SUPERCONDUCTING MATERIALS FOR PARTICLE ACCELERATOR MAGNETS

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INTRODUCTION

The support given to the production of superconducting composites by the needs of particle accelerators has been continuous over about 15 years, and many decisive advances, such as the development of twisted filamentary composites, have been made with this support. At present, interest appears to be focused on two areas: 5 Tesla conductors for present day accelerators such as the Energy Doubler and Isabelle, and 10 Tesla conductors for future very high energy machines. The Fermilab production program for the Energy Doubler conductor has enabled the costs, reliability, and production aspects of 5 Tesla conductors to be extensively explored with conventional Nb 46.5 wt%Ti. Applications at 8-10 Tesla will require Nb-Ti or Nb-Ti-Ta at sub-4.2K temperatures or Nb_3Sn. For both conductors there are present developments which appear capable of pushing current densities to higher values. The choice of superconductors for accelerator use is reviewed in these contexts.[1,2,3]

CHOICE OF MATERIALS

Present accelerator designs are clustered around a field of 5 Tesla (e.g. Fermilab, Brookhaven) with many future studies looking at the 8 - 10 Tesla range. There has also been some recent interest in low field, iron-dominated dipoles in which the superconductor will see a field of about 2 Tesla. The present range of interest is thus rather large but since the upper limit is about 10 Tesla, these demands can be met either by the use of Nb-Ti (or Nb-Ti-Ta) or Nb_3Sn. Both of these conductors are available in multifilamentary (MF) form from industrial sources and are suitable for accelerator magnets. As Table 1 shows, the upper critical field (Hc_2) and transition temperature (T_c) of both types of material cover the foreseeable range of demands for accelerator magnets. We shall briefly comment on some of the design compromises posed by the two materials later. It is appropriate to say here, however, that there is no magic new material on the horizon that is likely to replace Nb-Ti or Nb_3Sn. One class of materials that has a potentially exciting prospect is that of the ternary molybdenum sulphides: these can have an Hc_2 of > 50 T (although T_c is only ~ 14 K) which extends superconductivity into field ranges unattainable with A15 compounds such as Nb_3Sn. However, much development of this material is needed to produce conductors with useful current densities.[4] It does not appear, moreover, that such a material would offer any features not already possessed by Nb-Ti or Nb_3Sn in the field range presently of interest to accelerator designers.

Table 1. T_c and Hc_2 values

Material	T_c	Hc_2(4.2K)	Hc_2(2K)
Nb 46.5 wt% Ti	9.3K	10.7T	14.2
Nb 43 wt% Ti 25 wt% Ta	8.9	10.9	15.5
Nb_3Sn	18	~ 18	20
(Nb 1 wt% Ti)$_3$Sn	~ 17	20-22	22-23

Note that Hc_2 does not have a unique definition for heterogeneous, high J_c materials. Hc_2 is here defined by a current density extrapolation function - see ref. 1 and 2 for a discussion.

The ternary molybdenum sulphides are, like Nb_3Sn and the A15 compounds, brittle. Brittle conductors can be fabricated in multifilamentary (MF) form using various metallurgical tricks, of which the bronze route used for MF Nb_3Sn is the most well-developed.[3] A common feature of all such processes is that the superconducting phase (e.g. Nb_3Sn) is grown only at the final wire size. Growth occurs by a diffusion reaction (e.g. between Cu-Sn bronze and Nb) at elevated temperatures (typically 700°C). Since the breaking strains of the brittle superconducting phase are small (0.5 - 1%), only limited winding, thermal and electro-magnetic stresses can be applied to the wire and special techniques of insulation, fixturing and handling must be applied in order to control the applied strains. These techniques have been well developed for small magnets[5] and are presently being tested for model accelerator dipoles.[6]

CRITICAL CURRENT DENSITIES OF PRESENTLY AVAILABLE CONDUCTORS

Measurements

Before discussing reported values of current densities, it is necessary to make a few remarks on the subject of critical current (density) measurement. It is unfortunately true that many measurements made by different laboratories are not directly comparable. The reason for this is that most measurements are still made with only short lengths (~ 5 cm or less) exposed to the transverse field region of the test magnet and these short lengths do not permit the details of the resistive transition to be determined. A frequently chosen criterion of I_c is 1 μV/cm, a value which places the transition point well up on the internal heating-external cooling characteristic of the conductor. Since the external cooling characteristics of different laboratories' test fixtures can vary rather strongly, it is not surprising that the reported critical current can vary quite markedly from laboratory to laboratory.

