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| 著者 | 渡辺　和雄 |
|---|---|
| journal or publication title | IEEE Transactions on Magnetics |
| volume | 21 |
| number | 2 |
| page range | 316-319 |
| year | 1985 |
| URL | http://hdl.handle.net/10097/47223 |

# PROPERTIES AND PERFORMANCE OF THE MULTIFILAMENTARY $Nb_3Sn$ WITH Ti ADDITION PROCESSED BY THE Nb TUBE METHOD

S. Murase, H. Shiraki, M. Tanaka, M. Koizumi, H. Maeda and I. Takano
Toshiba R & D Center, 4-1 Ukishima-cho, Kawasaki-ku, Kawasaki 210, Japan

N. Aoki, M. Ichihara and E. Suzuki
Showa Electric Wire & Cable Co., Ltd., 2-1-1 Odasakae, Kawasaki-ku, Kawasaki 210, Japan

K. Noto, K. Watanabe and Y. Muto
Tohoku University, 2-1-1 Katahira, Sendai 980, Japan

## Abstract

MF $Nb_3Sn$ conductors made by titanium added niobium tube process have been developed for use in high fields. Composites, consisting of Nb-1.0wt.%Ti tube with a copper sheathed tin core inside and a high conductivity copper tube outside, were stacked together up to 264 filaments, then drawn to the final sizes without any intermediate annealings. As workability was further improved by the titanium addition, it was possible to decrease the Cu/SC ratio for the conductor down to 0.67 and to increase the tin concentration up to 30 wt.% in Cu-Sn inside the filament. An enhanced layer growth rate and a slightly increased grain size of $Nb_3Sn$ were observed in the titanium added conductors. compared with those made by use of pure Nb tube. The critical current density at high fields, which was more sensitive for heat-treatment condition, was superior to that of the other conventional processed conductors. Especially, the conductors with 30 % tin content have high critical current density for $Nb_3Sn$ layer and an excellent field dependence of critical current density without copper: 1550 A/mm$^2$ at 10 T, 600 A/mm$^2$ at 15 T and 350 A/mm$^2$ at 17 T. The critical current was also measured in the temperature range 1.88 - 4.2 K. A coil, aiming at 14 T, was fabricated using the conductor with 25 % tin to assure high level performance in practical situations.

## Introduction

$Nb_3Sn$ multifilamentary conductors have been developed for high field applications such as plasma confinement system in fusion reactor, hybrid magnets, NMR, laboratory use magnets for physics and so on. Recently, improvements in high field performance for the $Nb_3Sn$ conductors processed by the bronze method have been endeavored by the addition of the third element, for instance Ti, Ta, Ga, Mg or Hf, to the niobium filament and/or to the bronze matrix. Titanium is known as one of the most significant elements among additions.[1,2]

On the other hand, the conventional $Nb_3Sn$ conductor processed by the bronze method has imperfections: (i) necessary intermediate annealings due to work hardening of the bronze, (ii) low tin concentration (13-14 wt.%) and (iii) necessary diffusion barriers to protect the copper stabilizer. In order to eliminate those drawbacks, the $Nb_3Sn$ conductor processed by the Nb tube method has been developed.[3,4] The method offers conductors of tubular niobium filaments, each filled with a copper sheathed tin inside and embedded in a matrix of high conductivity copper outside. In the present method, (i) intermediate annealings are unnecessary because the composite which consists of pure metals mitigates the work hardening during the reduction, (ii) high tin concentration inside the niobium filament is possible and (iii) the niobium tube filaments play a role in diffusion barrier during $Nb_3Sn$ formation.

As a further modification, titanium addition to the tubular niobium has been carried out to enhance high field performance for the conductors processed by the Nb tube method.

This paper gives the results obtained from conductors fabricated using the Ti added Nb tube method, particularly, critical current densities and grain morphology in relation to the heat treatment condition and the tin concentration inside the tubular niobium. Preliminary results, obtained from a coil wound with the present conductor, are also described.

## Fabrication Process

A single core was fabricated using a Nb-1.0wt.%Ti tube with copper sheathed tin inside and copper outside. For fabricating MF conductors, single core wires were bundled, together 7 to 264 filaments in a copper tube, then drawn down to final sizes without any intermediate heat treatments and finally submitted to a reaction heat-treatment to form $Nb_3Sn$. The present method can vary tin concentration widely inside the filament, depending on the copper sheath thickness. Data on fabricated conductors are listed in Table I and a cross-sectional view of A1 conductor is shown in Fig. 1. As workability was further improved by the titanium addition, Cu/SC ratio was able to decrease down to 0.67 for conductors B and D, and tin content could be increased up to 30 wt.% in copper sheathed tin for conductors D and F1-3. A3 conductor was reduced to a rectangular cross-section for ease in winding a coil from a 1 km length.

