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Author
Pickus, M.R.

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M. R. Pickus, M. P. Dariel, J. T. Holthuis,
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MULTIFILAMENTARY Nb-Nb_3(Al,Ge) SUPERCONDUCTORS

M. R. Pickus, M. P. Dariel,* J. T. Holthuis,
J. Ling-Fai Wang and J. Granda

Materials and Molecular Research Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

ABSTRACT

Multifilamentary Nb-Nb_3(Al,Ge) superconductors were prepared using an infiltration process. The critical temperature of samples diffusion reacted in the 1250-1700°C temperature interval ranged from 16 to 19.0°K. A critical current density of $10^5 A/cm^2$ at 50 kOe was observed for samples reacted at the lower temperatures.

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* On leave from the Nuclear Research Center-NEGEV and the Department of Materials Engineering, Ben-Gurion University, Beer-Sheva, Israel.

Extensive research has been conducted in recent years toward the development of multifilamentary superconductors based on A15-type compounds. Some of these compounds have the highest known values of critical temperature (T_c) and critical field (H_c). They are, therefore, prime candidates for a variety of technological applications. A method commonly known as the "bronze process" has been successfully employed for the preparation of Nb_3Sn and V_3Ga composite superconductors.[1] This process which has reached the stage of commercial development is, however, adaptable to a limited number of systems. In particular, it is not applicable to the fabrication of multifilamentary superconductors of Nb_3Ga or $Nb_3(Al,Ge)$.[2] The latter compound with a critical temperature of 21°K[3] and a critical field of 400 kOe[4] appears especially attractive for high field applications such as controlled thermonuclear reactors, if suitable methods of fabrication could be developed.

Several methods for preparing $Nb_3(Al,Ge)$ superconductors have indeed been reported.[5-8] None of them, however, relate to the fabrication of multifilamentary conductors. It is well known that, as a result of stability considerations, the multifilamentary configuration is essential for optimal utilization of the A15 compounds.

In addition to the above mentioned bronze process, an alternative method of fabricating Nb_3Sn multifilamentary superconductors, based on an infiltration technique, has recently been reported.[9-10] The resulting material possesses a high field current carrying capacity which is as good or even slightly superior to that shown by samples prepared by the bronze method. The main significance of the infiltration process, however, lies in its versatility and its potential applicability to systems for which the bronze method cannot be used. The present communication summarizes our preliminary results in applying the infiltration method to the Nb-Al-Ge system.

The preparation of Nb rods with a controlled porosity has been previously described.[11] Briefly, it consisted of isostatically pressing fine Nb powder and sintering at 2250°C for 15 min in a vacuum of the order of 10^{-5}mm Hg. After furnace cooling and without exposing the sintered compact to room atmosphere, the porous rod was immersed in a liquid Al-Ge alloy bath of eutectic composition, maintained at 700°C. After immersion, the chamber was back-filled with helium at 1 atm. pressure.

The infiltrated Nb rod was doubly sheathed; first in Ta and then in monel tubing. This assembly was form-rolled, and after removal of the monel, was further reduced by flat rolling to 0.04-0.015cm thick tape. Sections, approximately 2cm long, were subjected to a series of reaction and post-reaction heat treatments. While only the results for flat-rolled ribbons are reported here, 0.025cm diameter multifilamentary wires were also fabricated.

The microstructure of the samples at various stages of their preparation was examined by optical and scanning electron microscopy. An EDAX attachment on the SEM and an electron microprobe were used for phase identification and for the determination of the compositional relationships in the various phases. Prior to the measurement of the superconductive properties, the Ta sheath was stripped off the samples. The critical temperature T_c was measured by a standard inductive method using a Ge thermistor for the temperature determination with an estimated uncertainty of 0.1°K. The reported values correspond to the points showing the highest rate of change of the inductance in the transition region. Spot welding was used to attach current and potential leads to the samples. The critical current density J_c at 4.2°K was determined by a pulse field technique in fields up to 150 kOe. The uncertainty of the reported values is 15%, mainly as a result of the difficulty in determining accurately the volume fraction of the current carrying A15 phase.

