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Nb_3Sn Superconductors Made by an Economical Tubular Process

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Abstract— **The tubular technique for economical production of Nb_3Sn material with large numbers of subelements is being explored further by Supergenics I LLC and Hyper Tech Research Inc.**

The number of subelements has been raised to 271 (246+25) by increasing the size at which the restacking is carried out. The product exhibited no fabrication problems and was drawn down and tested at a wire diameter of 0.42 mm, where the subelements are 18 µm in diameter.

Heat treatment of a 271 subelement restack has been varied and a pre heat treatment of 48 h at 575 °C followed by long times at 635 °C has been shown to be advantageous. Restacks with 217 subelements have been subjected to both compositional and heat treatment changes and the best results, closely approaching those found on PIT materials, have been found on samples given 500 h at 625 °C.

***Index Terms*— Copper, Niobium, Tantalum, Tin, Tubular superconductors,**

I. INTRODUCTION

THE two main processes for producing high current density (J_c) Nb_3Sn superconductors are the internal tin (IT) process [1] and the Powder-in-Tube (PIT) method [2]. For some time now the IT process has reported the highest J_c values, (>3000 A/mm^2 at 12T) [1] but recently the PIT process has shown J_c values above 2600 A/mm^2 at 12 T [3 & 4]. While much of the concentration has been on J_c at 12 T, interest has also centered on stability in the intermediate field range and J_c at fields in the 15 – 18 T range.

To achieve improved stability in the low and intermediate field range, the effective diameter ($d_{eff.}$) must be made as low as possible and the residual resistivity ratio (RRR) of the matrix maintained at a relatively high value [5]. The internal tin process has employed barriers of Nb or Nb alloys around each of the subelements and, in order to achieve the highest J_c values, the material has been heat-treated in such a way that all the filaments within a barrier are bridged together after they are reacted to A-15. The result of this is that more and more subelements are needed in order to meet the low $d_{eff.}$ requirement.

Fig. 1 shows a cross section of EG 36, a 61 (54+7) subelement material that we have made by this conventional technique. While this gave a J_c in the non-Cu, after a heat treatment of 80 h at 650 °C, of 2757 A/mm^2 at 12 T and a similar sample, HP 31, gave 3030 A/mm^2 under the same conditions [12], they both had d_{effs} of 70 µm, a size that is considered larger than is desirable for stable behavior in the intermediate field range. In our work on this internal tin approach we found that the cost began to increase and the fabrication of long continuous lengths became more of a problem as the number of subelements was increased. These ductility problems appeared to decrease however as the thickness of the barrier was increased and we decided therefore to retry the tubular approach where all the filaments were replaced by a single tube of Nb7.5wt.%Ta. This approach was tried many years ago [6]-[8] but abandoned for reasons of low J_c.

We have developed a simple process of inserting Cu clad Sn into Nb7.5wt.%Ta tubes and these in turn are inserted into larger Cu tubes. We have previously reported [9]-[11] restacking 127, 169 and 217 of these subelements and drawing

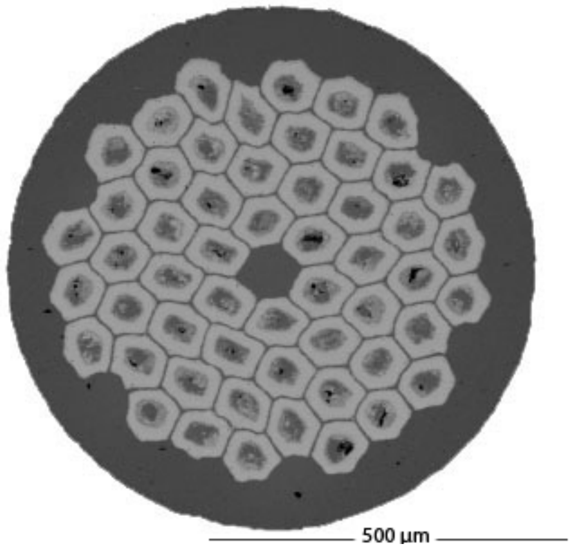

Fig. 1. EG 36 at 0.7mm diameter after 80 h at 650 °C.

