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Publication Date
1983-08-01

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Presented at the International Cryogenic Materials Conference, Colorado Springs, CO, August 15-19, 1983; and to be published in Applied Physics Letters

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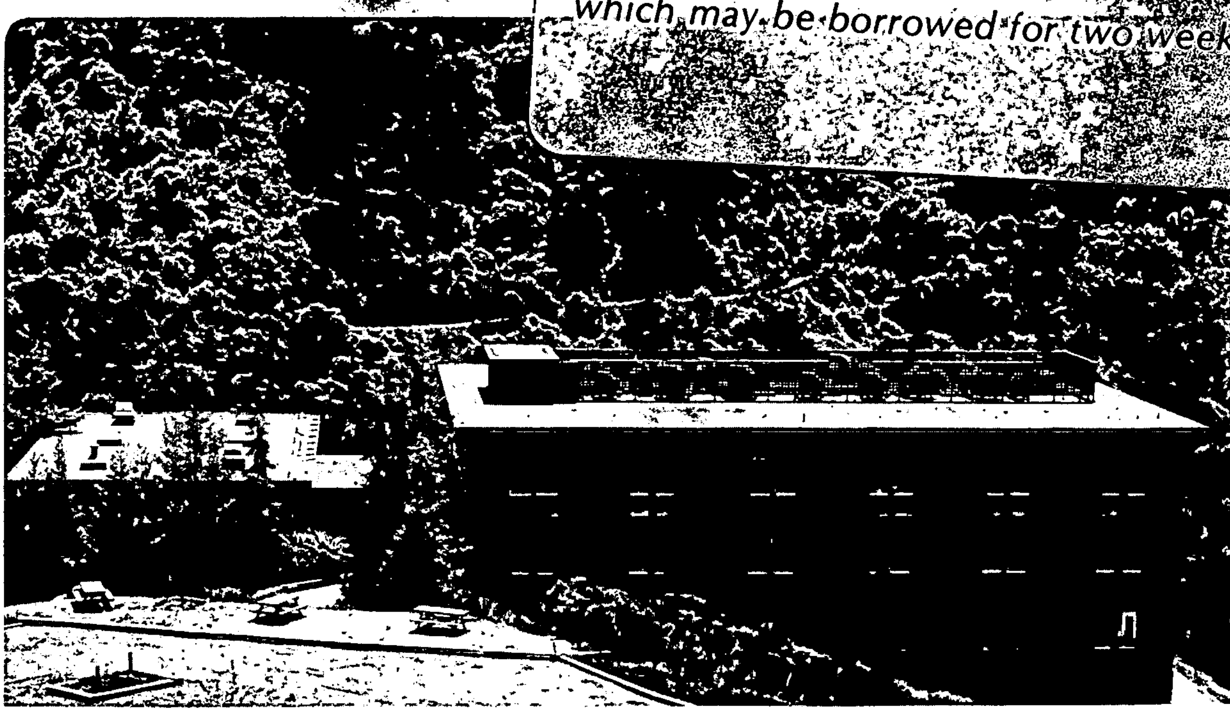
August 1983



Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

DISCLAIMER

THE CRITICAL CURRENT DENSITY OF BRONZE-PROCESSED MULTIFILAMENTARY Nb_3Sn WIRES WITH MAGNESIUM ADDITION TO THE MATRIX

by

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ABSTRACT

The effects of the addition of magnesium to the bronze of a bronze-processed multifilamentary Nb_3Sn conductor were studied. The bronze composition was varied from Cu-6.7Sn-0.0Mg to Cu-6.7Sn-0.62Mg. The magnesium was found to refine the Nb_3Sn grain size and improve the critical current density of the superconductor. The most heavily doped wire had an optimum critical current density three to six times better than the Mg-free control wire at all fields.

INTRODUCTION

Because of the relatively high critical field of Nb_3Sn, multifilamentary Nb_3Sn conductors have been proposed for a number of applications requiring fields of 8 to 15 Tesla. At the lower end of this range, NbTi is currently an economically competitive choice. At very high fields, the currently attainable critical current density of Nb_3Sn is not high enough to make an all superconductor magnet practical. As a result, there is a need for significant improvement in the critical current density of Nb_3Sn at all fields.

