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MICROSTRUCTURE PROPERTY RELATIONSHIPS IN Nb-Ti-Ta

P.J. Lee, D.C. Larbalestier*, J.C. McKinnell** and A.D. McInturff***
Applied Superconductivity Center
University of WI-Madison, Madison WI 53706

Abstract--The microstructures produced by conventional precipitation heat treatment of Nb-44.4wt.%Ti-15.4wt.%Ta have been observed and quantified. The precipitate morphology was found to be qualitatively the same as for similarly processed Nb-47wt.%Ti providing that a prestrain in excess of 6.5-7 was applied prior to heat treatment. 80hr/420C heat treatments produced ≈10% more α-Ti than in the binary alloy but the J_c at 4.2K was lower in the ternary.

I. INTRODUCTION

The upper critical field of binary Nb-Ti is normally suppressed by spin-orbit coupling. The paramagnetic limitation can be reduced, however, by adding a high atomic number element (Ta, Hf, V) to reduce the coupling by spin-orbit scattering. The most promising of these ternary alloys has been identified as Nb-Ti-Ta([1])([2]) and there is a long history to the development of this ternary for high field use ([3]). At 4.2K the enhancement of H_{c2} is small (~0.3T) but at 1.8K the H_{c2} can be raised from ~14.2 for Nb-Ti to 15.5 for the best ternary alloys([2]). The key property, however, in terms of successful application of a superconductor is the J_c, and, although good J_c values at high fields have been obtained (2075 Amm^{-2} at 10T, 1.9K using a $10^{-14}\Omega m$ criterion([4])), the J_c values have been as high as has been anticipated from the best binary alloy performance.

The factors controlling J_c for binary Nb-47wt.%Ti have been extensively studied. In order to produce high J_c:

1 The initial alloy must be of a high homogeneity (local chemical variation of less than 2 wt.%).

2. The degree of filament sausaging during wire drawing must be minimized by optimizing the local Cu to superconductor ratio for filament support and by the use of a diffusion barrier around the filaments to prevent hard Cu-Ti intermetallic formation.

3. Sufficient cold work must be applied prior to initial precipitation heat treatment to prevent the formation of the deleterious ω and Widmanstätten α-Ti phases.

4. Multiple heat treatment and strain cycles must be applied in order to maximize the amount of α-Ti. A linear relationship has been found between the volume % of α-Ti and the J_c up to approximately 26 vol.%Ti([5]).

5. A final cold drawing strain of at least 4 is required to bring the dimension of the precipitates close to those of the fluxoid lattice. In industrial scale composites the J_c increases with strain until the benefits of the microstructural refinement are overtaken by filament sausaging (typically at a final strain of ~5).

Over the past decade, improvements in the J_c of Nb-Ti-Ta have been obtained, as in the binary, by addressing these 5 items. The most recent Nb-Ti-Ta composites have benefited from the advances in binary alloy and composite processing to the point where an assessment of the intrinsic properties of Nb-Ti-Ta is appropriate.

This paper examines the microstructural properties of a high homogeneity Nb-44.4wt.%Ti-15wt.%Ta (Nb-64.2at.%Ti-5.9at.%Ta) alloy fabricated at TWCA for the Fermilab quadrupole development program and fabricated into conductor at IGC([4]), Supercon([6]), and at the University of Wisconsin. This alloy composition was chosen after an initial high homogeneity billet of Nb-41wt.%Ti-15wt.%Ta (Nb-60.6at.%Ti-5.9at.%Ta) alloy appeared to respond to heat treatment sluggishly. The higher Ti composition was intended to increase the volume of α-Ti. Some J_c results have been reported previously([4]). The conclusion of that report was that the additional Ti in this alloy had resulted in a lower H_{c2}, resulting in a 4.2-2K operating temperature/field advantage of only 3.2-3.4T, rather than the expected 4T advantage. Nevertheless a high J_c of 2075 Amm^{-2} at 10T, 1.9K, $10^{-14}\Omega m$ was achieved.

II. EXPERIMENTAL

Both a conventional Nb-46.5wt.%Ti alloy and the ternary Nb-44.4wt.%-15wt.%Ta alloy were fabricated and processed in parallel. Slices of the ingots were supplied to the UW for analysis of the chemical homogeneity using WDS on an ARL SEMQ microprobe. Flash radiographs supplied by TWCA were used to identify the areas of lowest homogeneity.

Cu-clad Nb barrier-wrapped Nb-Ti-Ta monofilamentary conductor was supplied to the UW by TWCA. The monofilament was processed both directly into wire and into small scale

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*Also The Department of Materials Science and Engineering.

