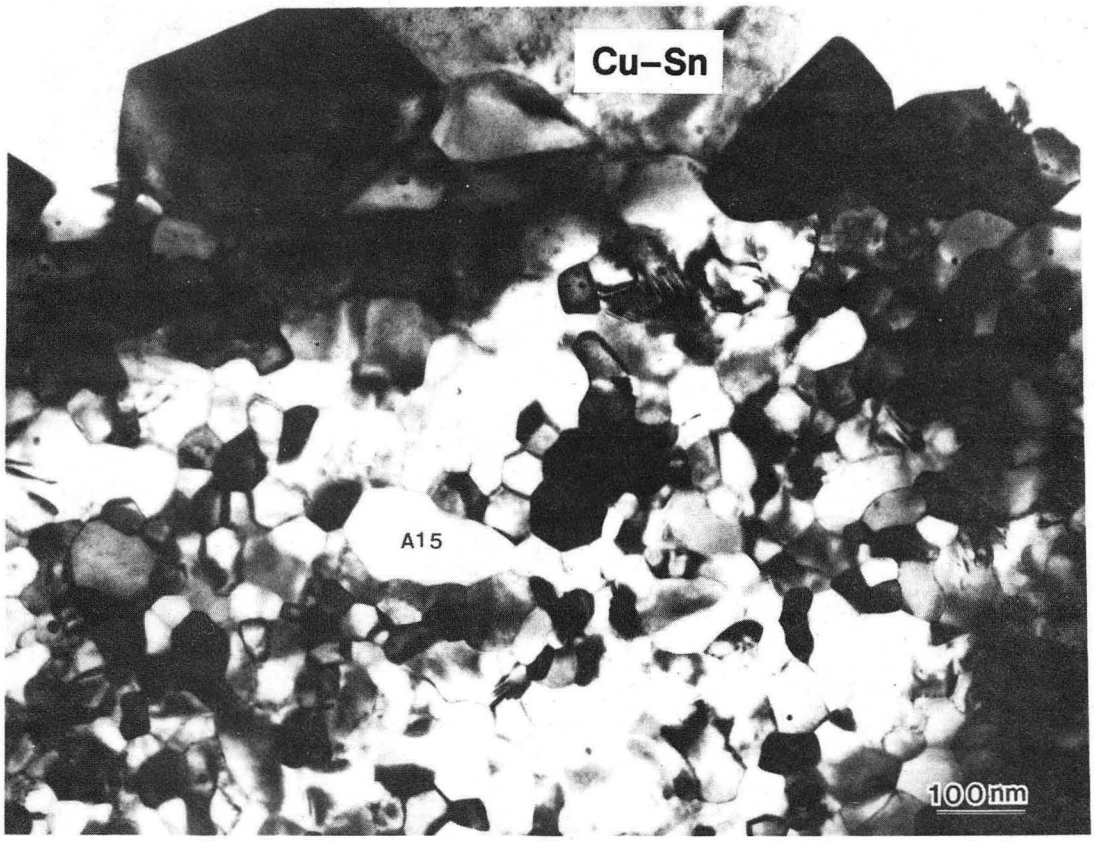
XBB 858-6103A

Fig. 1. SEM micrograph of an internal tin wire.