The metallographic examination of the Nb compacts following their immersion in the liquid Al-Ge alloy showed that the samples had been fully infiltrated (Fig. 1). Following the deformation stage, the Al-Ge infiltrate appeared in the form of filaments made up of individual Al and Ge particles aligned in the rolling direction. In efforts made to carry-out the deformation at elevated temperatures, it was observed that the compound Nb(Al,Ge)$_3$ nucleates and grows at rather low temperatures. In fact, a treatment of 12 hours at 400°C, resulted in the virtually complete conversion of the eutectic to this compound.

We have investigated samples reacted in the 1250-1700°C temperature range. In samples reacted at the higher temperatures, the A15 phase appeared in the form of bands occupying up to 50% of the total volume. In the samples reacted at the lower temperatures, the A15 phase appeared in the form of thin layers (1-2μm thick) interfaced between the Nb matrix and the phases rich in Al and Ge [Nb$_2$(Al,Ge) and Nb$_5$(Al,Ge)$_3$] - (Fig. 2). Pores were also apparent in the center of the A15 phase due to a large negative volume change associated with its formation from the constituent elements. The microprobe data showed that for a sample reacted at 1700°C for 15 min, the Al/Ge ratio was constant throughout the A15 bands and also through the A2 Nb(Al,Ge) solid solution. The results also indicate that the A15 phase after this treatment contains 79 at.% Nb. This concentration is slightly higher than reported by Müller[12] for the A15 monophase region in the isothermal section of the ternary diagram at 1840°C. The result is consistent with the general trend apparent in the Nb-Al diagram,[13] according to which the Nb content of the A15 phase increases with decreasing temperature.

The multifilamentary superconductor is formed as a result of a diffusion reaction. Its microstructure and superconductive properties are determined by the temperature and duration of the diffusion anneal, and also, by the local boundary conditions prevailing at the beginning of the reaction. The initial boundary conditions depend on the morphology and distribution of the phases, i.e., Nb and the Al-Ge eutectic, taking part in the reaction. These are determined by the local spatial configuration following the infiltration stage and by the nature and amount of deformation undergone by the sample. Considering the complexity of the system and the large number of parameters, only general conclusions can be drawn at this time. These conclusions reflect the major trends concerning the dependence of the superconductive properties on the processing parameters.

The measured critical temperatures of samples having undergone various treatments are listed in Table I. The data indicate an increase of T_c with increasing reaction temperature. However, even for a given reaction temperature, there is a range for the observed T_c's. Shorter durations and larger amounts of deformation tend to increase the T_c's. A post-reaction anneal at 750°C increases T_c, in agreement with published information.

The values of T_c observed, thus far, are lower than the best values reported for the Nb-Al-Ge system. The lower

T_c values of our present samples are probably due to the A15 phase having a composition off stoichiometry.

The results of some of our current carrying capacity measurements, as well as some of the previously published data are shown in Fig 3. It clearly appears that the critical current density is markedly improved by decreasing the temperature of the reaction anneal and its duration. This effect, however, is counterbalanced to a certain extent, by the reduced volume fraction of the A15 phase formed under these conditions, resulting in a decreased overall current carrying capacity. We can expect that by using an initially finer dispersion of the infiltrated phase followed by a more drastic deformation, the interphase area between the components taking part in the diffusion reaction will be increased. This, in principle, should result in an increased volume fraction of the A15 phase without having used lengthy anneals or elevated temperatures. The large volume fraction of A15 formed at temperatures in excess of 1600°C has a poor current carrying capacity as a result of its relatively coarse grain size. Grain boundaries are known to be effective pinning centers in A15 superconductors. In our samples reacted at low temperatures, the grain size is of the order of the filament diameter, i.e., 1-2μm.

The present results are significant in two respects. The method of preparation employed yields a truly multifilamentary morphology. Our measured critical current densities are appreciably higher than any of those previously reported for bulk superconductors in the Nb-Al-Ge system. At present, it appears that the highest current densities are associated with the lower critical temperatures. We can expect that further optimization of the processing parameters will lead to improved properties with respect to both the current carrying capacity and the critical temperatures. This will allow a closer approach to the realization of the full potential of this system.

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Table I. Critical temperature of Nb-Nb_3(Al,Ge) multifilamentary superconductors.

