

NEW DEVELOPMENTS IN NIOBIUM TITANIUM SUPERCONDUCTORS

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After a very active period of Niobium Titanium superconductor strand development in the mid 1980s, progress slowed as the SSC scaled up for production and emphasis moved towards guaranteeing, rather than enhancing performance. However, this period also produced several advances in the understanding of flux pinning in Niobium Titanium alloys and in the fabrication of composites containing artificial pinning centers (APC). This paper reviews some of this progress and attempts to assess its implications for future accelerator conductors.

I. STATE OF THE ART

A. Overview of the development of properties with time

The High Energy Physics (HEP) accelerator community has been the most consistent and effective driving force behind the major advances that useful superconductors have achieved in the last 30 years. The principal material advanced is Niobium Titanium of composition Nb-47wt.%Ti. There are at least three major reasons why the HEP community has played such a large role in superconducting wire development: One is that the accelerator community knows that increased magnet performance is always driven by an ever increasing critical current density (J_c), two is that the broader HEP community has stuck with superconductor development, even when there was not an immediate big accelerator project and three that there has been a commitment from key figures in the community to encourage productive links between the basic science of flux pinning in superconductors, the accelerator magnet community and the superconducting industry. In a series of workshops sponsored by the US Department of Energy, Division of High Energy Physics, held at least annually from 1983 until the present day, a community of US researchers, users and manufacturers have catalyzed each other in a continued attempt to wrest more performance from Nb-Ti conductors. Figure 1 collects information on some of the impressive advances in performance of material. The early large scale US procurements of conductors for Mirror Fusion Test Facility (MFTF) and for the Fermilab Tevatron in about 1980 developed the industry. Production of hundreds of billets generated genuine production statistics and also allowed researchers like ourselves to ask questions about which fabrication variables were really important in controlling their then rather variable properties. The Fermilab J_c (5T, 4.2 K) specification of 1800 A/mm² was less a carefully thought out attempt to determine what the material was capable of than an attempt to challenge production to a consistent, deliverable level. Its

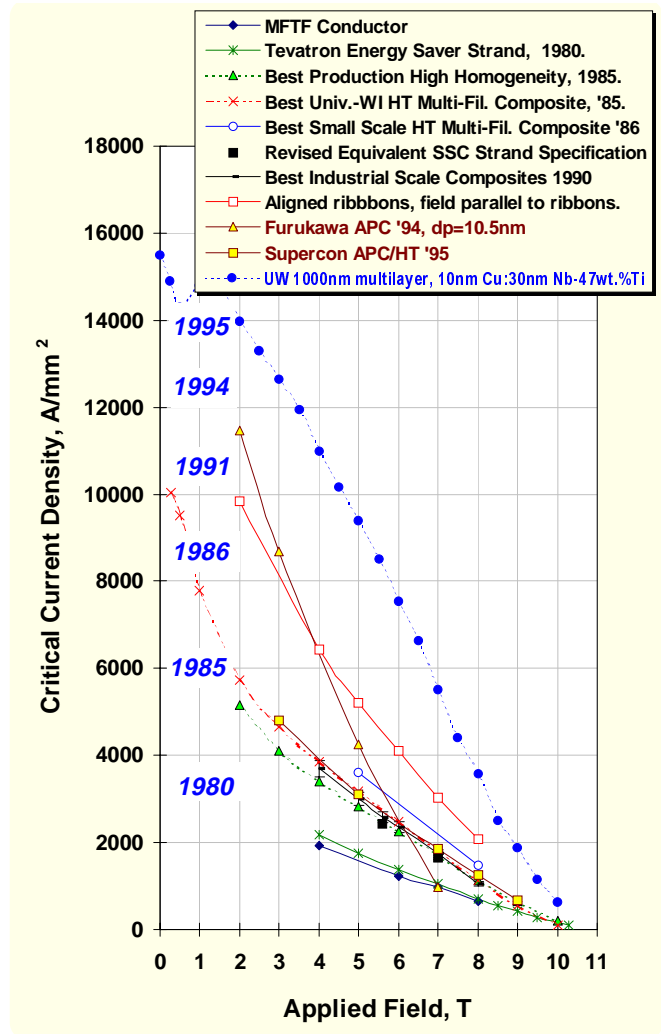


Figure 1 Advances in the Critical Current Density of Nb-Ti Based Superconductors. All data are at 4.2K. Key: MultiFil = Multifilamentary strand, HT = Precipitation Heat Treated, APC = Artificial Pinning Center (aligned ribbons ref. [1], Furukawa APC ref. [2], Supercon APC/HT ref.[3]).