Previous work on a commercial bronze-processed Nb_3Sn multifilamentary wire provided the theoretical basis for this work [1-3]. The relevant conclusions of that work are summarized briefly here. The reacted A15 layer is composed of three shells. Of these, the middle fine-grained layer provides most of the current-carrying capacity of the wire. The volume fraction of the fine-grained layer is largest at intermediate reaction temperatures. Low temperature treatments promote a finer grain size, while the stoichiometry of the A15 is maximized in the higher temperature treatments. Improved critical currents are obtained from double-aging treatments with an initial low temperature step followed by a shorter treatment at a higher temperature. However, in the optimized double-aging treatment, a large fraction of coarse grained material still remains. It seems unlikely that heat treatments can be devised which will suppress the coarsening of the outer grains of these wires without sacrificing the quality of the A15. Therefore, further improvements in critical current must come from chemically induced changes in the microstructure. Previous studies have suggested that magnesium additions to the bronze matrix prior to the formation of the Nb_3Sn phase fill this role. Magnesium apparently causes both a decrease in average grain size and a retardation of grain-coarsening. These microstructural changes are associated with a significant increase in critical current density for both composite-tape at 6.5T [4,5] and multifilamentary wire at 10 to 15T [6].

This work was undertaken to determine the influence of magnesium in combination with various heat treatments on the microstructure and corresponding critical current densities of Nb_3Sn. To isolate the influence of the magnesium the wires were all heat treated to almost complete reaction. A study of the evolution of the microstructure during heat treatment is in progress.

EXPERIMENTAL

The multifilamentary wires used in this study were manufactured in the laboratory from pure starting materials. The finished wires contained 133 filaments embedded in a bronze matrix 0.5mm in outer diameter. The compositions and other physical characteristics of the wires are listed in Table I. Sample cross-sections and a more detailed description of the fabrication process have been published elsewhere [6]. Although the characteristic geometry varies among the wires, the cross-section of each wire is fairly uniform along its length. Noticeable distortion of the filaments occurred only near the periphery of the wires with higher Mg contents. The unreacted filaments, pictured in Figure 1-a, showed only drawing marks; no evidence of the formation of Nb_3Sn during fabrication anneals was found by direct SEM observation, T_c measurement, or SEM/EDXS analysis.

To form the Nb_3Sn, the finished wires were heat treated in sealed quartz tubes backfilled with argon. The heat treatment temperatures were chosen to bracket the optimum reaction temperatures of 700 to 730°C determined previously for conventional bronze-processed wires. Both the initial rate of the reaction forming Nb_3Sn at the Nb-bronze interface and the reaction rate constant varied among the wires. The source of the difference is not clear because the kinetics of the reaction depend on the filament size and shape as well as on composition and temperature. However, the magnitude of the variation made it necessary to compare wires at the same state of reaction so that valid conclusions about the effect of magnesium on the microstructure and critical current density of the A15 layer could be drawn. The extent of the reaction was defined as the A15 area in the wire cross section divided by the area of the A15 + Nb core. Since the overall critical current density of the conductor is important in practice, ninety-five percent reaction was chosen so as to maximize the amount of superconductor without degrading its quality by overaging. Heat treatment times were varied accordingly (except at 650°C where the slow reaction rate dictated comparison at 65% reaction).

RESULTS AND DISCUSSION

The critical currents I_c of 30mm long wire specimens were measured at 4.2K in a transverse magnetic field at the Francis Bitter National

Magnet Laboratory. The four-point probe technique was used with the critical current defined as the current generating a potential of one microvolt between voltage taps placed 5mm apart. The critical current density J_c was determined by dividing the I_c by the cross-sectional area of the A15 phase.