Table I. Data on fabricated conductors

| Conductor Numbev | Wire Diameter (mm) | Number of Filaments | Filament Diameter ($\mu$m) | Cu/SC ratio | Tin Contents (wt.%) in Cu-Sn |
|---|---|---|---|---|---|
| A1 | 0.87 | 264 | 36.2 | 1.2 | 25 |
| A2 | 0.65x1 | 264 | ~36 | 1.2 | 25 |
| A3 | 1.25x2.5 | 264 | ~86 | 1.2 | 25 |
| B | 1.17 | 37 | 150 | 0.67 | 25 |
| C | 1.05 | 37 | 120 | 1.0 | 25 |
| D | 1.4 | 37 | 180 | 0.67 | 30 |
| E1 | 0.54 | 7 | 120 | 1.98 | 25 |
| E2 | 0.69 | 7 | 150 | 1.98 | 25 |
| E3 | 0.80 | 7 | 180 | 1.98 | 25 |
| F1 | 0.54 | 7 | 120 | 1.98 | 30 |
| F2 | 0.69 | 7 | 150 | 1.98 | 30 |
| F3 | 0.80 | 7 | 180 | 1.98 | 30 |

Manuscript received September 10, 1984

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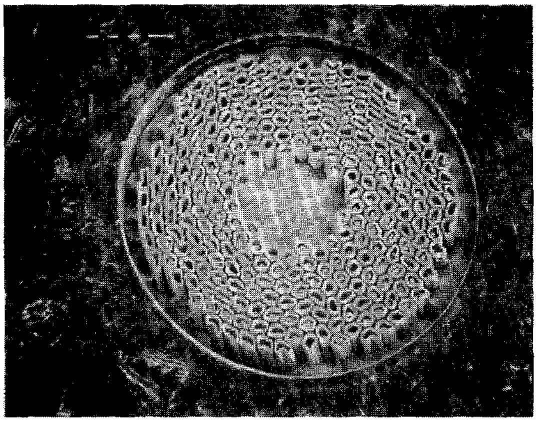

Fig. 1 Cross-sectional view of a conductor processed by the modified Nb tube method (A1 conductor)

### Experimental Procedure

The fabricated conductors were heat-treated in the range of 675 - 775°C for 1 - 312 h to form the $Nb_3Sn$ layer inside the niobium tube. The grain morphology was studied using an SEM and an optical microscope. The composition profile was analyzed by an EPMA. Critical currents ($I_c$) were measured in the transverse magnetic field from 8 to 20 T at 1.88 - 4.2 K using 1 μV/cm criterion. The hybrid magnet, which consists of a water cooled coil and a superconducting coil at Tohoku University, was used for $I_c$ measurement above 15 T.

### Conductor Characteristics

#### $Nb_3Sn$ Layer

The effects of Ti addition on the layer growth rate were investigated. Figure 2 shows $Nb_3Sn$ layer thickness versus heat-treatment time for A1 conductor, compared with a pure $Nb_3Sn$ conductor with the same configuration. In the wires with Ti addition, the time required for reaching the saturated layer thickness was 1/3 - 1/4 of the time for the pure $Nb_3Sn$. The result shows that $Nb_3Sn$ formation reaction occurred more actively by the titanium addition.