A second defect of this kind of test is that it defines the I_c at a power dissipation level which is substantially above that of the quench of high current density magnets. The effective quench resistivity of such magnets has frequently been shown to be on the order of 10^{-14} Ωm.[7,8] Measurements of I_c at 10^{-14} Ωm are not at all difficult for wires of ~ 1 mm diameter or less. For example, a 60 cm length of Fermilab strand (0.67 mm dia) can be wound as five turns on a 35 - 40 mm dia barrel for insertion into a modestly sized 50 mm dia solenoid. Such a sample will enable the I_c to be determined over the range ~ 10^{-15} - 10^{-12} Ωm (unless a sample quench intervenes), giving the full details of the resistive transition. Results drawn from a recent study of the Fermilab conductors show that if the 10^{-14} Ωm, 5 T I_c is ~ 245 A, then the 2×10^{-13} Ωm hairpin I_c is ~ 275 A and the quench ~

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310 A.[10] Variations of 12.5 - 25% in the reported I_c are thus shown to be possible even at ~ 0.5 Hc_2.

The barrel sample is still a good technical choice for such conductors, although the large winding radii and a lack of a large bore solenoid may dictate the use of straight samples in split solenoids with rather small constant field regions. If the latter type of test is used, it is to be expected that the measured I_c or J_c will be an overestimate of the 10^{-14} Ωm current density.

The preceding discussion has assumed that 10^{-14} Ωm marks the "true" I_c of a conductor. The justification for this is essentially empirical. Barrel sample measurements can conveniently measure at this level and high current density, poorly-cooled magnets quench at about this resistivity. It also appears that well-made conductors have resistive transitions which begin close to the 10^{-14} Ωm point. Although the choice of 10^{-14} Ωm as the criterion for I_c is thus somewhat arbitrary, it does appear to be empirically justified and all figures quoted in this paper will use this criterion, unless otherwise stated. It is unfortunate that the current draft ASTM standard is 1 μV/cm: fuller details are available from a recent NBS study.[11]

Current Densities in Niobium-Titanium Conductors

The principal example of Nb-Ti conductor is the Fermilab conductor for the Energy Saver/Doubler, over 800 such billets having been made.[2] Although high current density was never a major aim of this conductor, significant improvements in J_c have occurred, the J_c (5 T, 4.2 K) being raised from ~ 1800 to 2200 A/mm^2. Good manufacturing experience has been obtained with this conductor and its costs and cabling are well understood. An extensive study of the product variability of the conductor has recently been presented by Tannenbaum et al.[10] The effects of heat treatment and microstructure on the properties of near final size conductor have also been extensively studied in our laboratory.[12,13]

The Fermilab conductors have all used Nb 46.5 wt% Ti, which has become the de facto U.S. specification. However, it has been known for some time that multiply-heat-treated Nb 50 wt% Ti could have significantly improved J_c values of 2500 - 2600 A/mm^2 (5 T, 4.2 K) being possible.[27] Recent studies of ours have shown that standard Nb 46.5 wt% Ti can also give these values too.[24] Since this is a complex topic, however, the reader is referred to recent reviews for fuller discussions of the effect of composition and heat treatment on J_c.[1,2] Although some workers from Krupp[14] have reported values of J_c exceeding 3000 A/mm^2 (5 T, 4.2 K) in Nb 50 wt% Ti, such values have been considered inflated due to the measurement criterion used. A rather recent study by Li Cheng-ren et al. reports values of up to 4000 A/mm^2 (5 T, 4.2 K).[15] Measurements on one sample in our laboratory confirm a value of 3450 A/mm^2 (5 T, 4.2 K), a J_c which is a truly significant step forward in the further optimization of Nb-Ti.

