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Importance of local stresses and strains during the fabrication of Nb$_3$Sn composite superconductors.

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Abstract. The evolution of the distribution of strains and stresses within the multifilamentary composites designed for an improved bronze route is characterized all along their restack bundling process of fabrication. The results of numerous investigations of the microstructure are briefly compared to those of finite element simulations. The work compares the effects of the difference in mechanical behaviour of the various components to that of their repartition. The importance of the composite lay-out, that is to say of the relative disposition and of the volume of the bronze areas, as well as of the size, morphology and spacing of the filaments, for the Nb$_3$Sn formation is demonstrated. The consequences for both the Nb$_3$Sn grain size and the critical current are finally displayed.

1. Introduction
The multifilamentary Nb$_3$Sn composites are the foremost material candidate for the fabrication of high energy physics magnets. The ever hard to please adjustments between their electromagnetic properties requires continuous improvements in the wire design as well as in the definition of its forming parameters [1]. Their fabrication is a hard task because in particular of the high discontinuity of plastic flow among their components. The current paper emphasizes the importance of both the disposition and the differences in mechanical behaviour of the various constituents for the uniformity of deformation in a Nb$_3$Sn composite. The work further deals with the consequences of these features for the formation of Nb$_3$Sn and the superconducting behaviour. The present results will be the more pertinent as they are obtained with not too stringent composite designs characterized by reduced differences in mechanical properties among their components and by their strong bonding.

2. Experimental
The wire was manufactured by a restack bundle drawing process that comprises two steps. Each step was composed of an extrusion stage, defined by a true deformation of about 4 at 650°C, followed by a series of room temperature drawing operations. A true deformation of 0.8 was given during each drawing sequence that ended by a 1h at 580°C anneal. During the first step, a 6.1 true strain was applied at ambient temperature to a composite made of 54 filaments of niobium that are concentrically disposed in four rods within a 9.23at%Sn bronze matrix (see figure 1a). This first composite rod was finally cut in equal lengths that were restacked as 178 bundles of filaments in a new composite whose
architecture is depicted on figure 1b. In the final composite the bundles are regularly disposed in four concentric layers and again within a 9.23at%Sn-Cu matrix. Going from the centre towards the wire surface, these bundle layers are going to be designed by the letters A to D in the following of the paper (see Fig. 1b). Starting from the wire centre, the area fractions of the various components are 15.5% for the pure Cu core, 4% for the Ta layer, 5.3% for the innermost layer of bronze, 49% for the multifilamentary zone including the matrix and 26.2% for the outermost layer of bronze. At the end of metalworking, a prolonged heat treatment at 690°C was given at either the 0.3, 0.8 or 1mm wire diameter in order to transform the filaments of Nb into Nb₃Sn. The microstructural investigation of the local strains and stresses was performed with seventy samples cut along the fabrication and using a number of techniques [2]. A finite element analysis (FEM) was also used in order to identify the more effective parameters among the numerous interlocking variables intrinsic to the macroscopic parts of the cold working of composites [1]. The early calculations were performed by means of the Abaqus 6.7-1™ program and neglecting the thermal effect. The basic process variables were respected. The assumptions of the perfect bonding of the constituents, of their continuous work hardening and of the absence of friction at the die interface were also made. More details will be published elsewhere.

3. Results and Discussion

3.1. Primary design of composite

3.1.1. First extrusion. The composite deformation is non uniform from the end of the first extrusion. Thin foil transmission electron microscopy shows that the bronze matrix is not fully recrystallized as it contains a significant density of twins and stacking faults. In agreement with this observation, the Hv₁₀₀ microhardness of the matrix exceeds that of its fully recrystallized state by 45 units which indicates a close to 0.16 level of remaining deformation at room temperature. Such a result warrants the usual assumption according to which the level of remaining deformation at the end of a hot extrusion stage is negligible in front of the whole deformation by cold drawing.

3.1.2. Cold drawing stages and intermediate anneals. According to the rather constant value of the area of the filament cross-section depicted on Figure 1a, the deformation of the first design of composite is rather uniform at the end of the first stage of fabrication. The hardness data together with the results of the FEM analysis are however consistent with the occurrence of a maximum of flow in the part of the external bronze layer located in close contact of the outermost row of filaments. This peak of deformation is due to the crushing of the softer bronze layer against the central composite zone (see Fig. 2). It is also the cause of the slight elongation of the outermost filaments of row 4 in the hoop direction of the composite. In addition the systematic measurements of the area of the cross-section (S₀ and S, before and after deformation, respectively) of both filaments and wire lead to figure 3 that depicts the trend towards a reduced true deformation (ε = ln S₀/S) of the filaments compared to
that of the wire all along the first stage of deformation. The occurrence of such a behaviour during a cold drawing sequence between two intermediate heat treatments has also been predicted by FEM (see Fig. 2). The preferential deformation of the matrix has to do with both its almost full recrystallization during each intermediate anneal [2] and the difference in mechanical behaviour of both components.