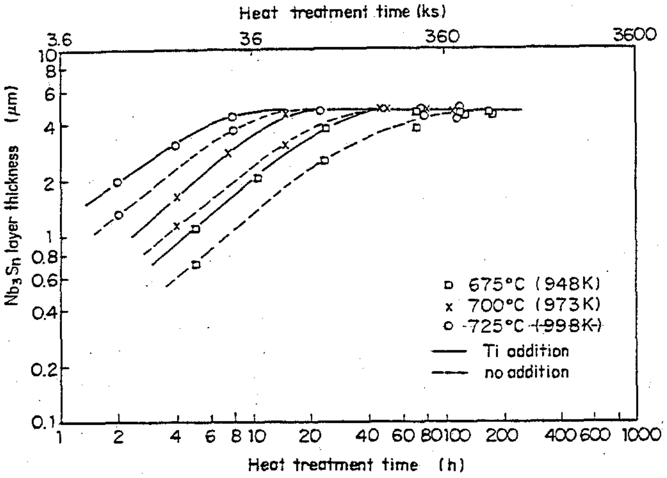

Fig. 2 Dependence of the $Nb_3Sn$ layer thickness on the heat-treatment time

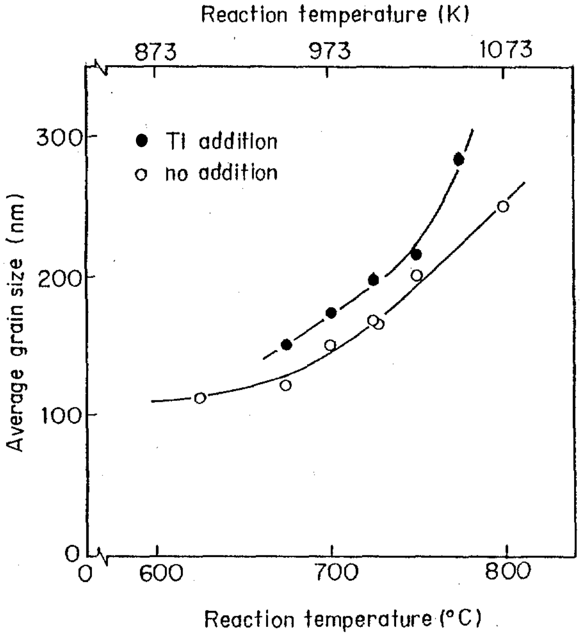

Fig. 3 Average grain size for $Nb_3Sn$ plotted against the reaction temperature

Figure 3 shows $Nb_3Sn$ grain size versus heat-treatment temperature. The $Nb_3Sn$ grains become coarse with increasing temperature. The grain size for titanium addition is slightly larger than the pure $Nb_3Sn$ conductor.

Titanium diffusion in the composite was studied by an X-ray microanalyzer for reacted A1 conductor. The results are shown in Fig. 4. Titanium diffused only into the $Nb_3Sn$ layer (1.1 wt.% Ti in $Nb_3Sn$) and was not traced in the copper stabilizer and reacted Cu-Sn alloy inside the filament. In the Nb tube method, the titanium diffusion mechanism is thought to be the same as that for the conductor by the bronze method.[1] Critical current density ($Nb_3Sn$ layer) at high fields increases as titanium content in the $Nb_3Sn$ layer increases, for the range of 0.5 - 1.5 wt.% Ti in $Nb_3Sn$, in the case of titanium addition to the niobium core.[1] As 1.1 wt.% Ti in $Nb_3Sn$ for the Nb tube method is larger than 0.5 % for the ordinary bronze processed wire, high performance is expected at higher fields for the Nb tube processed conductor.

### Critical Current Density

25 wt.% Sn conductor: In order to estimate the optimum heat-treatment condition for A1 conductor, characteristics of critical current density without copper ($J_c$) vs. heat treating time at several heat-treatment temperatures were obtained, as shown in Fig. 5. It can be seen that the optimum heat-treatment time decreases with increasing heat-treatment temperature. $J_c$ peaks at low fields were not observed remarkably as compared with the ones at high fields. It was considered that $J_c$ is more sensitive for heat-treatment condition at high fields than at low fields.

The magnetic field dependence of $J_c$ in A1 conductor heat-treated for 120 h at 700°C is shown in Fig. 6, compared with pure $Nb_3Sn$ conductor by the same process. Critical current density without copper of the conductor with 25 wt.% Sn was superior to pure $Nb_3Sn$ at high fields and also to the highest $J_c$ obtained in the conductors using the other conventional method.[1,2] $J_c$ values for conductors E1 and E2 treated for 240 h at 725°C, which were also plotted in Fig. 6, became slightly

