CRITICAL CURRENT, ELECTRO-MECHANICAL PROPERTIES AND SPECIFIC HEAT OF BRONZE NB_3SN CONDUCTORS

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Abstract
The fabrication process leading to a Nb3Sn wire by using the bronze route with 15.4 wt%. Sn is described. The critical current density, J_c, is studied as a function of the applied magnetic field, B, up to 25T; the uniaxial strain, ε, was measured up to 17. In the second part our device for measuring Ic(ε) is presented. The device is based on the concept of the Walters spring (WASP), which allows to measure long length wires (voltage taps distance up to 50 cm), up to 1'000 A and to obtain an absolute measurement of the strain value. It is thus possible to measure the voltage-current relation of technical superconducting wires and tapes down to 0.01 μV/cm, an important requirement for the characterisation in view of applications like NMR high field magnets which require persistent mode operation with high current densities. Finally specific heat measurements on Nb3Sn wires prepared at GAP have allowed to determine for the first time the overall distribution of Tc in the filaments. The onset of Tc was observed at 17.2 K, the Tc distribution being centred at 15.9 K. This analysis confirms the reduction of Tc due to the Ti addition and the presence of a distribution of Sn in Nb3Sn bronze wires.

1. INTRODUCTION

Currently the main commercial applications of LTS and HTS are magnets, which can be either solenoids, dipoles or multipoles (accelerator magnets). Up to now, Nb_3Sn is the best candidate for high field solenoids operating in persistent mode operation and high field dipoles. In the last years, various attempts have been made to enhance the critical current density, J_c and the upper critical field B_{c2} [1], introducing the additives Ta and Ti to the superconducting phase. There is still room for improvements for Nb_3Sn, mainly by increasing the Sn content and by improving the homogeneity of the A15 layer. In the last years, considerable progress has been achieved on the critical current densities of Nb_3sn wires produced by the Internal Sn Diffusion technique (ISD) [2] and the Powder-In-Tube (PIT) process [3]. The ultimate performances for a certain application can be reached also by better knowledge of the mechanical properties of the wires; in this way the design of the magnet could be adjusted in order to fully exploit the potentiality of the conductor. At GAP the activities are centred on the development of Nb_3Sn bronze wires and the advanced characterisation of the critical current as a function of field and strain. For applications at lower fields (12T-15T), i.e. for accelerator dipoles, bronze wires are actually not yet performing as well as PIT and ISD wires, but at fields exceeding 20T bronze wires constitute the best choice.

In the first part of the paper our activities on the development of bronze wires for high field applications (>21T) are presented. The design has been optimised to reach a higher uniformity in the degree of reaction of the filaments and several wires with different Ti, Ta, content have been manufactured to determine the optimum doping for high field applications [4].

The second part of the paper describes the device for the electro-mechanical characterisation of technical superconducting wires and tapes. In superconducting magnets the compressive and hoop stresses can reach very high values, and knowledge of the behaviour of the critical current as a function of strain and stress becomes an important issue for the optimisation of the magnets and of the conductors. Therefore it is important to have detailed knowledge of this behaviour to obtain strain limited, optimised Nb_3Sn wires for magnets with even higher fields.

In the last part we report about recent specific heat measurements on the Nb_3Sn wires presented in the first part. The specific heat gives information on the distribution of composition inside the whole superconducting sample, and it could be a very powerful tool for the optimisation of the conductors.

2. BRONZE WIRES FOR HIGH FIELD APPLICATIONS

2.1 Fabrication

The Nb_3Sn wires produced at GAP are based on Cu-Sn bronze produced by the Osprey technique. Here we present three wires: two (wire A and B) are quaternary wires and one (wire C) is ternary (see table I). For the fabrication of wire A the filaments consisted of Nb-7.5wt.%Ta rods with NbTi insertions, to form NbTa/NbTi composites [4]. Another way of adding Ti is to use bronze doped with 0.25% Ti and Nb-7.5wt.%Ta rods for the filaments; this method has been used for the fabrication of wire B. We have also manufactured a ternary wire, using Nb-7.5wt.%Ta rods as filaments and the same bronze as for wire A. A typical layout for the three wires is presented in fig. 1.