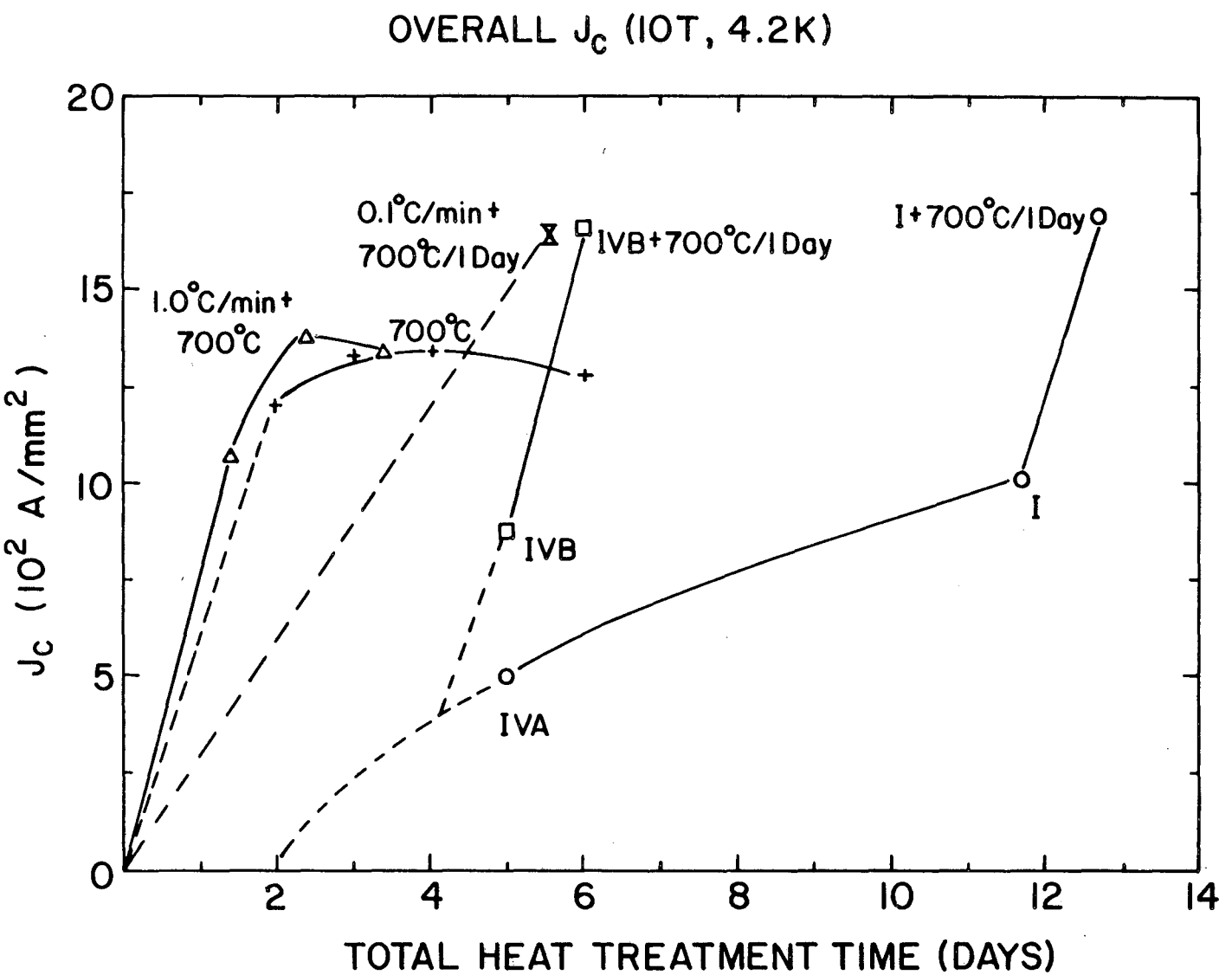
XBB 858-6105

Fig. 2. SEM micrograph of a reacted filament that has received heat treatment I. The sample has been etched to reveal the Nb_3Sn. Note the large grains at the filament periphery.



XBB 858-6099

Fig. 3. TEM micrograph from a sample with heat treatment I. Note the large grains (200-300nm) at the bronze interface and the small grained (40-45nm) region.



XBL 858-3756

Fig. 4. The overall critical current density (10T, 4.2K) vs. the total heat treatment time the wire has received.