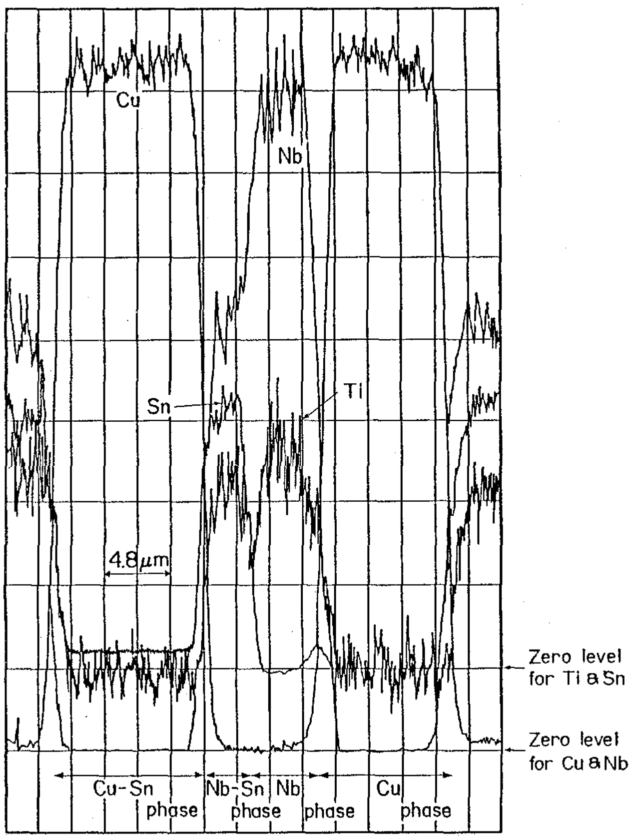

Fig. 4 Composition profile on the cross-section of A1 conductor

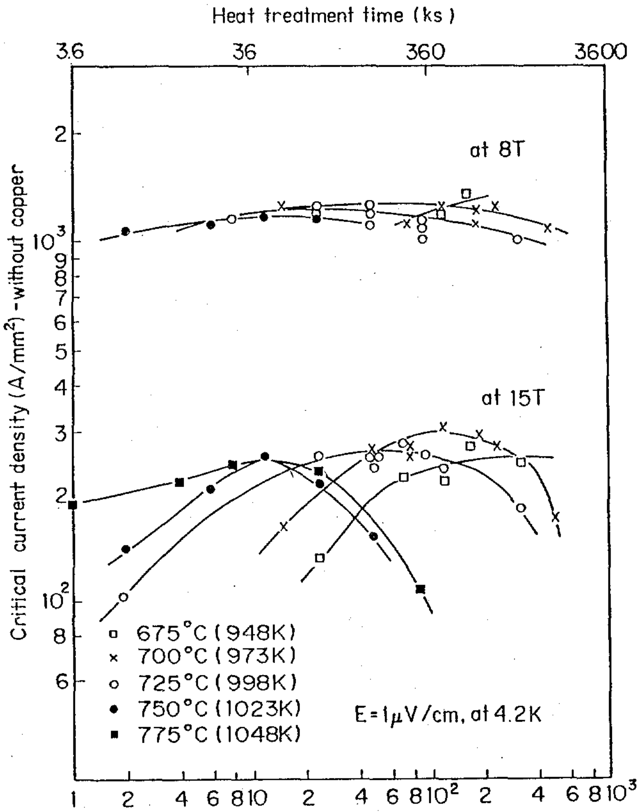

Fig. 5 Critical current density vs. heat treatment time at several heat treatment temperatures