**Now at Teledyne Wah Chang Albany.

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Table 1. Heat Treatment Schedules

Composite ID	Multi or Mono-filament	Strain from final recrystallization anneal and Heat Treatment.				
		1st	2nd	3rd	4th	5th
530x	Mono	■ 6.4	■ 7.5	■ 8.6		
9x38	Multi	■ 9.9	■ 11.1	■ 12.2		
9x58	Multi	■ 7.3	■ 8.5	■ 9.9	■ 11.1	■ 12.2
x10	Mono	◊ 7	☐ 8.4	■ 9.8		

■ = 80hr/420C, ☐ = 20hr/420, ◊ = 6hr/405

(61 filament) composites under controlled conditions using the heat treatment schedules described in Table 1. I_c testing was performed using 0.6m lengths of wire wound on barrels mounted coaxially inside the bore of a 12T solenoid. An overall wire resistivity of $10^{-14}\Omega$m (~10μVm^{-1}) was used to determine the critical current. All the measurements reported here were performed at 4.2K, unless otherwise stated.

Precipitate quantification was performed by multi-tilt atomic number enhancement of TEM images performed on a high resolution Megavision 1024XM image analysis system([7]).

III. RESULTS

The chemical homogeneity, as revealed by flash radiography coupled with electron microprobe data, was very good. The worst case inhomogeneity was ±5wt.% Ti, ±4wt.% Nb and ±4wt.% Ta. Depletions in Ta tended to coincide with excess Nb as shown in Figure 1.

In Figure 2 and Figure 3, the J_c vs strain curves for the monofilamentary and multifilamentary composites are shown. The highest J_c values for the Nb-46.5Ti composites were obtained using the 3 × 80hr/420C heat treatment schedule on the multifilamentary composite, 3490A/mm^2 at 5T, 1384A/mm^2 at 8T, with a significant decline (almost 200Amm^{-2}) with the addition of 2 additional heat treatments. A curiosity of the 3 x 80hrs/420C composite was that the J_c of the final wire could be raised an additional 300Amm^{-2} at 5T by a final wire anneal of 2hrs/250C (the J_c at 8T was reduced by 40Amm^{-2}) whereas the same heat treatment *reduced* the J_c at 5T of the equivalent ternary composite by the same amount. Figure 3 shows that the peak J_c values for both the 3 and 5 80hr/420C heat treatment ternary wires were approximately the same and both were well below those of the binary. The peak in J_c occurred at a much lower strain ($\Delta\varepsilon_f$~1) for the five heat treatment multifilamentary conductor than for the three heat treatment schedule. A minimized heat treatment schedule ([5]) also produced lower J_c values in the ternary than the binary, as shown in Figure 2 .

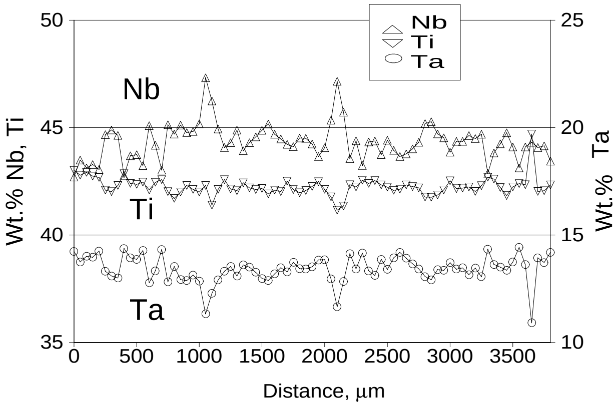

Figure 1 Electron microprobe WDS analysis of Nb-Ti-Ta billet at 6" diameter.

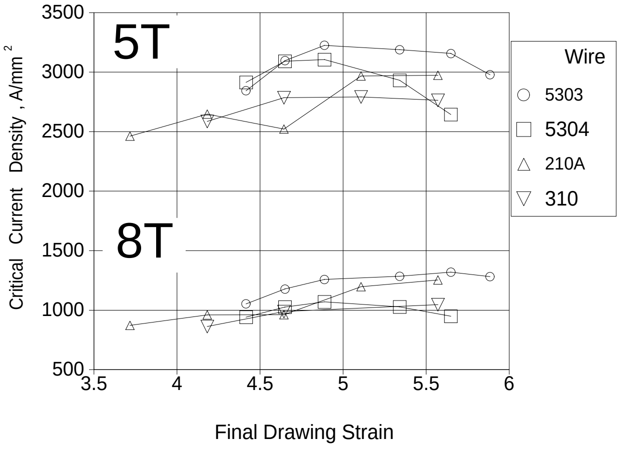

Figure 2 J_c versus final drawing strain for monofilamentary composites. Large symbols represent ternary alloy data (□5304 and ▽310).