Reaction Anneal		Post-Reaction Anneal		T_c
Temp. (°C)	Time (Sec)	Temp. (°C)	Time (h)	(K)
1700	60	-	-	17.4
1700	60	750	48	18.7
1300	900	-	-	15.4
1300	60	-	-	15.6
1300	60	750	48	16.3
1700	60	750	96	16.8
1250	60	750	96	17.2
1300	30	750	96	17.2
1300	30	750	250	17.4
1400	30	750	96	18.2
1500	30	750	96	18.5
1600	15	750	96	19.1
1700	15	750	96	18.7
1300*	30	750	96	17.5

* Sample thickness 0.015cm: for all others 0.04cm.

FIGURE CAPTIONS

Figure 1 - Sintered Nb compact infiltrated with an Al-Ge eutectic alloy.

Figure 2 - Scanning electron micrograph of $Nb_3(Al,Ge)$ filaments in a Nb matrix. Traces of Al and Ge rich phases surrounded by the A15 filaments are also visible.

Figure 3 - Magnetic field dependence of the critical current density for $Nb_3(Al,Ge)$ bulk superconductors.

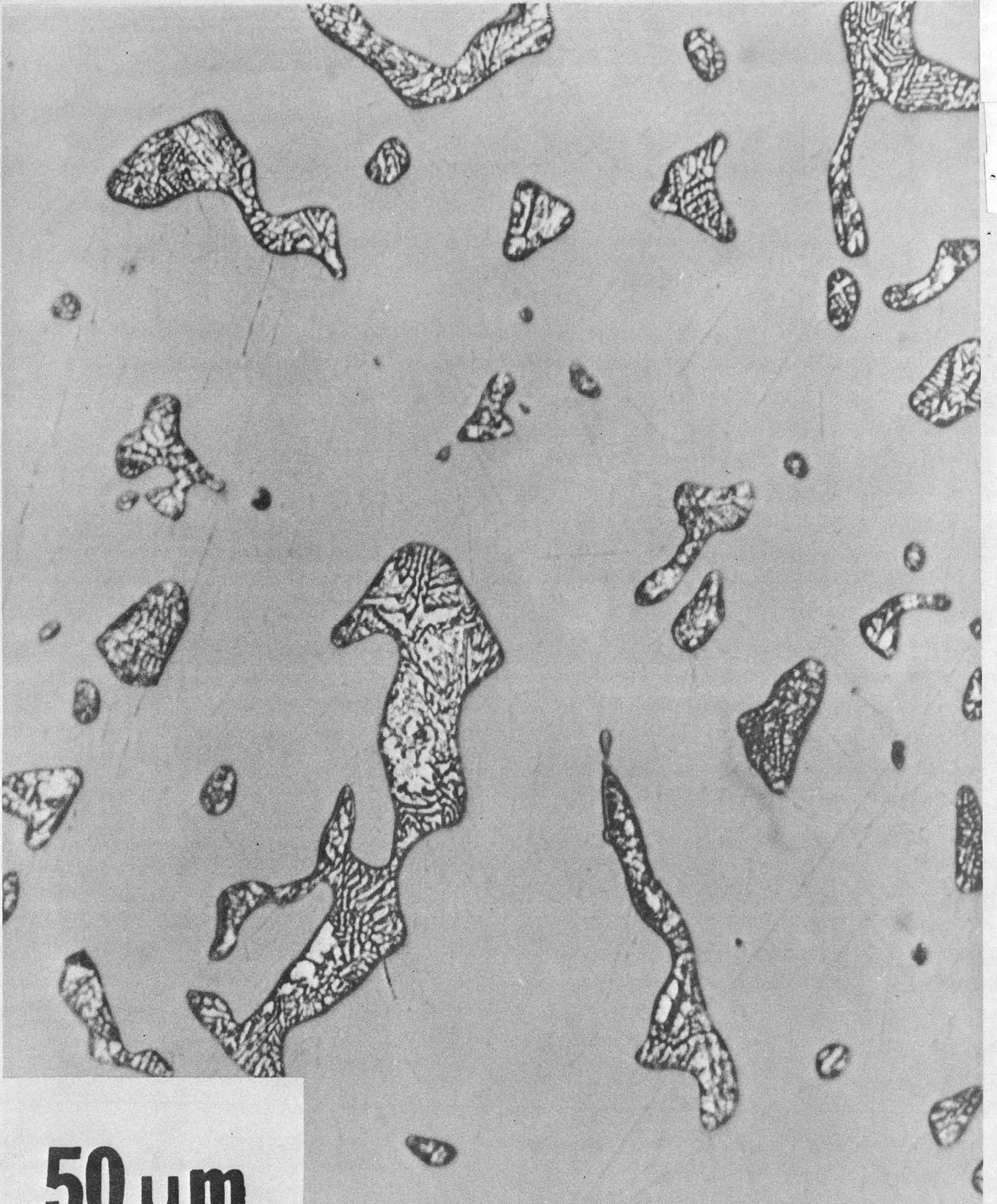

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Fig. 1 Sintered Nb compact infiltrated with an Al-Ge eutectic alloy