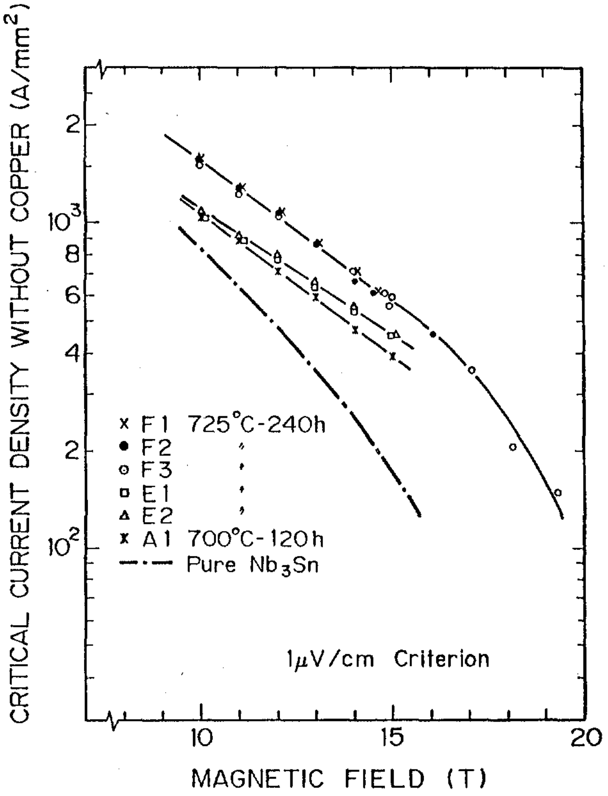

Fig. 6 Field dependence of $J_c$ in various conductors

higher than that for A1 at high field region: 450 A/mm$^2$ for E1&2 and 390 A/mm$^2$ for A1 at 15 T in contrast with 1050 - 1100 A/mm$^2$ at 10 T for A1 and for E1 and E2. Upper critical fields ($H_{c2}$), extrapolated from Kramer's plot, were obtained to investigate this difference further in detail: 26.5 T for E1 and for E2 and 24.5 T for A1 conductor. Since E1 and E2 conductors were subjected to longer term heat-treatment, it is considered that $H_{c2}$ increases and subsequently high $J_c$ at high fields is obtained as compared with A1.

30 % Sn conductor: High tin content (30 wt.%) conductors, F1-3, have performed excellent field dependence of critical current density: 1550 A/mm$^2$ at 10 T, 600 A/mm$^2$ at 15 T and 350 A/mm$^2$ at 17 T, as also shown in Fig. 6. Those values are larger than those for the ECN-Holec wire(5) at high fields above 15 T and are about 50 % up from those for conductors E1 and E2 with 25 % Sn content. $H_{c2}$ obtained for 30 % tin conductor from the Kramer's plot was 26.5 T, which was the same as that for 25 % tin conductor. From experimental results obtained for $Nb_3Sn$ layer thickness for 30 % tin conductor, which has the same thickness as that for 25 % tin conductor, it is considered that high $J_c$ in 30 % conductor would be caused by $J_c$ increase in $Nb_3Sn$ layer.

$J_c$ below 4.2 K: For A1 conductor heat-treated for 72 h at 725°C, field dependence of $J_c$ was studied below 4.2 K, as shown in Fig. 7. Normalized critical current, $I_c(2.13\ K)/I_c(4.2\ K)$, which increased with increasing the magnetic field, is 1.3 at 10 T and 1.5 at 15 T. The $H_{c2}$ increase from 4.2 K to 2.13 K, was 2.5 T, which was obtained from those data using Kramer's plot.

### High Field Solenoid Fabrication

In order to check the feasibility of fabricating a long wire, which is necessary for practical application,

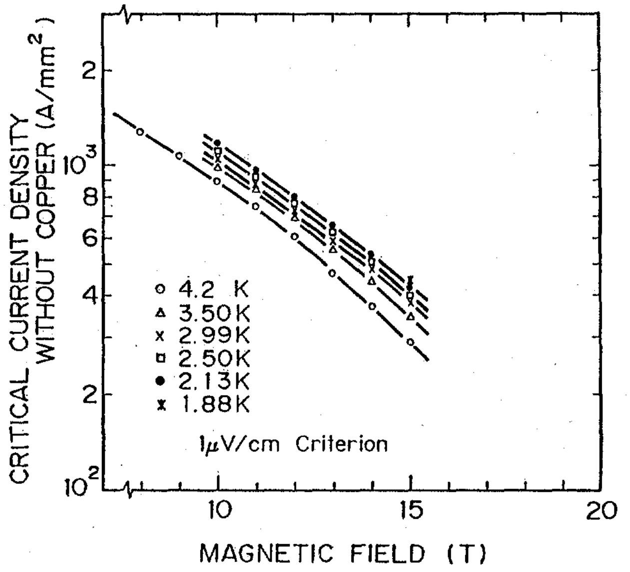

Fig. 7 Field dependence of $J_c$ below 4.2 K in Al conductor heat-treated for 72 h at 725°C

the authors fabricated a high field (13 - 14 Tesla) solenoid with the A3 conductor given in Table I. The solenoid is 40 mm in inner diameter, 140 mm in outer diameter and 140 mm in length. The conductor cross section was $1.25 \times 2.5$ $mm^2$; individual filament size was 86 μm. The magnet utilized a react after wind process; therefore, the conductor was insulated with quartz-fiber braids, while the coil form was insulated with heat resistant ceramic-fiber papers. After a heat treatment of 725°C x 181 h, the magnet was vacuum impregnated with epoxy resin. The solenoid was inserted into a bore of a NbTi backup magnet; 159 mm in inner diameter, 276 mm in outer diameter and 295 mm in length. Parameters for both magnets are listed in Table II. The magnet system was designed to generate 14.3 T at maximum, as shown in Fig. 8. Actually, it reached 13.6 T in a recent experiment in our laboratory, which is indicated by the closed circle in the figure.