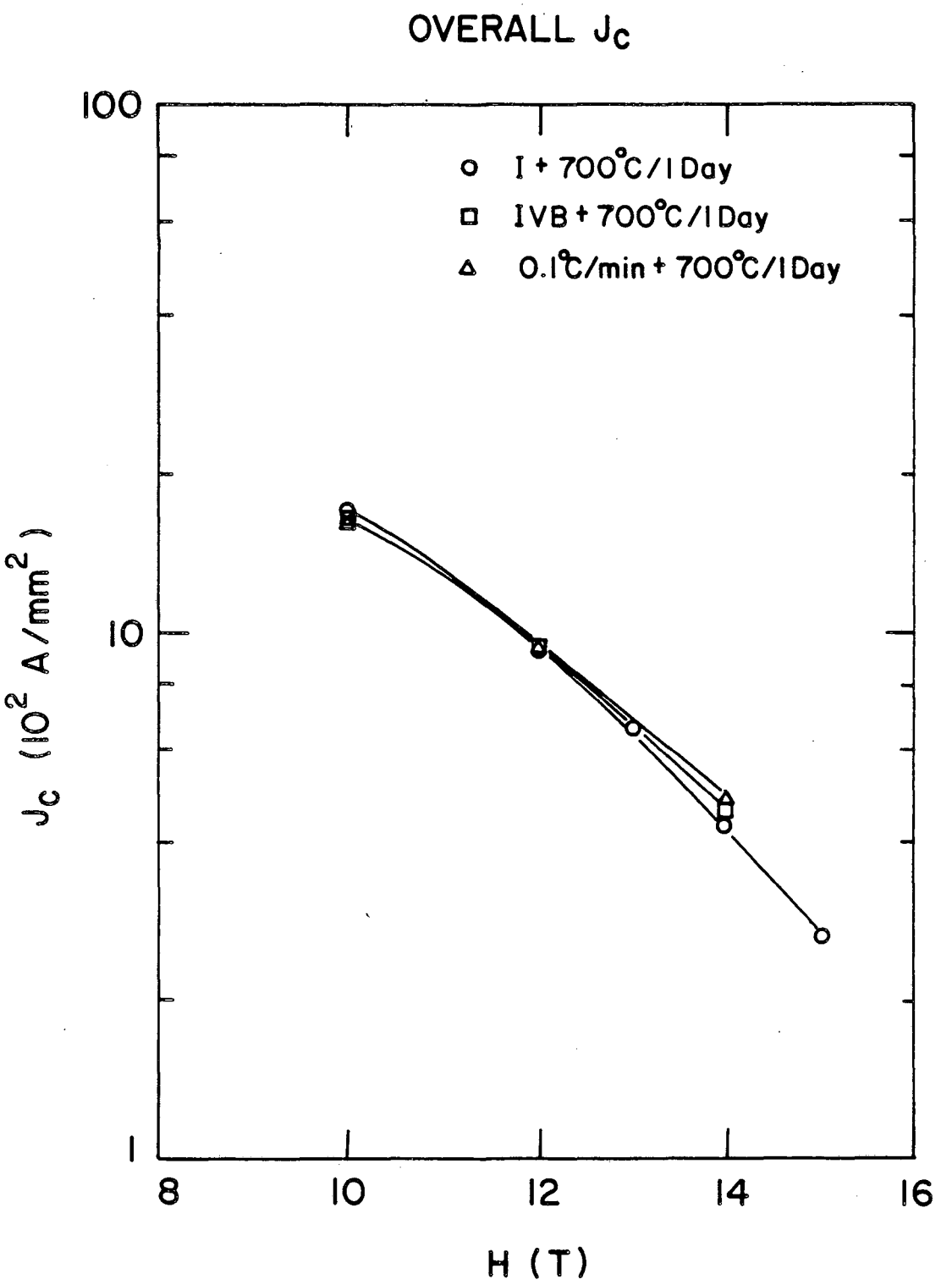


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Fig. 5. The overall critical current density (4.2K) vs. applied field.

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