Table 1

	A	B	C
matrix composition	Cu15.4Sn	Cu15.5Sn0.25Ti	Cu15.4Sn
filament composition	Nb7.5Ta1.0Ti	Nb7.5Ta	Nb7.5Ta
local bronze to filament ratio	2.2	2.2	2.2
global bronze to filament ratio	2.5	2.5	2.5
number of filaments	14'641	14'641	14'641
diameter of filaments	4.5 μm	4.5 μm	4.5 μm
filament spacing	2.1 μm	2.1 μm	2.1 μm
stabilizer material / content	Cu / 20 %	Cu / 20 %	Cu / 20 %
barrier material / content	Nb / 10 %	Nb / 10 %	Nb / 10 %

The deformation process to obtain wires with a final diameter of about 1 mm requires three hot hydrostatic extrusion steps. Before the third extrusion a Nb barrier and a copper stabilisation are added. After the third extrusion step, the rods consisting of 14'641 filaments were drawn into round conductors of 1.25 mm diameter and about one hundred metres length. The local bronze/filament ratio (after the second extrusion step) was chosen to 2.2, while the global ratio was 2.5. The filament diameter was 4.5 μm, the spacing 2.1 μm. Two different reaction treatments (670°C/100h and 600°C/100h-670°C/150h) were performed under a pressure of < 10-5 mbar. Heating and cooling rates were chosen at 30 °C/h.

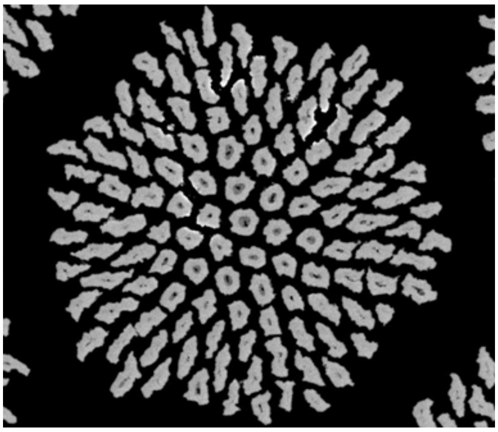

Fig.1. Filament bundle inside a bronze route Nb_3Sn wire (wire A) after reaction.

2.2 Critical currents

The critical current of the three wires has been measured on standard ITER barrels (∅ 40mm) at fields up to 17 T at 4.2K. The wire A was also measured up to 24T at the High Field Magnet Laboratory in Nijmegen; the upper critical field, B_{c2}, has been estimated to be 24T, i.e. markedly lower than the value extrapolated by the Kramer plot based on the data taken between 10 and 17 T. At 17 T, after a heat treatment of 700°C for 150h the critical current of wire B was about 20% higher than for wire A. A two step heat treatment (600°C/100h-670°C/150h) allows to obtain a further increase of 16% respect to the one step heat treatment. At 17T the wire C has almost the same critical current than wire A; at lower fields the I_c increase faster than for the quaternary wires, so we expect lower performances in high field with respect to the Ta,Ti doped wires.

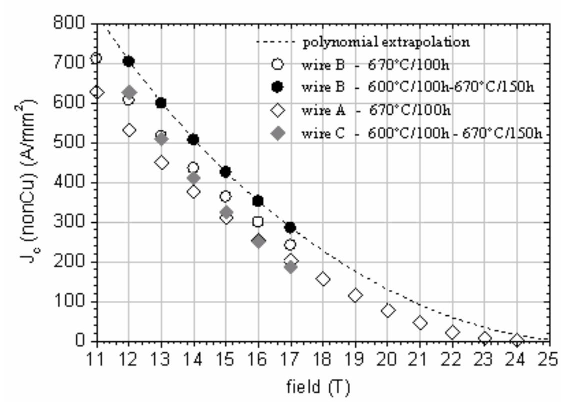

Fig.2. J_c versus B at 4.2K for bronze route Nb_3Sn wires; the J_c criterion is 0.1μV/cm (round wires, ∅ 1.25mm).

The usual method to extrapolate J_c values to higher fields is to fit the J_c data on a Kramer plot with a straight line. In some case this method gives J_c values higher the measured values. We have found that a polynomial fit of the J_c data on a Kramer plot gives values close to the measured ones. The critical current for the wire B has been extrapolated to higher fields fitting the Kramer plot with a polynomial curve (see fig. 2); this method allows more realistic values than the standard linear fit, especially in the 17T - 21T range. In the next section the behaviour of critical current under strain will be presented. Further analysis on the homogeneity of the A15 layers in the wires will be presented in the third section.

