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CRITICAL CURRENT MEASUREMENTS OF CVD PREPARED Nb_3Ge CONTAINING VARIOUS AMOUNTS OF SECOND PHASE (Nb_5Ge_3) MATERIAL*

R. V. Carlson, R. J. Bartlett, L. R. Newkirk and F. A. Valencia†

ABSTRACT

Data are presented which show a relationship between the critical current density and second phase (tetragonal Nb_5Ge_3) material in CVD deposited Nb_3Ge for deposition temperatures ranging from 750 to 900°C. The data were measured in the temperature range from T_c (≈ 21 K) down to 13.8 K and at 4 K for several of the samples. Also reported is the slope of the critical current density with respect to temperature, dJ_c/dT. Samples with the largest J_c at 13.8 K also had the largest (dJ_c/dT) and the lowest extrapolated intercept of the J_c curve with the temperature axis. J_c at 13.8 K as a function of second phase has a maximum of 2.5 x 10^6 A/cm^2 for approximately 2 to 3% Nb_5Ge_3 present in the coat. The temperature dependence of the critical current was represented by $J_c(T) = J_c(0)[1-(T/T_c)^2]$ for most samples.

I. INTRODUCTION

CVD produced A-15 Nb_3Ge can have a T_c above 20 K and sustain a large self-field current density (>10^6 A/cm^2) at temperatures as high as 16 K.

The large current densities observed indicate a high concentration of pinning centers much larger than could be expected to exist from grain boundary pinning in the relatively coarse grained (≥ 1 μm) CVD produced Nb_3Ge. It is the purpose here to investigate the possibility that the σ phase (tetragonal Nb_5Ge_3) dispersed in the superconductor gives rise to the high density of pinning centers.

II. EXPERIMENTAL PROCEDURE AND SAMPLE CHARACTERIZATION

The high T_c Nb-Ge samples were prepared using a chemical vapor deposition (CVD) technique described by Newkirk et al.[1] In the work reported here the Nb-Ge was deposited on the inside of 2.2 cm o.d. copper tubes with 0.09-cm thick walls at temperatures ranging from 750 to 900°C. The thickness of the deposited layer varied from sample to sample as well as along the length of each tube, in most cases, ranging between 10 μm and 60 μm. Once prepared the tubes were split lengthwise into two semicircular pieces. One half was used for T_c and x-ray measurements and the other half was used for critical current measurements. The lattice spacing was determined from Debye-Scherrer powder patterns with an accuracy of ± 0.001 Å. The second phase (Nb_5Ge_3) content was determined from x-ray phase analysis using predetermined calibration mixes. The calibration mixes were prepared from arc melted Nb_5Ge_3 and finely ground CVD Nb_3Ge (T_c = 22 K, a_o = 5.140 Å)[2] which had no detectable Nb_5Ge_3 diffraction lines when scanned on the most sensitive diffractometer scale. Samples containing various concentrations of Nb_5Ge_3 were prepared by mixing the appropriate weights of the two constituent powders and then grinding them together in a mortar to ensure thorough mixing. The powder was then applied to a glass slide using a thin layer of grease for bonding. Examination of the powder diffractometer patterns obtained indicated that the ratio of the Nb_5Ge_3 (411) peak to the A-15 (211) peak exhibited the smoothest variation with Nb_5Ge_3 concentration, and this ratio was used as a measure of the amount of Nb_5Ge_3. It is extremely important that the material to be examined be ground to a fine powder, as both Nb_3Ge and Nb_5Ge_3 frequently show a preferred orientation in the as-deposited condition. In addition, the Nb_3Ge used for calibration must be near stoichiometric as well as single phase.

For the critical current measurements the semicircular tubes were soldered into a sample holder with a low melting temperature indium-tin solder. In all cases the soldering temperature was below 150°C. Current was supplied to the sample from a 0-1500 A dc power supply and the voltage drop along the length of the sample was measured with a Keithley Nanovolt amplifier connected to contact probes spaced 1 cm apart along the sample. I_c was defined as that current which produced a voltage drop of 0.1 μV. Measurements were made in the temperature ranges from 13.8 to 21.0 K and 1.5 to 4.0 K. Pumped liquid hydrogen in contact with the specimen was used for the high temperature range while liquid helium was used in the 4.0 K range. An automatic data acquisition system (Hewlett Packard Model 3050A) was used to control the experiment and acquire the current-voltage characteristics. Temperatures were determined by a calibrated germanium resistance thermometer, and the temperature stability during data acquisition was better than 20 mK for the entire current range from 0 to 1500 A.

For the total semicircular sample the superconducting cross section was typically around 10^{-2} cm^2. The cross-sectional area of the samples was reduced by decreasing the width of the samples. After the final critical current measurement the cross-sectional area of the sample was determined from photomicrographs.