A compendium of some interesting recent results is shown in Fig. 1. Three sets of results are shown in the graph. The first is a recent example of a commercially produced (by IMI) Nb 46.5 wt% Ti conductor of high J_c (2600 A/mm^2 at 5 T, 4.2 K and 1640 A/mm^2 at 10 T, 1.8 K). These current densities are fully equal to the best Nb 50 wt% Ti conductors in regular production. The very high J_c Nb 50 wt% Ti conductor (J_c 5 T, 4.2 K of 3450 A/mm^2) falls off at high fields due to Ti depletion from the matrix which reduces Hc_2. The third alloy shown is a developmental Nb-Ti-Ta alloy fabricated in our laboratory in collaboration with A. D. McInturff, for which more information is given in the next and later sections. Fig. 1 contains results representative of the best J_c values currently obtainable. Additional information will be found

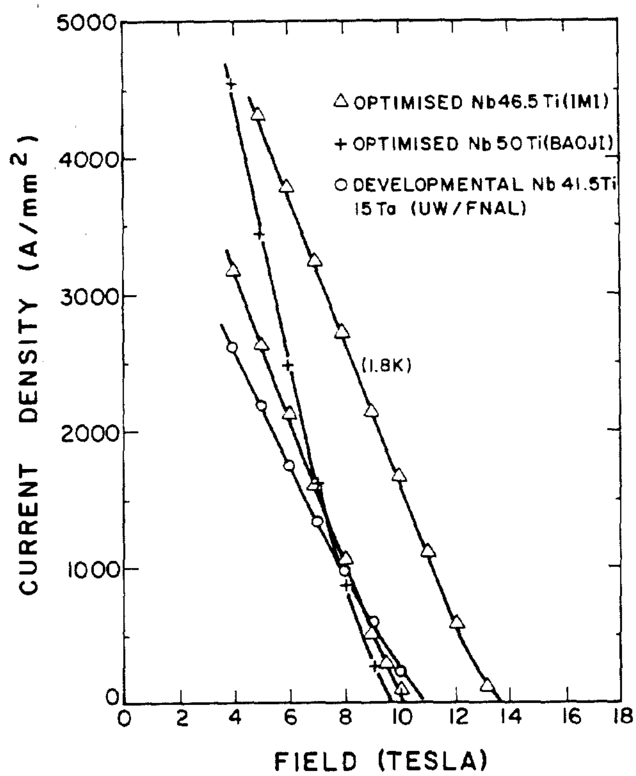

Fig. 1. J_c (10^{-14} Ωm) results on some recent high J_c conductors. Values are at 4.2 K except where noted.

elsewhere.[1] The figure shows how sensitive the high field J_c is to Hc_2, this fact being responsible for the interest in using Nb-Ti at 1.8 K or 2 K where Hc_2 is significantly increased (Table 1).

Current Densities in Niobium-Titanium-Tantalum

The reasons for interest in alloying additions of Ta are because of the higher Hc_2 conferred by the Ta additions.[1,9] An extensive investigation in our laboratory of the effect of Ta, Hf, Zr and V additions has shown that only Ta produces significant enhancements in Hc_2, the effect being maximum at about ε/a 4.37 for alloys in the vicinity of Nb 43 wt% Ti 25 wt% Ta.[17] The enchancement is small at 4.2 K (~ 0.3 T) but reaches 1.3 T at 2 K, where Hc_2 is raised from ~ 14.2 T (Nb 46.5 wt% Ti) to ~ 15.5 T. However, since Ta is expensive the most cost-effective addition may occur at lower values of alloy content.