realization required simultaneous work on the raw alloy, the copper, the extrusion process, large rod drawing and fine wire drawing and the heat treatment capabilities of a still small industry. Without a firm idea of what the empirical optimization processes employed by industry really were doing to the microstructure and thus to the flux pinning which controls J_c , it is not surprising that there was eventually a considerable extra margin of performance to be found in the material, the exploitation of which in the mid to late 1980s led to the SSC benchmark, now 50% larger, of about 2750 A/mm², while at the same time diminishing the filament size from ~10 to 6 μ m.

Underpinning the big jump from 1980 to 1985 was the discovery of the large role played by uncontrolled microscale chemical inhomogeneities in the raw Nb-Ti alloy[4]. When, with a great deal of help from collaborations with R. M Scanlan (LBL) and W. K. McDonald at Teledyne Wah Chang Albany (TWC) [5], the first high homogeneity Nb-Ti alloy was delivered, it became almost immediately possible to get a 50% increase in performance in an industrial scale billet. This improvement was based on a sound understanding of the factors controlling the microstructure [6,7,8]. Further exploitation of this understanding on a laboratory scale led to large filament conductors which could achieve ~ 3700 A/mm², although with larger filaments[9]. Since 1985-1986, engineering the very best laboratory scale properties into very long length (several km), very fine filament (6 μ m or even lesser diameters) cabled conductors has been the principal emphasis of the industry. This was triumphantly done in the latter stages of the SSC-CDG R&D program in a collaboration in which groups from the principal US industrial suppliers (IGC, OST, Supercon and TWC) worked together with teams at principally BNL, Fermilab, LBL and the University of Wisconsin to characterize, understand and to devise engineering solutions that stood in the way of realizing the scientific control of microstructure that underpinned the industrial fabrication process (these results are reviewed in greater detail elsewhere[10]).

Two recent benchmarks significantly lift the potential of Nb-Ti alloys. One was a proof-of principle experiment conducted in our group [1], aimed at answering the question of whether Nb-Ti had run out of capacity to develop any higher J_c . In this case we took an already very high J_c sample and rolled it to a high aspect ratio so that the pinning microstruc-

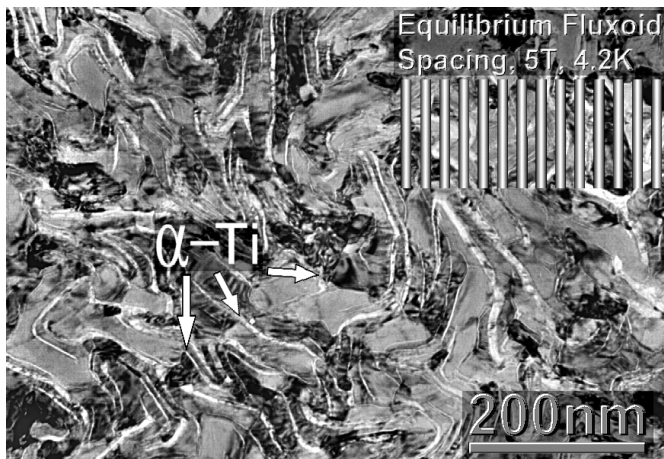


Figure 2 Typical Nb-47wt.%Ti high J_c , round wire microstructure (in transverse cross-section) showing the densely folded sheets of α -Ti pinning centers dispersed within the superconducting β -Nb-Ti matrix. Superimposed is a schematic illustration of the equilibrium fluxoid spacing and dimensions appropriate to Nb-47wt.%Ti at 5T, 4.2K.