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to wire of diameter 0.7 mm. This was accomplished without ductility and piece length problems. The best J_c reported was 2225 A/mm^2 at 12 T, and 1120 A/mm^2 at 15 T in material with 45 µm diameter subelements [10]. The process does not involve the use of the expensive powders that the PIT process employs. Nor does it require an electron beam (EB) welder, hot Isostatic pressing (HIP'ing) equipment or an extrusion press, all of which are used in the IT process. At the present time Nb based tubular material is more expensive and more difficult to obtain then the much smaller rods used in the IT process. There is no basic reason however that the relatively large diameter tubes should have these problems when the volume requirements are of the same order as those for the rods, Consequently the costs for the material and the processing will be lower than in the two other processes. Here we report increasing the number of subelements to 271 (246+25) and drawing down this material to a wire diameter of 0.42 mm where the subelement size was 18 µm. We have also changed the composition of the subelement and varied the heat treatment procedures in an attempt to improve the properties of a material with a low d_{eff}.

II. BROOKHAVEN NATIONAL LABORATORY (BNL) TEST RESULTS ON A 271 SUBELEMENT RESTACK

In our last paper [11] we showed a cross section of a restack with 217 subelements but we had not obtained properties on the material with that number of subelements at that time. This material was restacked in a Cu tube with an O.D. of 19.05 mm (0.75") and, while we had very few problems in assembling the array we felt that we were at the limit of the number of subelements that could be conveniently restacked in a tube of these small dimensions.

In order to explore the potential for increasing the subelement number still further we decided to make a 271 (246+25) restack (T 1489) using a 38.1 mm (1.5") diameter tube. This was drawn down to 0.7 mm (subelement size 31 µm) without any problems and a section drawn down further to 0.42 mm (subelement size 18 µm). Both of these were heat treated initially at BNL at 650 °C for 90 and 60 hours respectively. The J_c of the 0.7 mm material was 2030 A/mm^2 at 12 T and the J_s was 4170 A/mm^2. The J_c of the 0.42 mm material was 2050 A/mm^2 at 12 T and the J_s was 8495 A/mm^2. A cross section is shown in Fig. 2.

Based on their experience with Oxford internal tin material BNL suggested we try different temperatures for the heat treatment. We gave the 0.7 mm diameter material 144 h at 635°C and obtained a J_c of 2177 A/mm^2 at 12 T and a J_s of 4310 A/mm^2. When given 48 h at 665 °C a lower value, 1919 A/mm^2 at 12 T was obtained. These samples were given a lower temperature preheat treatment of 48 h at 575 °C.

Another two samples of T 1489 material were then prepared. One had an additional 144 h at 635 °C and the other a single heat treatment this time without the 575 °C preheat treatment. The extra time at 635 °C raised the J_c a small amount to 2222 A/mm^2 at 12 T. The sample without the pretreatment had properties similar to those of the material heat treated for 90 h at 650 °C, i.e. 2022 A/mm^2 at 12 T. This suggests the intermediate treatment may give an improvement in properties. These are summarized in Table 1.

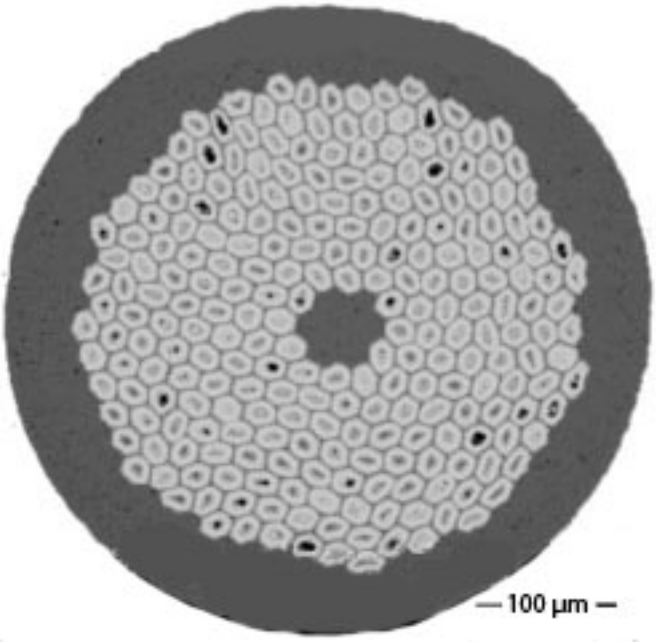

Fig. 2. T 1489 at 0.42mm diameter after 60 h at 650 °C.