The most important effect of the Mg additions on the microstructure at complete reaction is on the A15 grain size distribution. Previous studies of the grain structure of reacted Nb_3Sn in a commercial bronze-processed wire showed that the layer is divisible into three morphologically distinct regions: an inner layer of columnar grains growing out from the Nb interface, an intermediate layer of fine-grained material, and an outer layer of large, coarsened grains. The radial geometry of the filament means that the outer shell constitutes a large volume fraction of the A15. The undoped 6.7 at.% Sn wire of this study showed a similar A15 grain morphology. Figure 1-b shows the relatively fine grains at the A15-bronze interface of the reacted filaments of the Mg-doped wires. Comparison of Mg-doped and Mg-free wires at the same aging temperature and state of reaction shows clearly that the Mg suppresses coarsening of the outer grains so that the layer is uniformly fine-grained. The addition of Mg also seems to decrease the minimum grain size. However, preliminary studies of the grain size by SEM show that this grain size is not significantly smaller for high Mg wires than for low Mg wires.

The addition of Mg to the bronze increased the peak J_c over the range 8 to 15T for all aging temperatures. The incremental improvement in the J_c increased with Mg concentration. These results confirm qualitatively an earlier report that Mg additions improve J_c in these wires [6]. Previous work by Tachikawa on tapes of similar composition showed a slight improvement in J_c at 750°C and 800°C (~25% at 6.5T) [5,7]. The present report indicates that magnesium additions are considerably more beneficial at high fields. This finding is not necessarily in contradiction with Tachikawa's results, since the data also indicate that the effect increases with applied field. It should be noted that comparison of wires at the same state of reaction reveals a much greater benefit from the Mg than the isochronal comparison used previously. This discrepancy arises from variations in the growth kinetics of the Nb_3Sn among the wires. The time required to optimize a Mg-free wire is generally sufficient to overage the Mg-doped sample, thus degrading its performance. As a consequence, comparisons between equal aging times are not meaningful.

The improvement in critical current density from Mg additions is illustrated in Figures 2 and 3, which show critical current densities at 10 and 15T respectively. Data for the 6.7Sn-0.0Mg wire at 700°C has been omitted from the figure because of anomalously large scatter. At 10T the maximum critical current obtained for the 6.7Sn-0.62Mg wire represents an improvement of ~300% over the highest value achievable by a single-step heat treatment of the Mg-free wire; at 15T the increase is ~600%. The optimum aging temperature for the Mg-doped wires was between 700°C and 730°C at 10T and shifted upward to the range 730°C to 750°C at 15T. This shift with increasing field parallels a similar one in other Mg-free bronze-processed wires [1]. Because the J_c is more sensitive to the stoichiometry of the Nb_3Sn at higher fields, higher temperature treatments in which the Sn is better homogenized become increasingly favorable as the applied field is raised. The maximum J_c at a given field is determined by the balance between the improvement of the intrinsic properties of the Nb_3Sn at higher temperatures and the corresponding degradation of its overall properties by grain growth. The optimum aging temperatures for Mg-doped wires are expected to be higher than for Mg-free wires because grain coarsening is suppressed.

The suppression of grain coarsening found in the Mg-doped wires may account for a large part of the increase in J_c. It also apparently accounts for the shift in the optimal aging temperature. However, the J_c increases monotonically with Mg content, while the apparent grain size of etched filaments observed by SEM is not significantly smaller for the high Mg wires than for the low Mg wires. This observation indicates that Mg may contribute to the properties of the wire by some other mechanism. Preliminary measurements of T_c and H_{c2} of the fully reacted wires showed that the T_c midpoints of the Mg-doped and Mg-free wires are similar, and that H_{c2} increases only slightly with Mg addition. The increase in H_{c2} is far too small to account for a major part of the increase in J_c. These data indicate that Mg does not improve the inherent superconducting properties of the best A15 formed, but do not rule out the possibility that there is more material with optimal properties present in the Mg-doped wires. Tests of similar prototype wires with Sn contents near the solubility limit in Cu (7.8 at.%) show that increasing the Sn supply and presumably the quality of the A15 increases the J_c, although not to the level of the better Mg-doped wires.

A number of more subtle explanations may be postulated to explain the effect of Mg. One variable which affects the J_c is the state of strain in the A15. However, it is unlikely that strain effects account for the increase in J_c both because the H_{c2} does not change and because the wire should be relatively insensitive to slight changes in compressional stress because it has a high bronze to niobium ratio [7]. The most fundamental determinants of the critical current density of the superconducting layer are the strength and number of flux-pinning sites. These are fixed by its microchemistry and microstructure. In filamentary wires, grain boundaries are believed to dominate flux pinning [1,8]. A possible explanation of the effect of magnesium is that it provides other pinning sites in addition to the grain boundaries. A more likely theory is that the magnesium increases the strength of the grain boundary pinning. These questions are under study. Because the Mg does not appear to affect the intrinsic properties of the Nb_3Sn, the simultaneous addition of another element which improves the inherent properties of the superconductor appears a promising path toward the achievement of even higher critical current densities.