ture (Figure 2) was highly aligned for one axis of the wire. J_c (5T) was raised from about 3500 to 5300 A/mm² without any degradation at higher fields. This proved that there was a greater potential in the material. The second aspect studied after 1985 was the question of whether a better array of pinning centers could be incorporated into Nb-Ti than was supplied by nature, even in the highly optimized process that had evolved in the early 1980s. Artificial pinning center (APC) arrays were first introduced by Dorofeev et al. [14] at the 1985 Magnet Technology Conference, just as the conventional process was reaching its full understanding. The best realization of this concept is by Matsumoto et al. [2] from Furukawa Electric Co., as indicated by the 1994 plot in Figure 1. This composite was the first and is still the only round wire to have broken the 4000 A/mm² (5T) barrier. This result is all the more impressive in that the wire clearly lacks a competitive high field performance (it crosses over with standard wires at ~ 6 T), as we discuss later in this paper. In fact recent APC multilayers [11] and [12] have shown that highly aligned pinning layers can produce $>8,000$ A/mm², first at a similar expense to the high field J_c [11] and now without any compromise for the high field properties [12].

In the rest of this paper we briefly review some of the implications of these recent advances for new accelerator conductors, first briefly defining the difference between conventional and APC conductors.

B. Conventionally Processed Material

Virtually all material made up until now has been conventionally processed, where the term conventional means that the starting point is a Nb-Ti alloy which is sheathed in Cu, and after various compaction, extrusion and drawing processes is then heat treated multiple times within the $\alpha + \beta$ two phase field so as to produce a fine dispersion of α -Ti precipitates. These α -Ti precipitates (Figure 2) are the dominant flux pinning centers in conventional wire and their optimization is the goal of the complex fabrication procedures through which Nb-Ti composites are manufactured. Advanced accelerator strands optimized using such a process can today be expected to have critical current densities of the order of 2750-3000A/mm² at 5T, 4.2K and 1100-1400A/mm² at 8T, 4.2K and to consist of 1000-4000 6-10 μ m filaments of Nb-47wt.%Ti alloy, each clad in a thin Nb diffusion barrier and assembled within a Cu matrix for stability (Figure 3). Inherent limitations of the conventional process are that α -Ti is the pinning center and that the quantity of pinning center is limited to 20-25 vol. % for Nb-47Ti by the thermodynamic phase diagram of the Nb-Ti system. The shape of the pins is constrained to the ribbon-shaped pin morphology of Figure 2 by the plane strain deformation dynamics of the body centered cubic β -Nb-Ti alloy. It is an amazing gift from nature that it is possible to produce two phase nanometer scale microstruc-

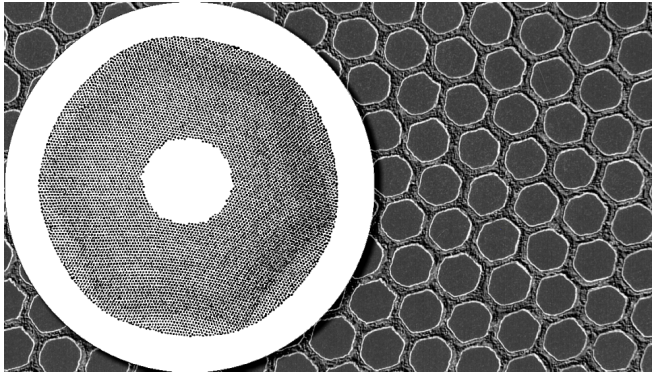


Figure 3 Cross-section of SSC-style multifilamentary superconducting strand at 26mm diameter superimposed on detail showing filaments and their diffusion barriers (fabricated by IGC as part of the SSC-CDG R&D program).

tures such as that seen in Figure 2 in complex many thousand filament composites such as that shown in Figure 3 and to have the piece length be several km. No wonder that competition with standard process Nb-Ti is so hard!

C. Artificial Pinning Centers (APC)

An alternative approach, first used by Dorofeev et al. [14], is to mechanically assemble the desired pinning structure in a composite at large size and then to reduce the structure to the dimensions of the fluxoid lattice by multiple extrusions and wire drawing. Such a process has great conceptual flexibility in the selection of pin, superconducting matrix, and pinning center geometry. One realization of such a composite is shown in Figure 4. The figure shows an important and common attribute of APC composites, namely that the final size nanostructure is not the idealized macrostructure that was assembled. In fact the final size nanostructure of APC composites is seldom explicitly studied and this is certainly one of the key drawbacks to much recent work on trying to understand why disappointing performance has so far been more common than outstanding performance. With these two concepts in mind we now turn to a wider discussion of the potential that the two processes offer for accelerator conductors.