TABLE I. BNL RESULTS ON T1489

	Wire OD	Pre HT	HT	J_c at 12 T (A/mm^2)
T1489	0.42mm		650 °Cx60h	2051
T1489	0.7mm		650 °Cx90h	2030
T1489	0.7mm	575 °Cx48h	665 °Cx48h	1919
T1489	0.7mm	575 °Cx48h	635 °Cx144h	2177
T1489	0.7mm	575 °Cx48h	635 °Cx288h	2222
T1489	0.7mm		635 °Cx144h	2022

The RRR's of all the samples were low indicating that Sn had entered into the matrix.

III. FURTHER TEST RESULTS

It was obvious from the RRR data that Sn was entering the matrix during the heat treatment of the T 1489 material and it was decided to vary the composition of the subelements in order to reduce the extent to which this would occur. Since the costs to carry out such an investigation on 271 subelement material would have been much greater than if the work was done on the 217 subelement material made in smaller tubes, this latter approach was chosen for the following work.

First a baseline material, T 1505, a 217 subelement restack was made with the same subelement composition as that of the 271 restack, T1489, and tested at OSU after similar heat treatment conditions. It had a subelement size of 35 µm, slightly larger than that of T 1489. The J_c of T 1505 after 100 h at 650 °C was 2247 A/mm^2 at 12 T and 1386 A/mm^2 at 14 T. A cross section of this material after 96 h at 650 °C is shown in Fig. 3. After 144 h at 635 °C it gave 2146 A/mm^2 at 12 T and 1268 A/mm^2 at 14 T. The time at 635 °C was increased to 200 h and a value of 2250 A/mm^2 at 12 T and 1460 A/mm^2 at 14 T was obtained.

A sample was then heat treated at 625 °C for 500 h and the J_c results are shown in Fig. 4, compared with those obtained after 100 h at 650 °C. The sample was damaged at

13.5 T and the low field data had to be estimated from an extrapolation of the Kramer plot. It appears that this material will exhibit 2400 A/mm^2 at 12 T, giving it properties very close to the best PIT material. Fig. 5 shows a cross section of this material after 500 h at 625 °C. This shows indications that not all the subelements have been heat treated to the point where the Sn is about to go into the matrix. This suggests that a modification of the composition and/or the heat treatment may improve the J_c even further.

Another sample of T 1505 at 0.7 mm was also heat treated for 500 h at 625 °C. It gave a J_c at 12 T of 2443 A/mm^2 and 1655 A/mm^2 at 14 T. The results are also shown in Fig. 4 and Table II.

Another restack, T1583, was made with a Nb7.5wt.%Ta/Sn ratio slightly smaller than both T 1489 and T 1505 and this was heat treated at 650 °C for 120 h and 635 °C for 200 h. The results were 2098 A/mm^2 and 1921 A/mm^2 at 12 T respectively. The ratio of Nb7.5wt.%Ta/Sn was increased in the next restack and the material, T 1581, heat treated at 650 °C for 84 h and 200 h. The J_cs obtained were both low, 1825 and 1883 A/mm^2 respectively but the material was very stable with an RRR of 96.5. Metallography showed that the Nb7.5wt.%Ta was incompletely reacted and affected very little by increasing the time of the heat treatment. It was concluded that the ratio of Nb7.5wt.%Ta/Sn in T1581 had been increased too much and, based on these results; we decide to make a material with an intermediate ratio. This material was designated T 1628.