## Conclusions

The modified Nb tube method, titanium addition to the niobium tube, offers MF $Nb_3Sn$ conductors with good workability and high critical current density. Several types of conductors have been fabricated, such as 0.67 - 1.98 Cu ratios and tin contents of 25 wt.% and of 30 wt.%. The present conductors were heat-treated for 1 - 312 h at 675 - 775°C to study grain morphology and superconductivity. Subsequently, $Nb_3Sn$ layer thickness, grain size, composition profile and critical current density at the field range of 8 - 20 T and at the temperature range of 1.88 - 4.2 K were measured for the present conductors. $Nb_3Sn$ layer growth rate was three or four times as large as the pure $Nb_3Sn$. Titanium in the composite diffused only into $Nb_3Sn$ layer (1.1 wt.% Sn in $Nb_3Sn$ after reaction) and was not traced in the copper stabilizer and reacted Cu-Sn inside the filament.

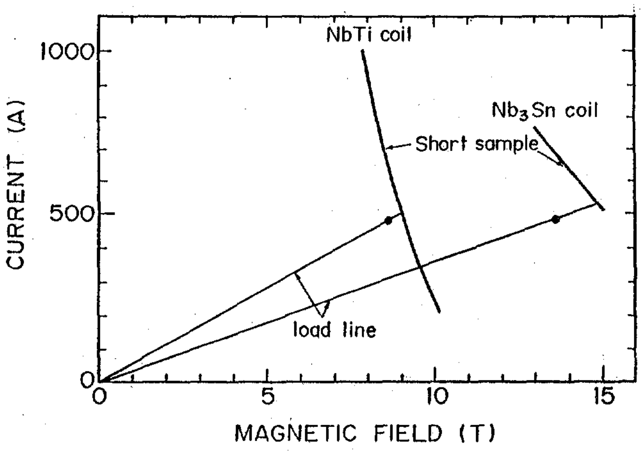

Fig. 8 Coil characteristics

Table II. Coil parameters

| Items | $Nb_3Sn$ coil | NbTi coil |
|---|---|---|
| Conductors | | |
| Conductor cross section ($mm^2$) | 1.25 x 2.5 | 1.2 x 2.4 |
| Copper ratio | 1.2 | 1.0 |
| Insulation | Quartz fiber braids | Formvar |
| Coil | | |
| Inner diameter (mm) | 40 | 159 |
| Outer diameter (mm) | 140 | 276 |
| Coil length (mm) | 140 | 295 |

$J_c$ became more sensitive for heat-treatment condition with increasing the magnetic field. $J_c$ for 25 % tin conductor was superior to pure $Nb_3Sn$ at high fields and also the highest $J_c$ obtained in the conductors using the other conventional processes. Conductors with high tin content (30 %) have shown an excellent field characteristic of $J_c$ values, such as 1550 $A/mm^2$ at 10 T, 600 $A/mm^2$ at 15 T and 350 $A/mm^2$ at 17 T, which are larger than those for ECN-Holec wires at high fields above 15 T and about 50 % up from those for conductors with 25 % tin content. $J_c$ below 4.2 K was also measured. Finally, the coil fabrication, aiming at 14 T, was achieved using the conductor with 25 % tin content to assure high performance of practical conductors.

## References

1. K. Tachikawa, H. Sekine and Y. Iijima, "Composite-processed $Nb_3Sn$ with Titanium Addition to the Matrix", J. Appl. Phys., Vol. 53, (1982) pp.5354-5356.
2. K. Kamata, N. Tada, K. Itoh and K. Tachikawa, "High-Field Current-Carrying Capacities of Titanium Bronze Processed Multifilamentary $Nb_3Sn$ Conductors with Pure and Alloy Cores", Proc. ICMC-CEC in Colorado Springs (1983).
3. Y. Koike, H. Shiraki, S. Murase, E. Suzuki and M. Ichihara, "Fabrication of Multifilament $Nb_3Sn$ Conductors", Appl. Phys. Lett., Vol. 29 (1976) pp.384-386.
4. S. Murase, M. Koizumi, O. Horigami, H. Shiraki, Y. Koike, E. Suzuki, M. Ichihara, F. Nakane & N. Aoki, "Multifilament Niobium-Tin Conductors", IEEE Trans. Magn., Vol. MAG-15, (1979) pp.83-86.
5. J. Williams and L. Neuringer, Annual Report July 1982 through June 1983, Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, (1983) p.92.