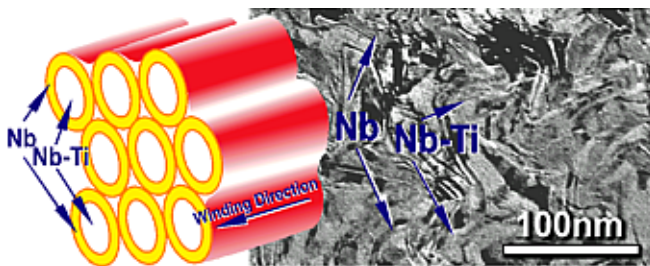


Figure 4 TEM image of original IGC APC at final size (in transverse cross-section) compared with the original "flux tube" design from the patent of Zeitlin et al. [13].

II. OUTSTANDING ISSUES FOR FUTURE DEVELOPMENT

Three issues underlie the development of higher J_c : The first is that of understanding the flux pinning mechanism, the second is ensuring that the upper critical field H_{c2} is not degraded by the choice of pinning center and the third is making composites that are fabricable in useful lengths at appropriate cost. We shall aim our discussion towards these 3 points.

A. Flux-pinning.

The keys to our present understanding of flux-pinning came from careful correlation of superconducting property measurements to a quantitative microstructural description. This turns out to be particularly necessary for understanding the APC composites, many of which do not have well-understood nanostructures [15]. The Volume Flux Pinning Force, F_p ($F_p = J_c \times B$) is a particularly valuable optimization parameter. The microstructural and flux pinning basis of present optimization was described in the studies of Meingast, Lee and Larbalestier [16]. There are systematic changes in the shape of F_p as the wire and its precipitates are drawn to different sizes, as is shown in Figure 5. Two important issues of great practical relevance are that drawing the wire to its optimum size raises F_p at all fields and that the maximum in F_p occurs at about $0.5H_{c2}$. This behavior is unlike that of APC Nb-Ti and almost all other high field superconductors. Having F_p peak at high fields is the reason why Nb-Ti magnets are effective so close to their upper critical field. Two other very important results [17] are that F_p and J_c are linearly dependent on the volume percentage of pinning material and that the H_{c2} of the optimized composite is the same as that of the homogeneous starting alloy. This result was extremely surprising when first obtained because the matrix of the superconductor is strongly depleted of Ti by the formation of about 20 vol.% of α -Ti. It occurs because the optimum size precipitates (Figure 2) are thinner than the superconducting coherence length, ξ . Thus there is a remixing of matrix and pin so far as T_c and H_{c2} are concerned. The enormously beneficial surprise of this result is that this remixing does not in any way lose the heterogeneous nanostructure required for optimum flux pinning. This principle has further important implications for the optimization of ternary Nb-Ti-Ta compositions and APC composites.

The question posed at the start of this section is whether flux pinning is well understood. At the conclusion of [16/21], the answer appeared to be a qualified yes. The microstructure and the flux pinning force had been directly correlated for the very highest J_c composite over more than an order of magnitude change in pin dimension and F_p . By correlating these two it was possible to deduce the thickness dependence of the elementary pinning force and to compare this to calculations

of the vortex core interaction with the pin. These calculations correctly predicted the observed linear dependence of the elementary pinning force f_p on pin thickness up to a pin thickness of 2ξ , the vortex diameter. This was a considerable success. However, there was one flaw in the agreement.