ACKNOWLEDGEMENTS

The authors are grateful to Dr. L. Rubin and his associates at the Francis Bitter National Magnet Laboratory. One of the authors, I.W. Wu, would like to thank the IBM Corporation for financial aid through the Thomas J. Watson scholarship. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division and the Director, Division of High Energy Physics, U.S. Department of Energy under contract #DE-AC03-76SF00098.

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Table I. Composition, nomenation, filament size (D) and bronze to Nb ratio (R) of the experimental wires.

Composition (at.%) Cu-6.7 Sn-x Mg	Nomenation[1]	D (μm)	R
0.0	6700	11.4	14
0.1	6710	13.4	10
0.2	6720	10.9	15.3
0.35	6735	10.7	15.9
0.62	6762	12.5	12

[1] The first two digits of the code refer to the Sn content in atomic percent, the second two to the initial Mg concentration in the bronze.

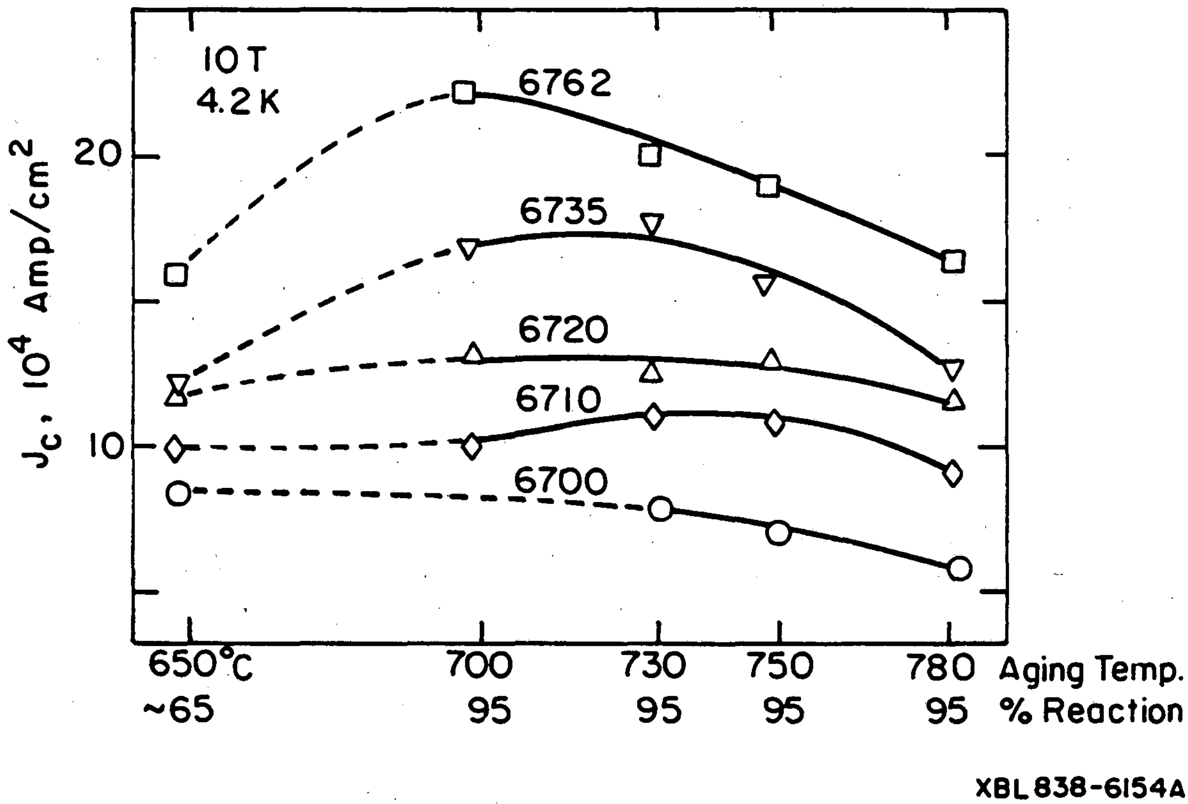

Figure 1. Critical current densities at 10T for ~95% reacted wires aged at various temperatures. Data for wires aged to ~65% reaction at 650°C are shown for comparison.