3. ELECTRO-MECHANICAL PROPERTIES

Various probes for the measurement of $I_c(\varepsilon)$ have been described in the literature. Ekin et al. [5] developed a sample holder which fits into very high field hybrid magnets. A modified design has been reported by ten Haken et al. [6]. The design of Specking et al. [7] allows higher forces by using a split-pair magnet, but the field strength was limited to 15 T. A common drawback of these methods is the short sample length of a few cm (corresponding to the size of the magnet bore). Voltage taps for the I_c measurement are typically separated by 5 to 10 mm. Due to this short distance, it is difficult to apply the usual 0.1 μV/cm criterion of the standardised critical current measurements [8] and the 1 μV/cm is thus currently used. Another difficulty due to the small high field magnet bores, is the short distance between voltage taps and current contacts (transfer length) [9]. Recently at the University of Twente a new device has been developed, which allows measuring longer samples; the distance between voltage taps is few cm and the current transfer length is of the order of 10 cm.

3.1 Description of the Walters spiral

All the problems due to short length can be overcome by the ingenious design of Walters et al. [10] who wound the wire on a spiral in order to introduce long wire lengths in the reduced high field space. A modified version of the Walters spiral (WASP) has been constructed in Geneva. By keeping one end of the spring in position and by rotating the other end, the wire can be strained either under compression or under tension. Because the spring is made of Ti6Al4V, reversible and linear strains up to 1.4% can be applied at 4.2 K [11]. Due to the sample length of the order of 80 cm, there is no current transfer problem and even the 0.01 μV/cm criterion can be applied. The method of Walters has not been used extensively because of the unreliable soldering process between the superconducting wire and the Ti-alloy forming the WASP. This problem was recently circumvented by using CuBe instead of the Ti-alloy, which however, limits the maximum strain to 0.7% [12]. On the other hand, soldering the total length allowed compressive strains to be applied too. In our implementation of the Walters spring the above mentioned problems have been solved; not only the zero strain on the wire can be determined precisely, but there is also the possibility to solder on the Ti alloy spiral, thus allowing to access to the compressive regime. In addition the effect of the different thermal contractions during the cool-down run can be controlled.

The WASP consists of a Ti alloy; at both ends of the spiral the wire is soldered to copper blocks, which act as current terminals. The wire sits in a grove that has been machined on the spiral; several spirals with different groove size have been built for measuring either rectangular or round wires with different cross sections and also for measuring tapes (up to 4.5 mm in width). The essential part of the modified WASP is shown in fig. 3. The bottom is fixed and the top can be rotated by a d.c. motor located outside of the cryostat. Close above the WASP, but still in the liquid helium bath, a piezoelectric sensor measures the torque. The angular position of the motor can be determined by an optoencoder [13].

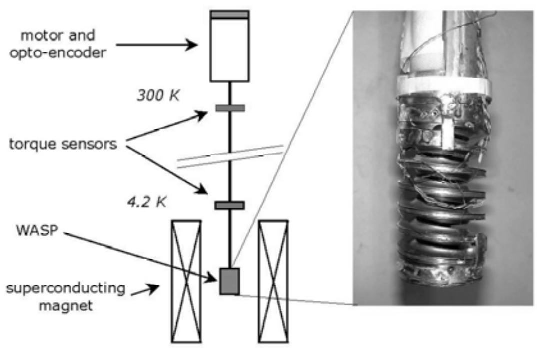

Fig.3 Set-up of Walters spiral (WASP), Geneva design.

3.2 Electro-mechanical properties: $J_c(B,\varepsilon)$

The following wires have been characterised: the wires A, B and C (described in the previous section) and a round binary wire (0.8mm diameter) manufactured by Furukawa for the ITER benchmark test [14].

The variation of the critical current as a function of strain at 17T is shown in Fig. 4a and 4b for the wires A, B and C. Strains ranging from –0.5% to 0.4% has been applied to wire B. The Ekin model has been used to fit the critical current; one of the parameters used in the Ekin model is the critical field, B_{c2}, which can be found by fitting the $I_c(\varepsilon)$ curve. In the case of wire B the transport measurements gave a value of 24T for the critical field. The critical field determined by the Kramer extrapolation of transport measurements in the range 10T-17T is 27T. The critical field obtained by the Ekin model is about 25.5T; this is the B_{c2} at $\varepsilon=\varepsilon_m$. When correcting for the prestrain effect (ε=0.25%) the value of 24.3T is found, which is closer to actual value than the Kramer extrapolated value. We believe that the dependence of the critical current from strain could be used for the determination of the critical field when a direct measurement in high magnetic is not available. This method could be more accurate that the standard Kramer extrapolation.