![Figure 2. FEM simulation of the evolution of the equivalent strain along a radius of the first design of composite during cold drawing.](image)

**Figure 2.** FEM simulation of the evolution of the equivalent strain along a radius of the first design of composite during cold drawing.

![Figure 3. Comparison of the average true strain of filaments (points on unbroken curve) with that of the composite (dashed line) along the first stage of fabrication. Each arrow indicates the existence of an anneal of stress relief.](image)

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3.2. Final design of composite

3.2.1. Effect of the Ta barrier. The presence of the Ta barrier with a far higher mechanical resistance than that of the other components [3] is extremely detrimental to the uniformity of plastic flow.

At the end of the second extrusion, the Ta barrier already exerts a strengthening effect at least in the non-Cu part of the wire. Going from the outer surface towards the barrier, the Hv100 hardness remains almost constant and around 184 up to the bronze interval between the innermost rows B and A (see figure 1b); after which it continuously increases up to the Ta barrier where it peaks at about 240.

In accordance with this hardening effect, figure 4 proves that at the lowest levels of deformation, the Ta layer and with a lower importance its adjoining components, i.e. the Cu-core and the innermost bronze zone, are less deformed than the remainder of the composite. Figure 4 displays the effect of the cumulative strain on a deformation factor defined by the ratio: (\(\varepsilon_{\text{wire}} - \varepsilon_{\text{component}}\))/\(\varepsilon_{\text{wire}}\), where \(\varepsilon_{\text{wire}}\) and \(\varepsilon_{\text{component}}\) are the true strains of the whole composite and of one of its component, respectively. The hindering action of the Ta layer on the plastic flow of its adjoining components is again obvious from the results of the three-dimensional finite element simulations (see Fig. 5). In a more general way, FEM has further proved that the magnitude of the differences of deformation increases with the volume fraction of the strong barrier. Moreover, and fortunately for the process, figure 4 exhibits a clear trend towards an uniformity of deformation in the various zones of the main part of the composite with the progress of the second step of metalworking. All the previous results verify the importance of the choice of the chemical nature and microstructure, and therefore mechanical behaviour of the various components together with their volume fraction and disposition for the homogeneity of deformation of a composite during metalworking. More particularly, the choice of an as-cast Ta barrier that possesses as a rule an extremely large grained microstructure is the more problematic as it gives rise to a non uniform mechanical strength and therefore to a heterogeneous deformation of the barrier [4].
3.2.2. Effect of the components lay-out. The location of the various constituents together with their dimension and morphology exert a strong effect on the kinetics of Nb$_3$Sn formation. The histograms in figure 6 prove that for defined conditions of heat treatment, the mean value of the volume transformed per filament reaches a maximum for the D and A rows of bundles that are in close contact with the peripheral layers of bronze. Such a result obviously arises from both the proximity and the total solute content of the reservoirs of tin within the composite. It must however be noticed that this is not the only reason of this lack of uniformity of the reaction; because if it was, the transformation should be more extended in the D than in the A bundle row. Just before the final heat treatment, the tin content of the immediate vicinity of the D row is indeed somewhat higher than in the case of the A layer [2] which should give rise to a higher kinetics of Nb$_3$Sn formation in the D row [5].

The degree of transformation of the filaments is also assisted by either a reduction or an asymmetry of their cross-section [2]. The first conclusion is obvious from the figure 7 data that exhibits the consequence of the variation of the mean filament cross-section according to its layer. Such a change of filament diameter "d" again arises from the hindering of the plastic flow by the Ta barrier. On the other hand, the plastic flow of a bulk bronze area is easier than that of a composite zone. Due to this feature, the cross-section of the filaments situated at the border of a bundle, is far more elongated than

Figure 4. Comparison between the mean deformation of the wire and that of its various components at increasing levels of applied deformation during the final stage of metal working. The inset indicates the cumulative strain of the wire.

Figure 5. FEM simulation of the shape of a tail of rod of the second design of composite during wiredrawing.

Figure 6. Comparison of the area of Nb transformed into Nb$_3$Sn per filament with the primitive area of the Nb filament.

Figure 7. Correlation between the filament cross-section and its degree of transformation.
that of the innermost filaments. Such an asymmetry reduces the thickness of niobium to transform, i.e. the length of atomic diffusion and the duration of the reaction in the short dimension of a filament. The outermost filaments of a bundle are therefore more transformed than those on the inner side.

3.2.3 Effect of cold work. Besides on the disposition of the components and the filament morphology, the kinetics of formation of Nb₃Sn further depends on the diameter and spacing of the filaments, and hence on the magnitude of the applied strain.