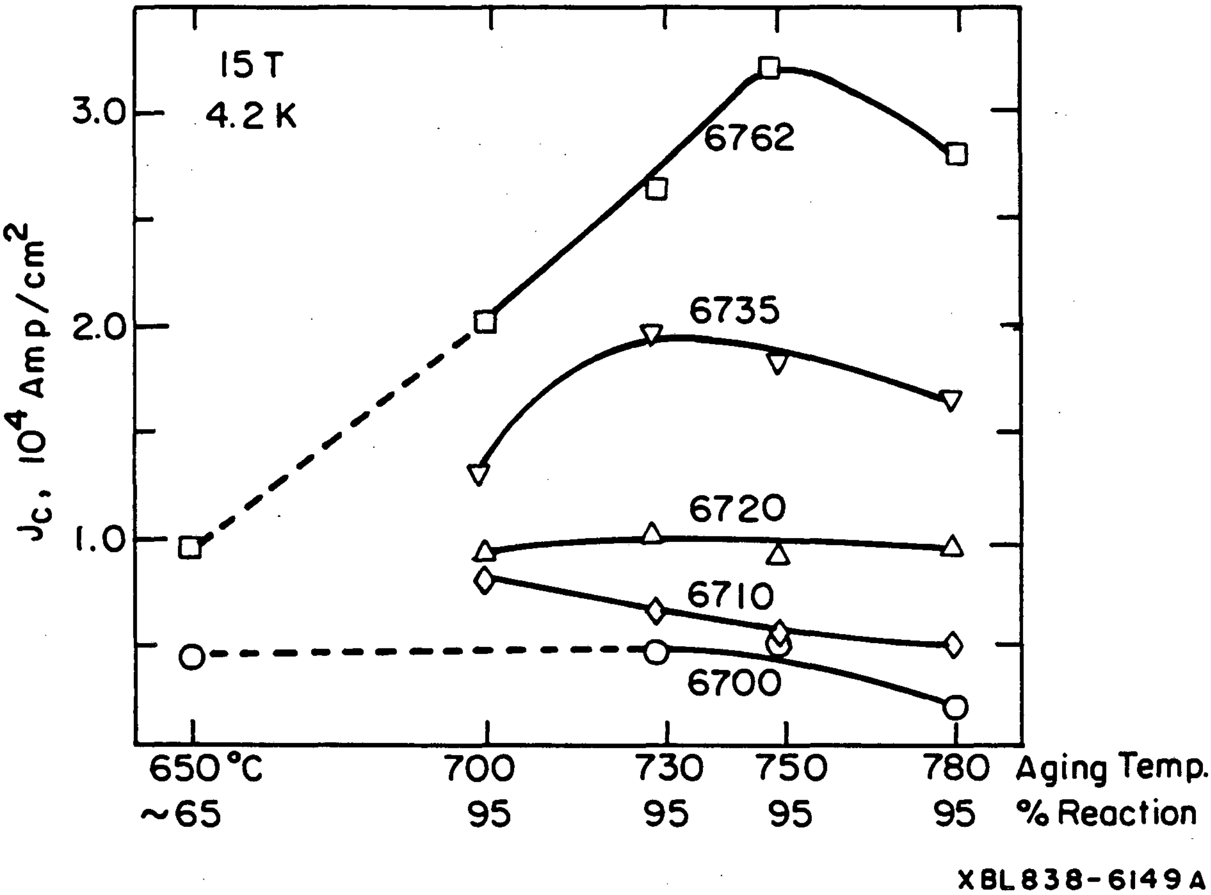

Figure 2. Critical current densities at 15T for ~95% reacted wires aged at various temperatures. The range of values for wires aged to ~65% reaction at 650°C is indicated for comparison.