The first magnet use of the Nb Ti Ta conductor was for the GA 12 Tesla Fusion Test coil, which has just been wound and is awaiting test.[18] Conductor current densities were disappointing (~ 1000 A/mm^2 at 10 T, 1.8 K) due to filament sausaging produced by compositional inhomogeneities.[19] Japanese remakes of this alloy have been promising[20] (J_c ~ 1400 A/mm^2 at 10 T, 1.8 K), although the criterion used for I_c (5 μV over 2.5 cm) is sufficiently high to make comparison to 10^{-14} Ωm difficult. In collaboration with A. D. McInturff of Fermilab, we have recently fabricated a 361 filament Nb 41.5 wt% Ti 15 wt% Ta conductor with J_c properties of 2200 A/mm2 (5T, 4.2K), 1320 A/mm2 (10

T, 2.25 K) and an extrapolated 10 T, 1.8 K J_c of 1600 A/mm^2. This thus already matches the very best binary NbTi alloy at 10 T, 1.8 K. Further adjustments of processing should permit further significant increase in J_c.

Current Densities in Nb_3Sn Conductors

The true current densities of Nb_3Sn conductors are considerably more uncertain than those of Nb-Ti, due to the greater breadth of the resistive transitions: differences of 20-30% in J_c may not be at all significant, even for undamaged samples. Two examples of the J_c (the J_c is normalized to the original bronze + Nb cross-section) obtained from well-optimized conductors in long sample measurements are shown in Fig. 2. The first is from recent studies of conventionally produced bronze route conductors produced by Airco[22] (the criterion used here was not 10^{-14} Ωm but 3 μV over 350 mm which is believed to be close to 10^{-14} Ωm) and the second from studies of jelly roll conductors produced by Wah Chang.[23] Both conductors gave very similar J_c values of 800-900 A/mm^2 at 10 T, 4.2 K.

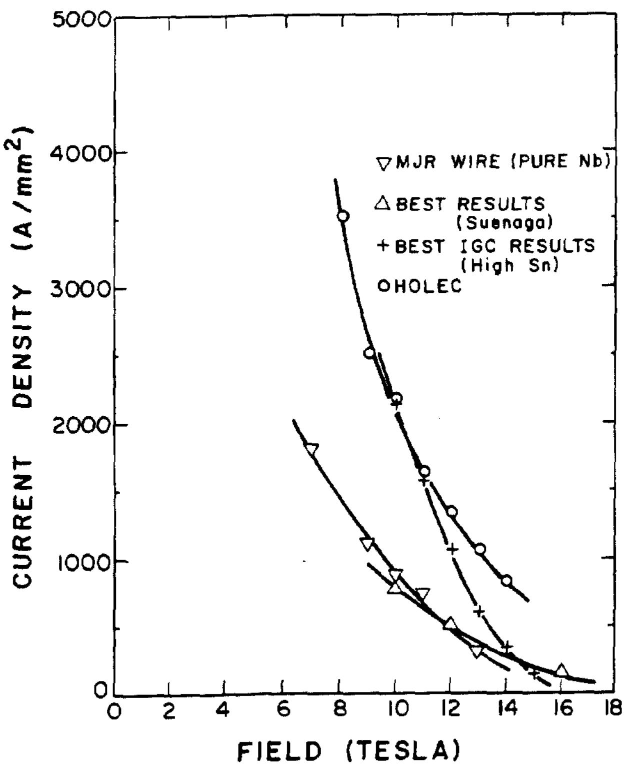

Fig. 2. J_c results on some recent high J_c Nb_3Sn conductors. J_c values are at various criteria (see text). Data is included from references 22, 23, 32, 33. The lower two curves are for bronze route conductors, the upper for high Sn process material.

Significant possibilities exist for making further important increases in critical current density by utilizing processes which use tin-rich phases, as will be discussed in a later section.