III. RESULTS AND DISCUSSION

The critical currents of 25 Nb_3Ge samples with varying amounts of Nb_5Ge_3 were measured as a function of temperature. Figure 1 shows the results for a typical sample over the range from 13.8 K to 20 K. As can be seen there exists a "tail" in the J_c data which we believe to be due to inhomogeneities in the sample material. In most cases the data can be fit using a simple model based on a material composed of constituents with

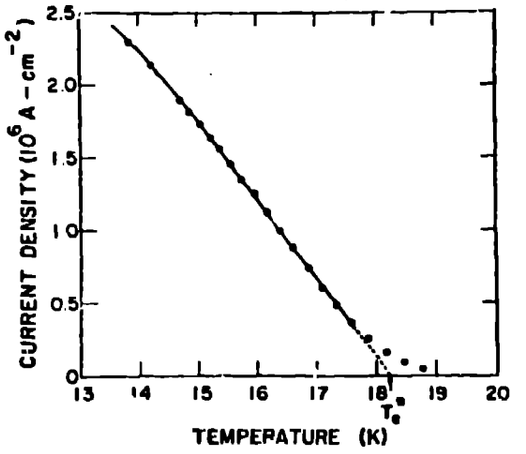

Fig. 1. Temperature dependence of the critical current density J_c. The dashed line shows how the extrapolated temperature T_c* is defined.

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varying T_c's with each constituent following a T^2 temperature dependence.[2] Also shown is a linear extrapolation of the data (disregarding the tail) through the zero current axis, where the intercept is defined as T_c^*. For the samples tested T_c^* varied from about 16 K to 19.7 K. In those samples showing the highest T_c^*'s, the tail was virtually nonexistent; however, for the low T_c^*'s the tail covered a temperature range of up to 4 K. For those samples where the inductive transition temperature was measured, T_c^* was 1.5 to 3.5 K lower than the inductive onset.

Such inhomogeneities should be reflected in broadening of the x-ray lines. The variation of linewidth with diffraction angle θ was examined for 9 of the 25 samples and was found in all cases to vary as tan θ. This functional dependence implies that a distribution of lattice spacings exists about the mean value determined from a Debye-Scherrer powder pattern. Furthermore, the rate at which the linewidth increases with tan θ, $(\Delta\beta/\Delta\tan\theta)$ is a measure of the width of the distribution of lattice spacings. This distribution of lattice spacings reflects compositional inhomogeneities, microstrain, or related effects all of which can cause variations in the superconducting properties. The samples measured indicated that T_c^* is an inverse function of $\Delta\beta/\Delta\tan\theta$, implying that good homogeniety is essential in obtaining high T_c^*'s. In general we have found that the best T_c^*'s are associated with the highest deposition temperatures (up to 900°C); however, the existence of several exceptions to this trend suggests that this effect may be due to sample preparation rather than an intrinsic property of Nb_3Ge.

In Fig. 2, J_c vs T^2 is plotted for two characteristic samples; one deposited at 900°C and having a high T_c^* with little or no "tail" and the other deposited at 850°C exhibiting a distinct "tail". The data for the sample deposited at 900°C shows a T^2 temperature dependence over the temperature range from 4 to 19 K, while the other sample deviated from this behavior. Both the T^2 temperature dependence and the small "tail" suggest that the samples deposited at higher temperatures are more homogeneous. This generally was the case over the total range of deposition temperatures studied. Again this may be associated with problems in sample preparation and not an intrinsic property of Nb_3Ge.

High critical current densities (≈ 2.5×10^6 at 13.8 K and 4.5×10^6 A/cm at 4 K) were observed only in those samples with small amounts of Nb_5Ge_3. In Fig. 3

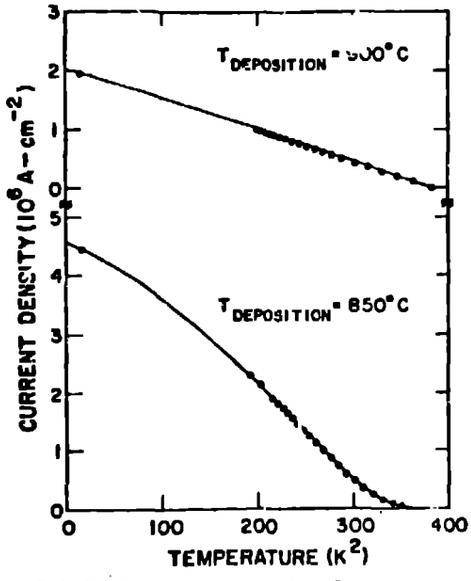

Fig. 2. Critical current density plotted as a function of T^2 for two deposition temperatures.

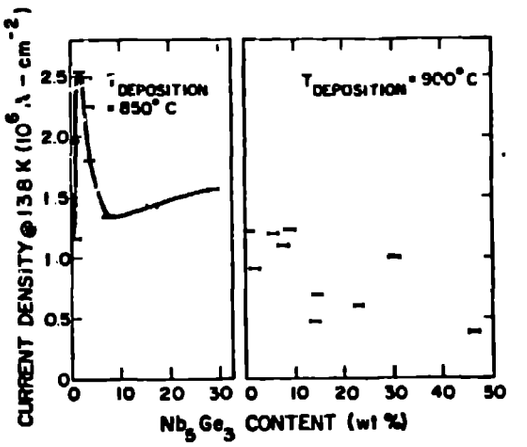

Fig. 3. Critical current density at 13.8 K plotted as a function of wt.% of σ phase (Nb_5Ge_3) for two deposition temperatures. The solid line in the first figure is drawn to suggest the apparent dependence of J_c on Nb_5Ge_3 content.

this dependence of J_c on the σ phase Nb_5Ge_3 is shown for two deposition temperatures. Figure 4 shows the effect of the σ phase content on dJ_c/dT at 13.8 K. For both J_c and dJ_c/dT a relatively sharp peak occurs at ∿ 2.5 wt% Nb_5Ge_3 for the material deposited at 850°C, and a somewhat less pronounced peak at 5 wt% second phase is evident in the material deposited at 900°C. There also seems to be a secondary maximum for much larger amounts of second phase material (≈ 30 wt%).