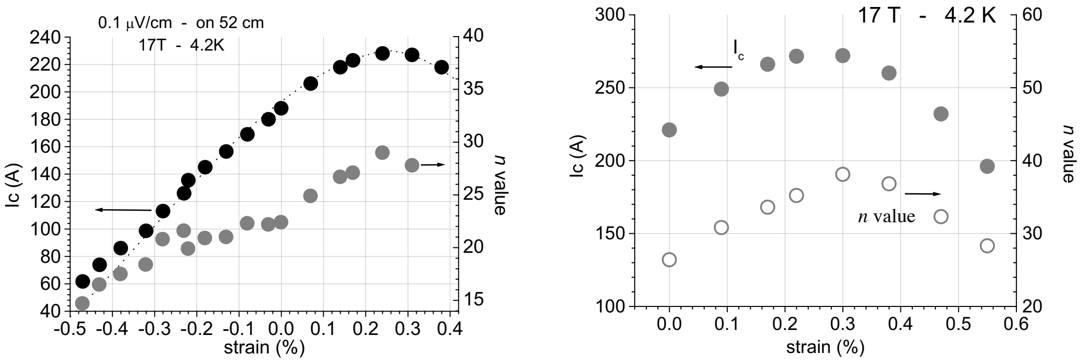

Fig.4a-b Critical current (0.1 μV/cm criterion) and *n* value for the $(Nb,Ti,Ta)_3Sn$ bronze wires (A on the left and B on the right) manufactured at the University of Geneva; both are square wires, 1.37X0.92mm^2, twisted. The dashed line is Ekin's model: p=0.5; q=2; n=1; u=1.7; $a(\varepsilon<\varepsilon_m)$=1250; $(\varepsilon>\varepsilon_m)$=900; B_{c2}=25.5T.

The Furukawa wire was measured following the same procedure as the wire above; in this case the field was 12T. Strain up to 0.9% was applied to the wire and the voltage-current relation was measured over three decades.

3.3 $J_c(B,\varepsilon)$ with a criterion of 0.01 μV/cm: a test for possible wire degradation

On the Furukawa wire it was possible to determine the critical current at 1 μV/cm, 0.1 μV/cm and even at 0.01 μV/cm. The *n* values have been calculated doing a linear fit of the V-I curves in the region between 0.1 μV/cm and 1 μV/cm. All the data have been obtained for both the loading and unloading process, in order to study the effect of the filaments cracking on the V-I curves. In fig.5 the critical current determined with the three criteria are plotted as a function of strain. The ascending branches of the $I_c(\varepsilon)$ curve are very similar, regardless of the criteria. Apparently the maximum value of I_c decreases slightly when increasing the criterion. At the maximum applied strain (0.9%) the I_c depends strongly on the criterion: for instance the 1 μV/cm I_c is reduced of 60%, while the 0.01 μV/cm I_c is reduced of 80%. The large dependence from the criterion is still present when the strain is released back to 0%. The 1 μV/cm critical current suffered a degradation of 5%, while for the 0.1 μV/cm and the 0.01 μV/cm criteria the reduction was respectively 20% and 40%. The high voltage sensitivity of our device allows to detect the formation of crack, which cause an irreversible behaviour of the critical current.

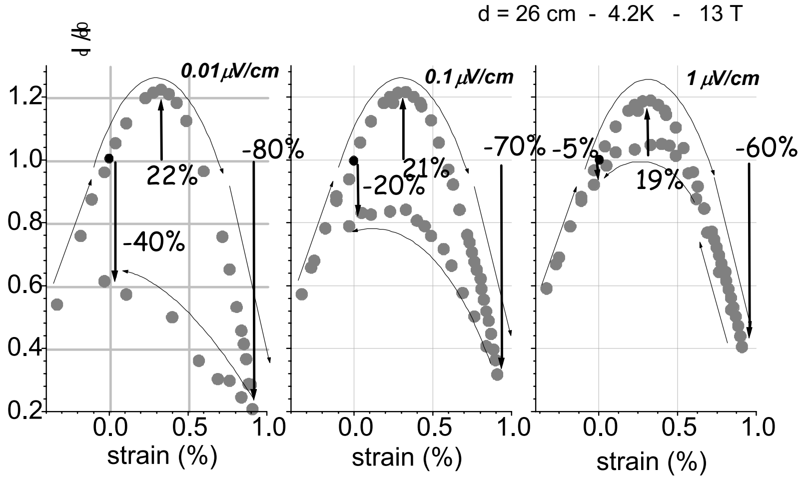

Fig..5 Critical current (0.01 μV/cm, 0.01 μV/cm and 0.01 μV/cm criterion) for the Furukawa ITER wire. The degradation of the critical current at 0.9% and after the strain recovering depends strongly on the voltage criterion: the recovered I_c is 5% lower than the initial value for 1 μV/cm but it is 40% lower for the 0.01 μV/cm.