NEW DEVELOPMENTS

A major reason for optimism that there are significant developments possible is that, after a decade or more of essentially empirical development of J_c, there is now appearing the outline of a quantitative microstructural description of well optimized conductors which appears capable of forming the basis of a full scientific understanding. This is a complex problem, both for Nb-Ti and Nb_3Sn, since the critical current densities are determined by the interaction of the fluxoid lattice and the microstructure on a scale of approximately 5 - 20 nm.[2]

Niobium-Titanium

A major recent advance, provided by the electron microscopy of Anne West in our laboratory, has been to determine the morphology of the precipitates responsible for high J_c.[24] An example of these in high J_c Nb 50 wt% Ti alloy is shown in Fig. 3. The plate morphology of these precipitates, which are approximately 3 nm thick by 20 to 50 nm wide, is common to all of the high J_c conductors so far examined by us. Our recent investigations have shown an approximately inverse relationship between the plate separation and J_c. When the plate separation becomes less than about 20 nm, however the J_c begins to decline, presumably due to the proximity effect.

Fig. 3. α-Ti precipitates in a transverse section of an optimized Nb 50 wt% Ti composite (J_c 5 T, 4.2 K = 2610 A/mm^2). Courtesy of Anne West.

A crucial additional observation has been that significant local composition gradients (up to about 10 wt% Ti) can exist in Nb-Ti alloys.[19] These gradients have their origins in the coring of the alloy as it freezes. It is evident that the microstructure of Fig. 3 is a rather regular one. This requires uniform precipitate nucleation, which in turn requires an alloy of uniform composition capable of permitting the development of a regular sub-band structure. Our conclusions are that compositional inhomogeneities perturb the sub-band structure, as well as causing irregular nucleation of precipitates.[19] Both effects degrade the J_c and tend to produce large α-Ti precipitates which produce cabling and drawing breaks.[24] In cases where the segregation is particularly large, gross filament sausaging can occur too.[24]

Niobium-Tin

A major variable in Nb-Sn is undoubtedly that of prereaction. Under adverse circumstances, grossly irregular filaments result. The filament J_c is then significantly degraded due to the locally reduced cross-section (Fig. 4). Recently reported results of ours have shown that over-annealing near final size (when the filaments are small) can be very deleterious.[21] As few as four anneals for 1 hr at 550°C (rather than 1 hr at 450°C) were responsible for halving the current density, due to the production of irregular Nb_3Sn layers. Fortunately the solution to

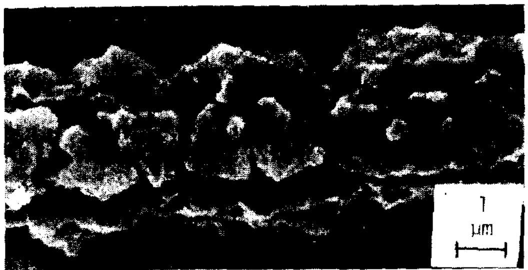

Fig. 4. Reacted Nb_3Sn filament from a composite having initially strong pre-reaction. The uneven cross-section is very evident. Courtesy of David Smathers.

this problem is straightforward, namely to reduce the in-process anneals to as low a temperature as possible (450 - 475°C). An encouraging new area of study is that of the local chemical composition of the Nb_3Sn. It is now possible to study the concentration gradients within the filaments using both Auger and electron microscopy.[25,26] Measurements by David Smathers in our laboratory show that local gradients can be both variable and significant and that it is difficult to react conductors to a uniform composition. All conductors as normally reacted have a significant spectrum of T_c, Hc_2 and J_c, thus making it difficult to interpret the globally averaged values[2,30] resulting from the measurements. Such local measurements will be needed in order to properly understand the effects of different conductor makeup parameters and heat treatments: both are areas which presently appear to be closer to black magic than to scientific understanding.