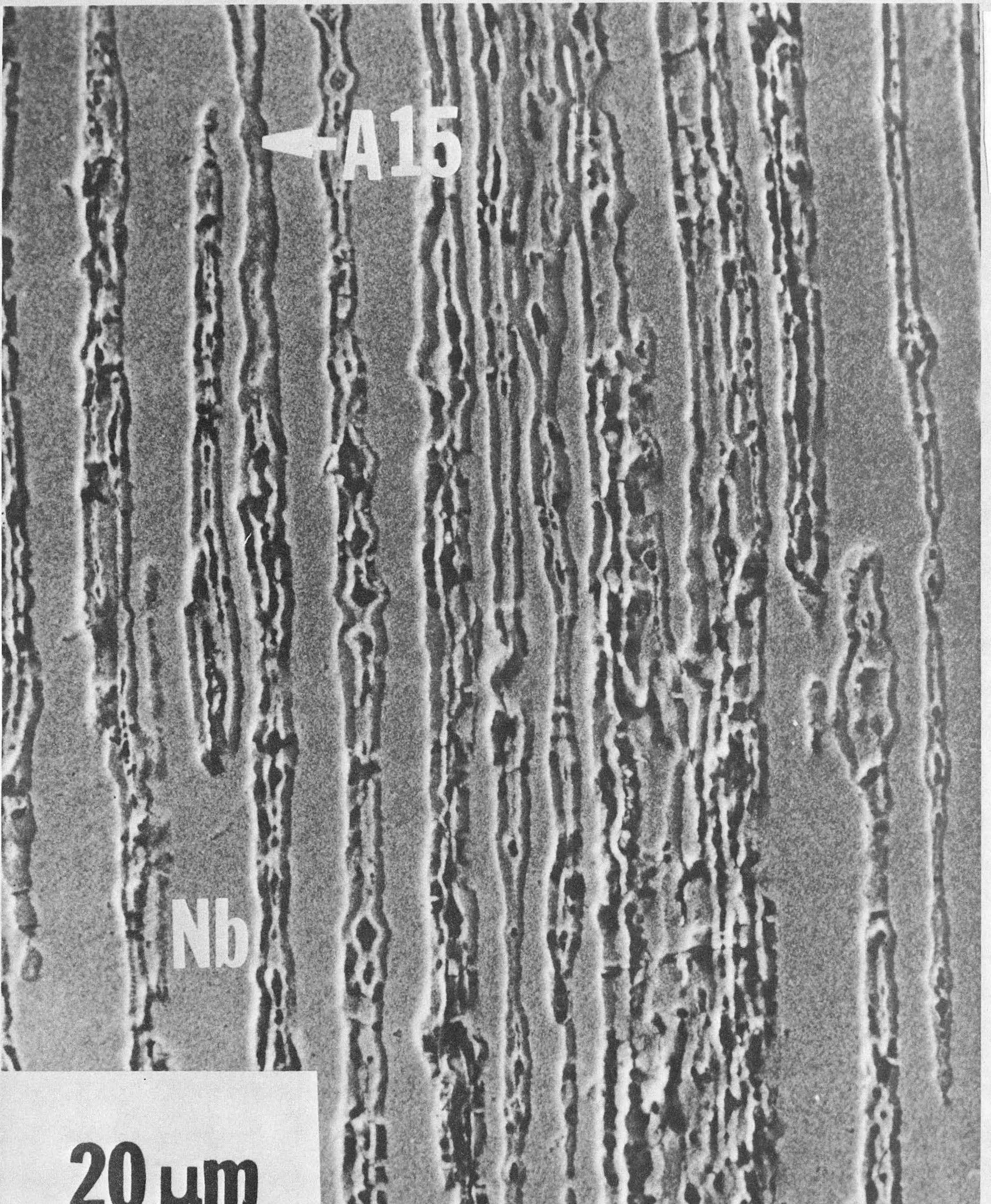

Fig. 2 Scanning electron micrograph of Nb3(Al,Ge) rich phases surrounded by the A15 filaments are also visible