The basis for flux pinning was long believed to be a core pinning interaction between the normal α -Ti precipitate and the normal vortex core. However, the α -Ti pins are so narrow that they are proximity coupled to the matrix, thus creating an important theoretical complication not treated in the simple models used to compute f_p . A second issue is that the thickness dependence of f_p was found to fit best in the limit of T tending to T_c , where the field dependence of F_p was not a good fit to the field dependence of the order parameter. This apparently small inconsistency was attacked by Cooley et al. [18] in the context of the shape of the $F_p(H)$ curves of APC conductors, virtually all of which exhibit a peak at low fields, about $0.25H_{c2}$, rather than the $0.5 H_{c2}$ of the conventional materials (see Figure 1 and Figure 5). The proposal was made there without much justification that the pinning mechanism in the APC was a magnetic one, rather than a core pinning one. In fact it now seems that this may be the dominant mechanism in conventional Nb-Ti too [19,20]. A key prediction of the new model [20] is that optimum f_p occurs when the pin is decoupled from the matrix. The thickness of the pin and its electron mean free path determine this.

Within this model it is easy to rationalize the poor high

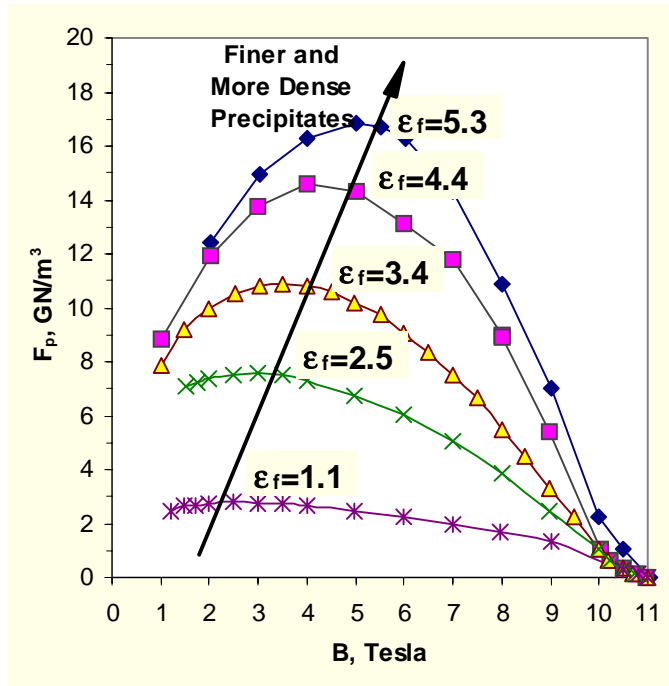


Figure 5 For conventionally processed Nb-Ti the bulk pinning force increases in magnitude with drawing strain after the last heat treatment. The increase occurs at all fields as the precipitate size and spacing are reduced to less than a coherence length in thickness (see Figure 2) (data from [21]).

field performance of most APC conductors: The reason is that their pins are pure Nb, which has a much longer mean free path than the alloyed α -Ti precipitate of the conventional process. The consequence of being optimized for thicker ($\sim 2-4 \xi$) pins is that the pin density is lower than for thinner pins, thus reducing the number density of vortex-pin interactions. Thus the lower field pinning benefits strongly but the higher field pinning suffers. The remedy is thus to dirty up the pin. Experimental tests of this hypothesis are now underway.

This is not quite the whole story since Figure 1 shows that the H_{c2} of the very good APC wire is $\sim 7.5T$, rather than the $10.5T$ of conventional wires. This is principally the consequence of the billet being experimental. Nb-47wt.%Ti was used for the matrix and Nb for the pins, meaning that the overall composition was very far from the optimum H_{c2} composition. As the correct choice of pinning center becomes better understood, this aspect of the reduced high field performance will be designed out by making sure that the overall composition of matrix and pin is one with a high H_{c2} .

One final note about APC wires should be made. The most successful one practically has been the jellyroll design of Supercon [3,22] that uses co-wound sheets of pure Nb and Ti. This technique produces a strand that is claimed to be cost competitive with conventionally made material. Both dipole and MRI magnets have been made with the design. The properties are very similar to conventional Nb-Ti but do not exceed the normal material. At this stage the nanostructure produced in the wires is not very clear.