Higher Critical Current Densities

The recent work by Li Cheng-ren showing that very high J_c values can be obtained in binary Nb-Ti alloys is extremely important. Our investigations of the microstructure show that the high J_c is not obtained with a higher density of precipitates (the spacing is in fact about 32 nm, considerably above the point at which the critical current density peaks in the previous alloys examined by us) but by thickening of the precipitates.[31] Now that we have a tool to observe the microstructure, it becomes possible to design treatments to produce the desired microstructure. Without such microstructural information, optimization can only be empirical; it is interesting to note that the microstructure that we observe in all high J_c materials is quite different from the "string-of-pearls" morphology that had been assumed for these materials.[14,27]

In considering the advances in J_c possible for Nb-Ti, Nb-Ti-Ta and Nb_3Sn we do not have any good theory to guide us. There appears to be no good flux pinning theory which can treat the dense, strong pinning situation found in the well-optimized conductors. Proceeding by progressive refinement of the microstructure appears to be a promising method[28] for Nb-Ti(-Ta). As we have discussed elsewhere,[1,28] the high J_c shows no saturation (in contradiction to the predictions of Kramer's often used theory[29]).[15] The significance of the very high J_c Baoji material[15] is that it raises the level of microstructural flux pinning still further. There seems to be no reason why the same level of microstructural pinning should not be put into Nb-Ti-Ta alloys too. So far as the 10 T, 1.8 K J_c of Nb-Ti(-Ta) is concerned, the J_c is determined first by the Hc_2 and second by the degree of optimization of the microstructure. With respect to the Hc_2, it is immediately apparent from Fig. 1 that the Baoji Nb 50 Ti is not optimum, its Hc_2 being about 9.5 T, as opposed to the 10.5 T of the Nb 46.5 Ti. We have argued elsewhere[1] that the sequence of heat treatment-draw cycles is more important than the base alloy composition in determining the flux pinning. However, the Hc_2 is determined by the residual matrix composition, after the Ti rich precipitation has taken place. We have used the above approach in order to estimate the potential of Nb-Ti and Nb-Ti-Ta, assuming that (i) the matrix composition is adjusted to that of optimum Hc_2 after precipitation and (ii) that the same degree of flux pinning can be achieved in the Nb-Ti and Nb-Ti-Ta. We have explained this procedure in detail elsewhere:[9] for high field use, the procedure requires only lines drawn parallel to the high field portion of the Baoji (2.21 K) pinning force (J_c x B) vs. field curve shown in Fig. 5. The two parallel lines drawn correspond to the peak Hc_2 values exhibited by binary Nb-Ti (~ Nb 46.5 wt% Ti) and ternary Nb-Ti-Ta alloys (~ Nb 43 wt% Ti 25 wt% Ta). The 10 T, 1.8 K pinning forces obtained are 19 and 25 GN/m^3, which correspond to 10 T, 1.8 K J_c values of 1900 and 2500 A/mm^2. These values are not to be interpreted as limits: further improvements may follow from better microstructural control.

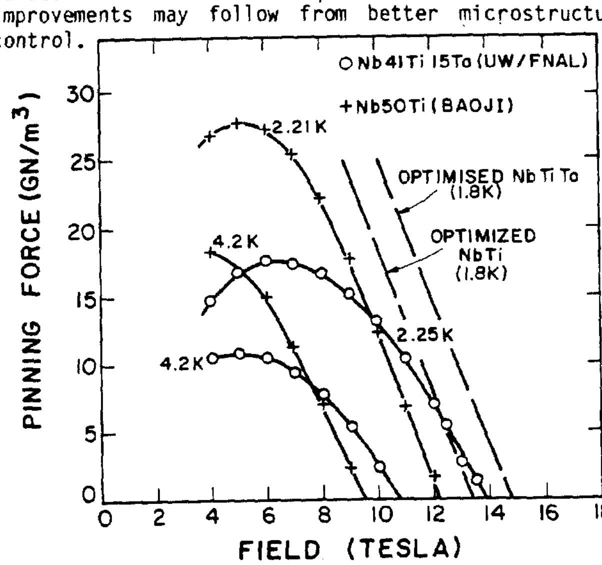

Fig. 5. Pinning force (J_c x B) curves for Nb-Ti and Nb-Ti-Ta conductors (see also Fig. 2). The dashed lines are scaled from the 2.21 K high field slope of the Baoji conductor, using the maximum Hc_2 values expected from binary and ternary alloys at 1.8 K (see text).