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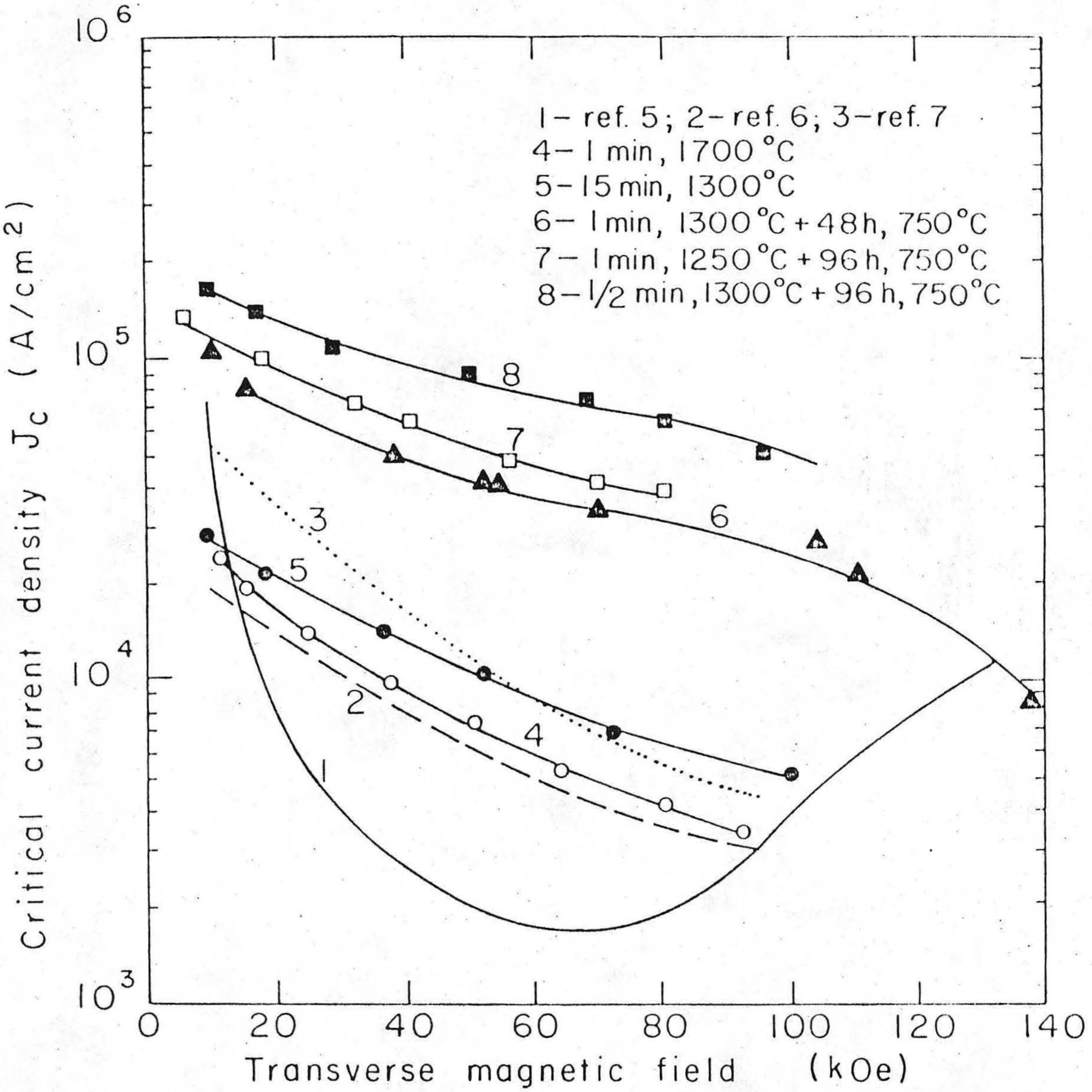

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Fig. 3 Magnetic field dependence of the critical current density for $Nb_3(Al,Ge)$ bulk superconductors.

TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720