Distinct correlations between the lattice parameter, a_o, and material properties (such as T_c, T_c^*, J_c, dJ_c/dT) and deposition parameters such as substrate temperature were obscured by the lack of uniform samples. However, the data did exhibit certain trends which may be significant. In particular the lattice parameter was constant for all samples made at the same deposition temperature as long as some second phase material was present. This behavior is shown in Fig. 5 for samples deposited at 850°C. There also seemed to be an inverse relationship between T_c^* and a_o superimposed on the linewidth effect discussed above; however, no direct relationship between a_o and J_c, or dJ_c/dT was found for the limited range of lattice spacings in these samples (5.141-5.145 Å). Although the data shows substantial scatter, there seems to exist an inverse relationship between T_c^* and dJ_c/dT for the materials used in this

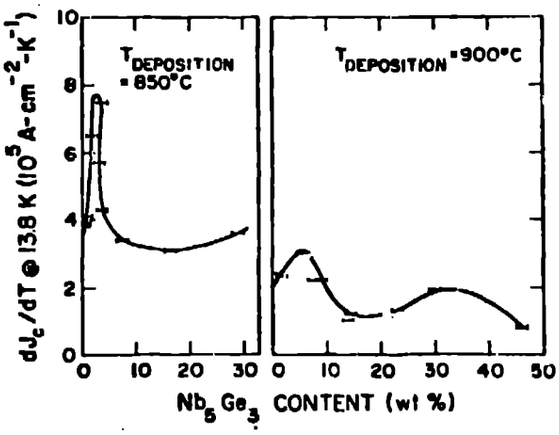

Fig. 4. dJ_c/dT at 13.8 K versus Nb_5Ge_3 content at two deposition temperatures. The solid lines are drawn to suggest the dependence on σ phase.

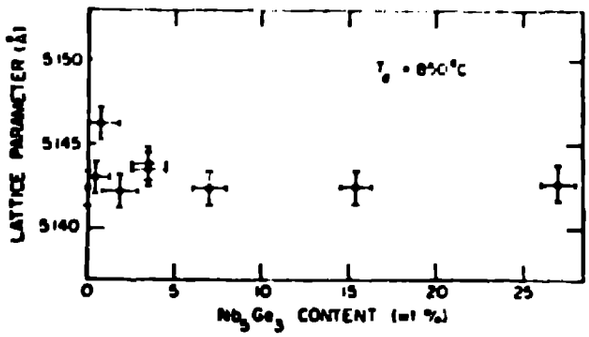

Fig. 5. Lattice parameter as a function of Nb_5Ge_3 content for a deposition temperature of 850°C.

study. This relation would be consistent with the idea that the pinning is increased due to inhomogenieties in the microstructure while these same inhomogenieties are reducing T_c*. If these two parameters, T_c* and dJ_c/dT, are directly related in this manner then it may be necessary to optimize the material for a particular operating temperature. However, we have found that T_c* is independent of Nb_5Ge_3 content over the range considered in these studies (0-30 wt% Nb_5Ge_3) indicating the possibility of preparing material with both a high dJ_c/dT and a high T_c*.

Grain sizes determined metallographically varied from <1 μm to as large as 5-10 μm with no clear relationship to material properties or deposition conditions (over the limited ranges examined). Several of the samples deposited at 900°C early in the study contained substantial amounts of O_2 (up to ~2 wt%); however, the majority of the samples, (including all those deposited at 850°C) were prepared under improved conditions and had low O_2 contents ranging from 0.1 to 0.2 wt% as measured by the platinum fusion technique. No substantial effects were observed as a result of lowering the O_2 to this level.

IV. CONCLUSION

At the present time it is not clear exactly how the second phase material is enhancing the flux line pinning since optical and SEM work does not reveal how the second phase is distributed for samples containing small amounts. However, based on the observation that small amounts of Nb_5Ge_3 substantially increase the flux line pinning but are not visible using ordinary metallographic techniques it seems reasonable to speculate that the second phase material may be finely dispersed throughout the Nb_3Ge grains. In addition, the Nb_3Ge is relatively course grained which also implies that the pinning is due to the precipitated σ phase and that the grain boundaries play a secondary role. Furthermore, it is not clear that the decrease in T_c* associated with the increase in dJ_c/dT represents a relationship intrinsic to Nb_3Ge, and it is expected that further work may result in material with both a high T_c* and dJ_c/dT.

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