The prospects for increases in the J_c of bronze route Nb_3Sn are less clear. We know little about the flux pinning interaction in grain boundaries of Nb_3Sn and many aspects of composite design are unclear. It seems likely on purely general grounds that further improvements are possible, particularly now that the deleterious effects of pre-reaction are understood.[21] Bronze carries Sn rather inefficiently, since the maximum solubility of Sn in Cu is ~ 14 wt% and the depleted bronze left after reaction is too resistive to be an efficient stabilizer or quench protection for the coil. An obvious way to increase the J_c (normalized to the total package of Nb and Sn carrier necessary to produce the Nb_3Sn) is to provide a more efficient Sn carrier.[3] Three recent examples of MF conductors are the composites produced by HOLEC[32], IGC[33] and Wah Chang.[34] The HOLEC process uses Nb tubes filled with $NbSn_2$ and a little Cu powder, while the other two processes use internal Sn (perhaps with a little Cu) contained within a Cu-Nb composite. A cross-section of an IGC composite made is this way is shown in Fig. 6.

The very beneficial effect which these high Sn processes have on the overall J_c is shown in Fig. 2. Although a variety of J_c criteria are used for the high Sn conductors, it is clear that the processes appear to offer considerable prospects, since J_c values of ~ 2000 A/mm^2 at 10 T, 4.2 K were reported. Present evaluation of the reproducibility of long lengths and J_c evaluation at high sensitivity will yield valuable information on the magnet capability of these new processes.

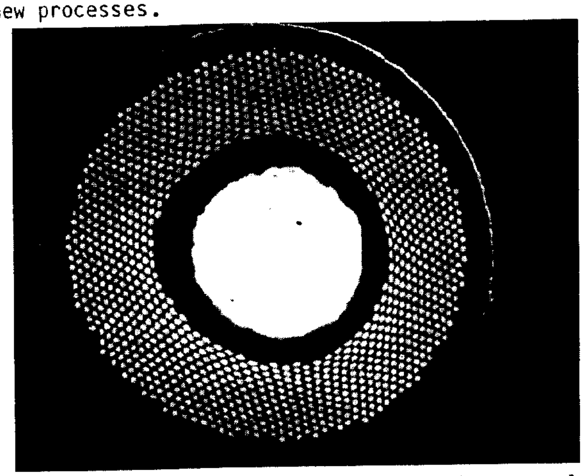

Fig. 7 Tin-core conductor made by IGC. The central core is Sn, the outer section containing Nb filaments embedded in copper. (Courtesy of R. E. Schwall.)

Summary and Conclusions

Critical current densities of Nb-Ti, Nb-Ti-Ta and Nb_3Sn conductors have shown significant recent increases. High J_c is available both from Nb 46.5 wt% Ti and Nb 50 wt% Ti. Recent conductors made at Baoji have particularly high J_c at 5 T, 4.2 K, although not optimized for 10 T, 1.8 K use. Present values of J_c 10 T, 1.8 K in Nb-Ti exceed 1600 A/mm^2 (10^{-14} Ωm) and values of 1900 A/mm^2 appear feasible. The higher Hc_2 alloys of Nb-Ti-Ta appear capable of 10 T, 1.8 K J_c values of 2500 A/mm^2.

The J_c of Nb_3Sn conductors is less well-established. Values of ~ 900 A/mm^2 (10 T, 4.2 K, 10^{-14} Ωm) are presently available from clean, well-optimized bronze route conductors. New high-Sn processes offer promise of J_c values approximately twice these. Few measurements of 10 T, 1.8 K J_c have been made for Nb_3Sn: increases of ~ 15-20% over the 10 T, 4.2 K values seem possible.

A scientific understanding of the microstructures of both Nb-Ti and Nb_3Sn composites is being developed. Good prospects exist for further developments in J_c as we acquire an understanding of the fluxoid-microstructure interaction in these high Hc_2 materials.

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