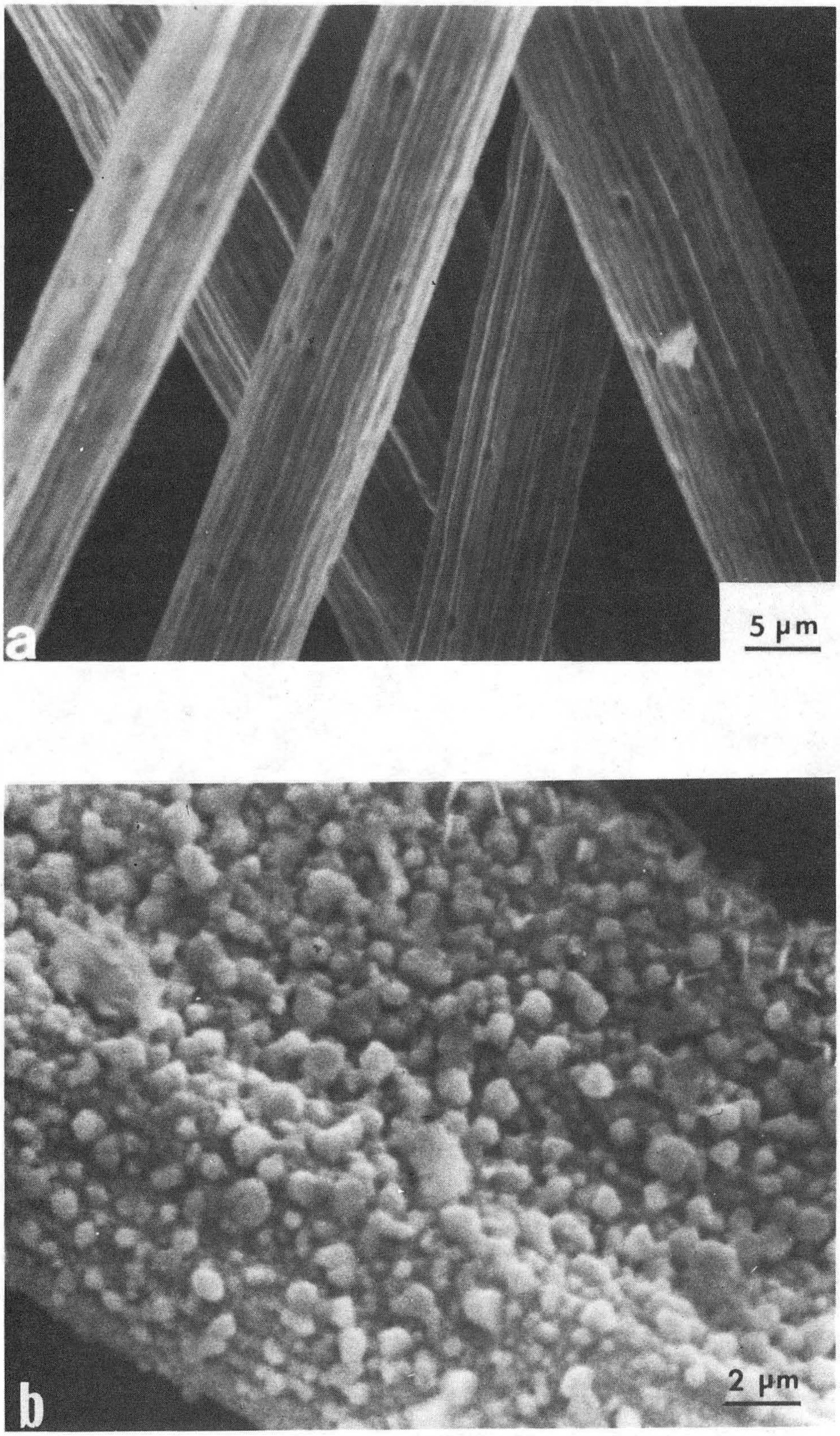

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Figure 3. Scanning electron micrographs of the filaments of the wire after the bronze matrix has been removed with an acid etch. a) Unreacted niobium filaments of 6.7Sn-0.62Mg wire b) The Nb_3Sn filaments of the wire after aging at 730^oC to ~95% of complete reaction.

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