B. Upper Critical Field.

A contour plot of H_{c2} at 2K for alloys in the ternary Nb-Ti-Ta system is shown in Figure 6. Ta and Nb are mutually soluble, similar chemically and mechanically and H_{c2} ($T < 4.2K$) is greater in the ternary than in the binary. As indicated by the figure, $H_{c2}(2 K)$ can be increased from a maximum of 14.25 to $15.5T$. These data indicate the prime reason for using Nb-Ti-Ta. In most other respects except one, the alloy processes well and can be used to replace the binary alloy when $1.8-2 K$ operation is required. So far at least 4 fairly large billets have been made of different alloys in the system:

Nb- 43wt.% Ti-25wt.% Ta	1980 UW[23,24]
Nb- 41wt.% Ti-15wt.% Ta	1983 Fermi/UW[4]
Nb- 44wt.% Ti-15wt.% Ta	1987 Fermi/UW[25,26]
Nb- 37wt.% Ti-22wt.% Ta	1992 Kharkov[27]

None of these has had the maximum H_{c2} composition, principally because of the desire to minimize the more expensive Ta.

Any new alloy must strike a balance between ensuring that there is enough Ta to raise H_{c2} and enough Ti to get sufficient α -Ti for pinning. In fact the best Nb-Ti-Ta alloys made so far have had $J_c(4.2K)$ values which are a little lower than

the binary but they do improve at 2 K. There have been few problems with Nb-Ti-Ta alloy, making it a rather simple retrofit if 2K use is being considered.

C. Fabricability

Conventional Nb-Ti and Nb-Ti-Ta fabricate extremely well, often producing piece lengths exceeding several km. Thus there is no concern about them. By contrast, APC conductors are sometimes quite hard to make. The problems are of several types. First is the fact that multiple extrusions are required to make an APC, except in the special case of the Supercon design mentioned above. Since each extrusion has a yield of ~85% at maximum, it is easy to see that a design requiring 3 or 4 extrusions has an inevitably low yield. An additional problem is that the composite is two-phase from the very beginning of fabrication and this leads to early work hardening and sometimes to fracture. This does not have to lead to problems but it is probably true that wire breaks are more common than with the standard process. These issues will take a greater importance as more of them made in an industrial environment.

III. SUMMARY

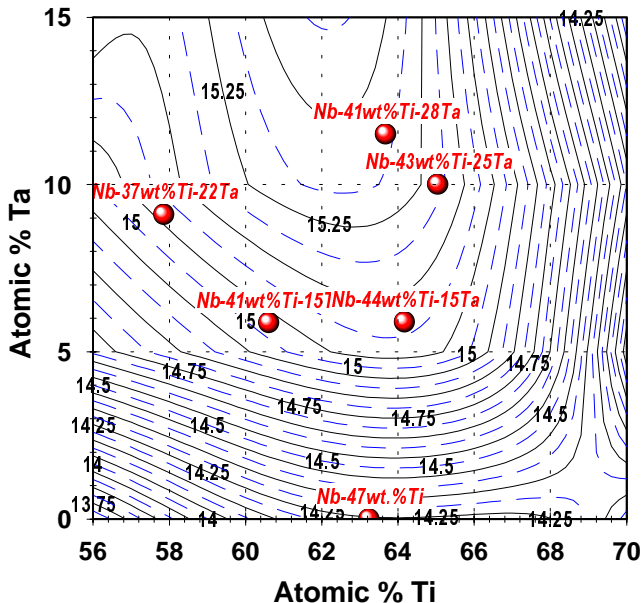


Figure 6 Contour plot of H_{c2} at 2K for Nb-Ti-Ta from the data of Hawksworth [23] and Muller [28]. The investigated alloy compositions are superimposed on this plot along with the standard binary Nb-47wt.%Ti. We use atomic composition here rather than the commonly used weight percentage because the all important volume % of α -Ti is more closely related to the atomic % Ti and the weight percentage values for the ternary are often misleading in this respect.

Recent advances in Nb-Ti alloys have been reviewed. A better understanding of flux pinning in the system offers opportunities for new and better prototype designs, perhaps of multilayers and APC conductors. APC composites as presently made suffer from having the wrong overall composition and perhaps also from having pure metal pins. Ternary alloys should be capable of further optimization since too few having close to optimum compositions have yet been made. Near term designs of specialty magnets for low beta quadrupoles may provide a stringent test of the potential of these new concepts, since applications of this type are not too sensitive to conductor cost. It does appear that Nb-Ti conductors with better properties can still be developed. The challenge is to integrate the lessons of the much better flux pinning properties of multilayers into more conventional round wire designs.

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