In a given condition of heat treatment (30h at 630°C + 120h at 690°C) the mean value “F” of the fraction of filament transformed into Nb₃Sn increases from 91 to 96% when “d” is reduced from 5.2 to 4.2 micrometers by an augmentation of the cold work strain. Such an auspicious effect of a diminution of “d” for the Nb₃Sn formation has already been established in the bronze and internal-tin cases [2]. It must however be noticed that a close examination of the results obtained with the wire of largest diameter clearly proves that this influence is of secondary importance compared to that of the volume and proximity of the Sn reservoirs. In this condition, and in contradiction with the effect of the filament diameter (see Fig. 6), F is actually a few larger in the more central A bundle layer than in the B one.

The filament spacing is another important parameter to control because of its effect on both the amount of solute atoms available for the Nb₃Sn formation and the actual expansion of the Nb₃Sn compound within the matrix. In this regard, the comparison of figures 8 and 9 shows that in case of partial transformation, the filament dilatation is strongly enlarged by an increase of its diameter “d”, and therefore of the filament interval. Moreover, two astonishing results are worthy to underline. First, in a fully reacted wire, the volume fraction of the Nb₃Sn filaments does not derive from that of the primitive Nb filaments. Secondly, a prolongation of the heat treatment of formation of Nb₃Sn entails an augmentation of the Nb₃Sn volume that does not match at all with the increment of the transformed volume within the filaments. All these results seem explainable by the influence of the filament spacing on, on the one hand, the stresses within the matrix, and on the other hand on the plastic deformation of the matrix. Taking account of the change of the crystal lattice from Nb to Nb₃Sn and of the simultaneous contraction of the matrix due to its Sn impoverishment, the transformation of the Nb filaments into Nb₃Sn generates hydrostatic compressive stresses within the matrix. The smaller the filament spacing or the thicker the Nb₃Sn layer, the higher these stresses should be with the consequence of an increase of their adverse effect on the filament expansion within the matrix. Such an expansion is on the contrary expected to be enlarged by an increase of the filament spacing that promotes the accommodation of the filament expansion by the plastic flow of the matrix. The latter assumptions can actually explain the experimental data. As a matter of fact, the comparison between the results obtained with the various bundle layers clearly shows that for the reduced filament spacing, the expansion of the filaments within the matrix correlates with the thickness of the Nb₃Sn layer but not with the filament spacing. The reverse is observed with the larger filament interval [2].

![Figure 8. Effect of cold work on the filament spacing.](image1)

![Figure 9. Consequence of cold work for the filament expansion.](image2)
3.2.4. **Superconducting behaviour.** The dispersion of the rate of formation of Nb$_3$Sn between the bundle rows depicted in figure 7 is prejudicial to the superconducting behaviour as it gives rise to distinct values of the Nb$_3$Sn grain size and therefore to various densities of flux pinning centres between the bundle rows. Although most favoured for the phase transformation because of both its proximity with the external bronze reservoir and the smallest filament cross-section, the D bundle layer is also the worst for flux pinning as it possesses the biggest grain size, for instance 125 nm against about 110 nm for the other layers in the 0.8mm-diametered wire (30h/630°C + 120h/690°C).

Otherwise, the present increase of strain during the last cold working sequence is strongly favourable to the non-Cu critical current density (Jc) at low magnetic fields up to 12T (see, Fig. 10a). Such improvements are due to both 1) a marked decrease of the mode and width of the distribution of the Nb$_3$Sn grain size that has to do at least with the existence of compressive stresses during its formation (see Fig 10b), 2) the reduction of the diversity of deformation of the filaments with the cold work strain and 3) to the significant reduction of the filament diameter that promotes the advancement of the Nb$_3$Sn formation. This enhancement is observed in spite of the trend towards columnar grains.

![Figure 10](image_url)

**Figure 10.** Effect of filament spacing on the non-Cu Jc vs. applied magnetic field curves (a) and on the histogram of the Nb$_3$Sn grain size (b).

4. **Conclusions**

The results obtained with the present not too demanding composite design and fabrication process clearly show that the uniform deformation of an Nb$_3$Sn composite manufactured by the restack bundle drawing process is not easily accessible. The success has mainly to do with the differences in mechanical behaviour of its components while the location of the components is of secondary importance. The component lay-out is on the contrary of the first importance for the kinetics of formation of Nb$_3$Sn and thereby for both the grain size and Jc. The kinetics of formation of Nb$_3$Sn increases more with the volume of the Sn sources and with a decrease of the distance between the sources and the filaments than with a decrease of the filament diameter. The consequent dispersion of reaction rate is deleterious for the critical current as it gives rise to a non uniform repartition of the flux pinning centres. By way of contrast, a decrease of the filament spacing that reduces the filament expansion within the matrix is the cause of a marked diminution of the Nb$_3$Sn grain size. At last the comparison of the experimental data with the results of the FEM simulations strongly proves the valuable contribution of this approach for the improvement of the metalworking process.

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