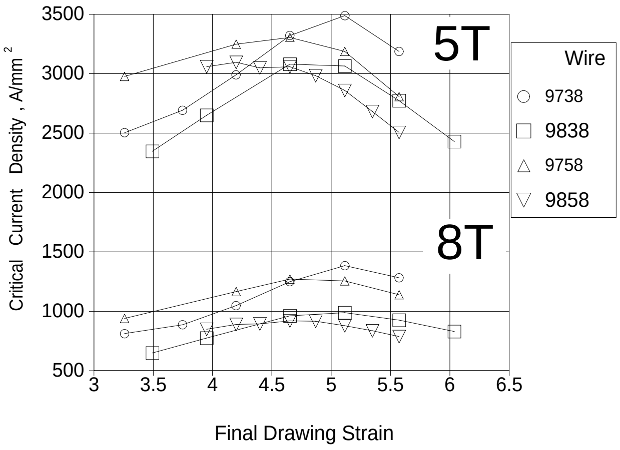

Figure 3 J_c versus final drawing strain for multifilamentary composites. Large symbols represent the ternary alloy (□9838 and ▽9858).

Table 2. TEM Analysis of α-Ti Dimensions and Volume for Transverse Cross-Sections.

Composite	Alloy, wt.%	Heat Treatment Conditions		Strain Before Heat Treatment	Volume % α-Ti	Mean α-Ti CSA, nm^2	Mean α-Ti diam.*, nm
		Time, hr	Temp., C				
9738	46.5	3 × 80	420	9.9	21	15500	140
9758	46.5	5 × 80	420	7.6	24	29660	194
9838	44Ti15Ta	3 × 80	420	9.9	25	32300	203
9858	44Ti15Ta	5 × 80	420	7.6	27	20870	163
UW210B	46.5	6, 20, 80	405, 420, 420	7	20	23630	173
UW310	44Ti15Ta	6, 20, 80	405, 420, 420	7	21	10540	116

* = calculated from the CSA assuming a circular cross-section.

In the 5304 monofilament, which received its initial heat treatment at a prestrain of 6.4, some Widmanstätten α-Ti was observed. This compares with a prestrain of 4.5-5 required to suppress the deleterious Widmanstätten α-Ti precipitate morphology in Nb-46.5wt.%Ti([8]).

Significantly more precipitate was produced for each heat treatment schedule in the ternary than for the binary (Table 2). The lower strain to peak J_c in the five heat treatment ternary composite coincides with a relatively small precipitates. However, the mean α-Ti transverse cross sectional area, CSA, for the three heat treatment multifilamentary binary composite is ~50% that of the ternary. Thus the lowered final strain to peak J_c in the five heat treatment material cannot be entirely attributed to precipitate size. In fact, there was no consistent trend in precipitate size with alloy or heat treatment. EDS indicated that there was less than 5wt.%Ta(2at.%) in the α-Ti, the Ta staying in the β matrix. The opposite was found for ternary additions of Zr([9]).

IV. DISCUSSION

Despite the reduction in wt.%Ti from 46.5wt.%Ti in the binary to 44.4wt.%Ti in the ternary, the volume % α-Ti was increased for all of the heat treatments (ranging from very aggressive to minimized). However, the use of weight % is misleading, as the atomic % of Ti rises from 62.8%Ti to 64.2%Ti. However, the increased volume % of α-Ti in the ternary was not sufficient to raise the J_c at 4.2K above the binary alloy and the J_c values were typically 6-10% less in the ternary than in the binary. This result is unexplained.

How then can the J_c of the ternary be improved? Reviewing the factors important to raising J_c outlined in the introduction:

1. High homogeneity Nb-Ti-Ta ingots were been produced. This material yields higher J_c than earlier inhomogeneous composites([3]).

2. Filament sausaging can be controlled by the use of good diffusion barriers, just as in binary Nb-Ti. All the composites fabricated here had high n-values. This result emphasizes the unexpected failure of the 5 heat treatment sample to have higher J_c than the three heat treatment sample. Copper poisoning of the near barrier region, ([10]), seems ruled out by the large filament diameters (> 50μm) of the 61 filament composites.

3. The large prestrain 6.4-7 required to suppress the deleterious ω phase and Widmanstätten α-Ti morphology is disappointing. Even if comparing the ternary to a binary of the same atomic % Ti (Nb-48wt.%Ti), a prestrain of only ~5.5 would be required ([11]). Unfortunately, restrains greater than 5 are difficult to incorporate into the restricted strain space of commercial practice.