4. DISTRIBUTION OF T_C BY SPECIFIC HEAT ANALYSIS

The measurement of low temperature specific heat is particularly appropriate to the determination of the critical parameters of superconducting materials, as it indicates the properties of the whole volume, without being influenced by screening effects. In particular it allows to probe multifilamentary wires and to obtain reliable measurements of the distribution of the critical temperature inside the filaments. Specific heat has been measured on a short piece of the wire A and wire C at 0 and 14T, and a distribution of critical temperature has been calculated. The same analysis has been carried out on very homogeneous bulk sample with a composition of 24.8% Sn as determined by the value of residual resistivity and of the ratio c/a of the tetragonal deformation [15]. The results are presented in fig. 6: the T_c distribution of the wires is centred at a lower temperature respect to that of the bulk sample; the reasons for this shift are the lower overall Sn content (this effect is present in both wires) and the presence of Ti which is known to lower T_c [4] (in the case of wire A). In addition the width of the distribution is much larger than for the bulk sample. The reason is the inherent Sn gradient of the A15 layer in bronze route filaments with unreacted Nb cores [16]. The width of the distribution is similar in both wires, so apparently the Ti doping seems to have no influence on the homogeneity of the A15 layer.

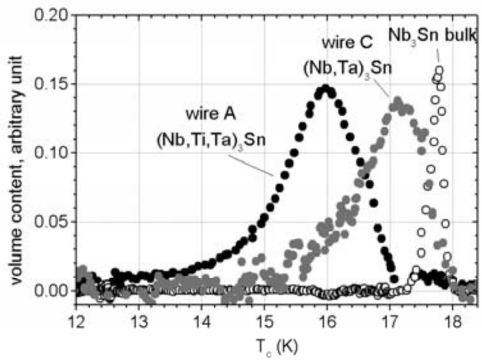

Fig. 6 Critical temperature distribution for the wires A and C, compared to the bulk sample [15].

5. CONCLUSIONS

In the present paper we have described the fabrication process of bronze $(Nb,Ta,Ti)_3Sn$ wires optimised for high field applications. The variation of J_c has been measured as a function of field and uniaxial strain: at 17 T, the non-copper value of J_c reached 285 A/mm^2, the *n* factor being as high as 50. The maximum of J_c vs. strain was determined to $\varepsilon_m = 0.26$ % by means of the modified Walters spiral.

A device to measure the critical current of long length technical superconducting wires and tapes (up to 0.8 meter) carrying up to 1000 A at 4.2 K in fields up to 17 T has been developed. The advantage of this apparatus with respect to the usual linear devices resides in the longer wire length, thus allowing one to use a more stringent criterion for the determination of the critical current (down to 0.01 μV/cm instead of 1 μV/cm). It is also possible to determine the true strain acting on the wire and to apply compressive strain. Another advantage is the high measuring current (up to 1000 A); no problems arise from the current transfer length, the distance between the soldered ends and the voltage taps being ≥ 10 cm; moreover measurements on long lengths are also more representative in the case of twisted wires. The device can be used to better characterise commercial superconducting wires, in particular Nb_3Sn wires for high field NMR applications, and it can be used to study the effect of strain on critical current from the fundamental point of view.

These measurements will be extended to higher fields, a new 21T magnet being recently installed in our laboratory.

Low temperature specific measurements have been carried out to determine the T_c distribution inside the A15 layer of our bronze route Nb_3Sn wires. The same analysis has been done on a homogeneous bulk sample, with a Sn content of 24.8 at. %. The T_c distribution of the wires peaks at a lower T_c value and is considerably wider than that of the bulk sample. The lower T_c onset for the filaments is due to addition of Ti, while the larger width is due to the Sn gradient inside the filaments. Taking into account these results, and knowing the relationship between T_c and the Sn content, it appears that the Sn distribution in the A15 layer of bronze route Nb_3Sn wires can still be optimised, thus letting room for further improvements of J_c.

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