None of the above samples tested by OSU had been given the 575 °C preheat treatment that improved the J_c of the BNL T 1489 samples. A sample of T1505 was then given such a treatment followed by 300h at 635 °C. The properties were very similar to those obtained after a straight 200 h at 635 °C.

The T 1628 material was then heat treated for 200 h at 650°C, 250 h at 635 °C and one for 48 h at 575 °C followed by 300 h at 635 °C. The results showed that this material had properties slightly inferior to those of T 1505 showing that the composition, and/or the heat treatment still requires more refinement.

A sample of T1505 is being heat treated for 700 h at 625 °C and will be sent to BNL The data should be available in the near future.

Some of the J_c properties of T 1505, T 1583 and T 1628 are summarized in Table II.

IV. SUMMARY

A low cost tubular process has been developed that makes subelements by simply putting Cu clad Sn into a Nb7.5wt.%Ta tube, inserting this into a Cu tube and drawing the assembly down. These tubular subelements are then assembled in large numbers, introduced into another Cu tube and the assembly drawn to fine wire.

We have made 271 subelement restacks and drawn them down to 0.42 mm diameter wire without significant problems. We therefore feel that manufacture of restacks containing 469 or more subelements and reducing these to 0.7 mm to yield $d_{effs.}$ of 18 μm will not offer any serious problems.