4. The response of the ternary to multiple heat treatment is similar to the binary, although the ternary alloy produces ~10% more precipitate for the same heat treatment schedule.

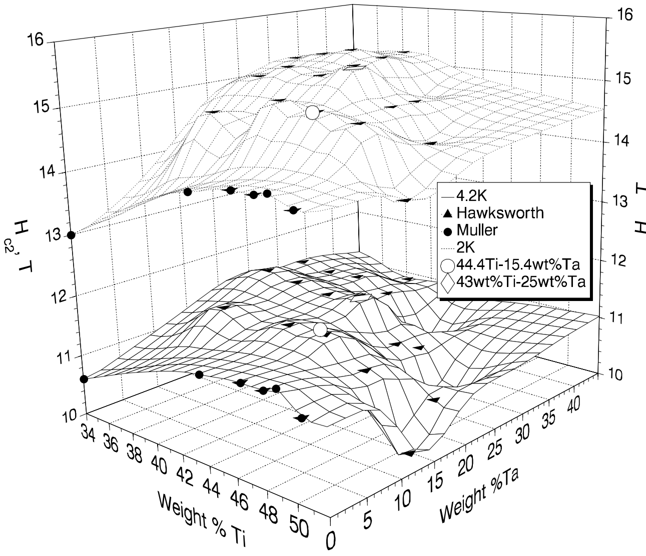

Figure 4 2K and 4.2K H_{c2} surfaces for Nb-Ti-Ta calculated from the data of Hawksworth and Larbalestier ▲([2]) and Muller ●([12]) using an inverse distance weighting algorithm.

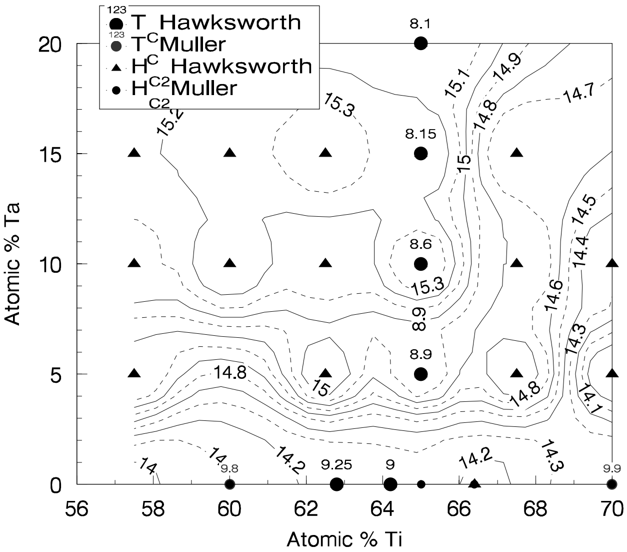

Figure 5. A contour plot for H_{c2} at 2K generated from the data of Hawksworth and Larbalestier ▲([2]) and Muller ●([12]).

5. The final drawing strain to peak J_c for the ternary is typically slightly lower than the equivalent binary($\Delta\varepsilon_f$=~.25). The precipitate sizes for the two alloys were different and had no clear trend.

With regard to these five criteria this 15wt.%Ta ternary composition appears to have been pushed close to its optimum performance. Gains in the performance of Nb-Ti-Ta, must come from the choice of a different alloy composition. The variation in H_{c2} with ternary composition is illustrated in the surface plot shown in Figure 4 calculated from the ternary data of Hawksworth and Larbalestier ▲([2]) and the binary data of Muller ●([12]). A contour map has been generated, Figure 5, for the variation in H_{c2} with atomic composition. Superimposed on Figure 5 are some T_c values (●) from Hawksworth and Larbalestier([2]) and Muller([12]). This plot reemphasizes the earlier conclusions of Hawksworth and Larbalestier([2]) and Segal et al.([3]) that optimum H_{c2} is found for higher Ta contents. The maximum H_{c2} is found for Ta contents of approximately 10-18 atomic %. Provided that the Ti content is at least 63 at.%Ti compositions in this range should produce sufficient α-Ti for high J_c.

V. CONCLUSIONS

1. The Nb-44.5wt.%Ti-15wt.%Ta produced ~10% more precipitate under the same condition than Nb-46.5wt.%Ti.

2. The J_c produced in the ternary is 5-10% lower at 4.2K than in the binary, despite the larger volume of precipitate.

3. With the high levels of α-Ti produced in these alloys it appears safe to replace more Ti with Ta in order to further raise the H_{c2}.

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