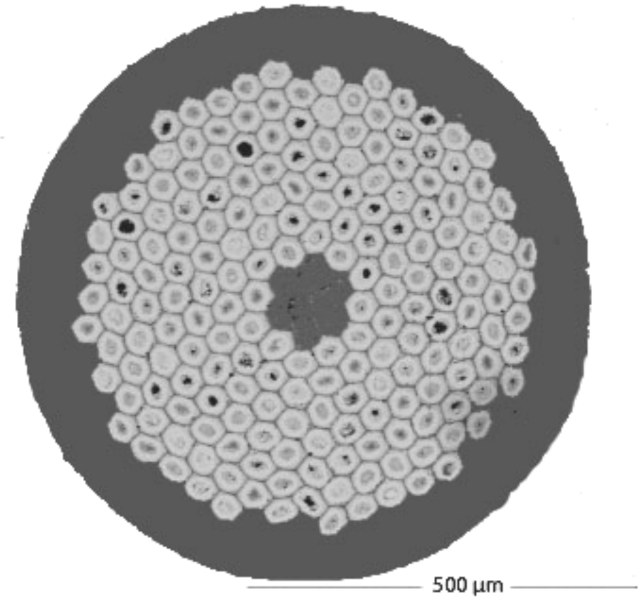

Fig. 3. T 1505 at 0.7 mm diameter after 96 h at 650 °C

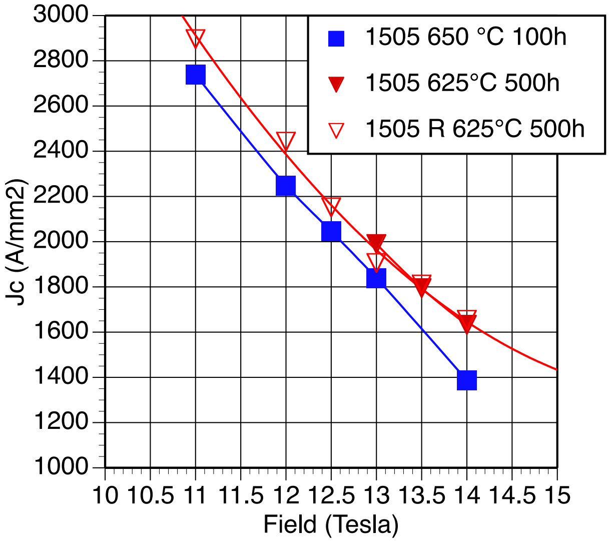

Fig. 4. J_c of T 1505 after 100 h at 650 °C & 500 h at 625 °C

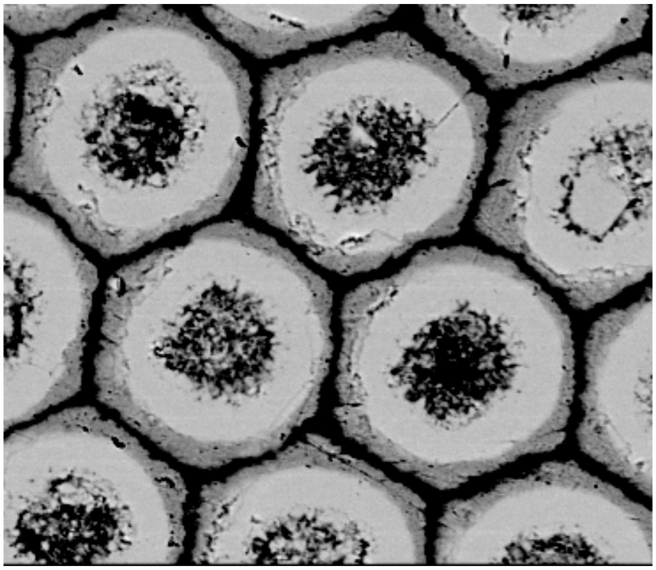

Fig. 5. T 1505 after 500 h at 625 °C

Working with 271 subelement restacks where the $d_{eff.}$ was 31 µm we have manufactured material with a J_c at 12 T of 2250 A/mm^2. With 217 subelement restacks we have started to explore the effects of compositional and heat treatment variables on J_c and have achieved a value of 2443 A/mm^2 at 12 T and 1655 A/mm^2 at 14 T. Under these conditions this material does not appear to be reacted completely and as a result we feel raising the J_c another 10% should be possible in the near future.

TABLE II. OSU RESULTS ON T 1505, T 1583, AND T 1628

	Wire OD	**HT**	**J_c at 12 T (A/mm^2)**	**J_c at 14 T (A/mm^2)**
T1505	0.7mm	650 °Cx100h	2247	1387
T1505	0.7mm	635 °Cx144h	2146	1269
T1505	0.7mm	635 °Cx200h	2250	1460
T1505	0.7mm	625 °Cx500h	2400*	1633
T1505	0.7mm	625 °Cx500h 575 °Cx48h	2443	1655
T1505	0.7mm	635 °Cx300h	2214	1426
T1505	0.7mm	635 °Cx144h	2022	1269
T1583	0.7mm	650 °Cx120h	2098	1351
T1583	0.7mm	635 °Cx200h	1921	1186
T1628	0.7mm	650 °Cx200h	2047	1308
T1628	0.7mm	635 °Cx250h 575 °Cx48h	2003	1335
T1628	0.7mm	635 °Cx300h	2113	1309

*value from extrapolation of Kramer plot

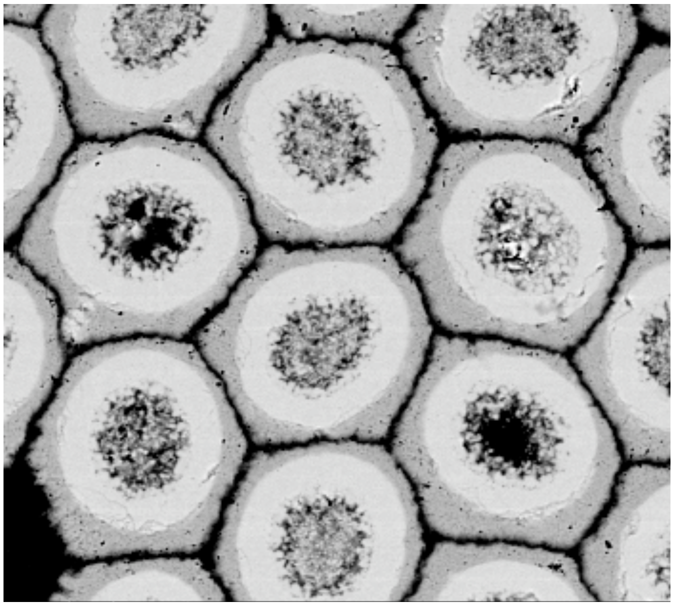

Fig.6. T 1628 at 0.7 mm diameter after 200 h at 650°C

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