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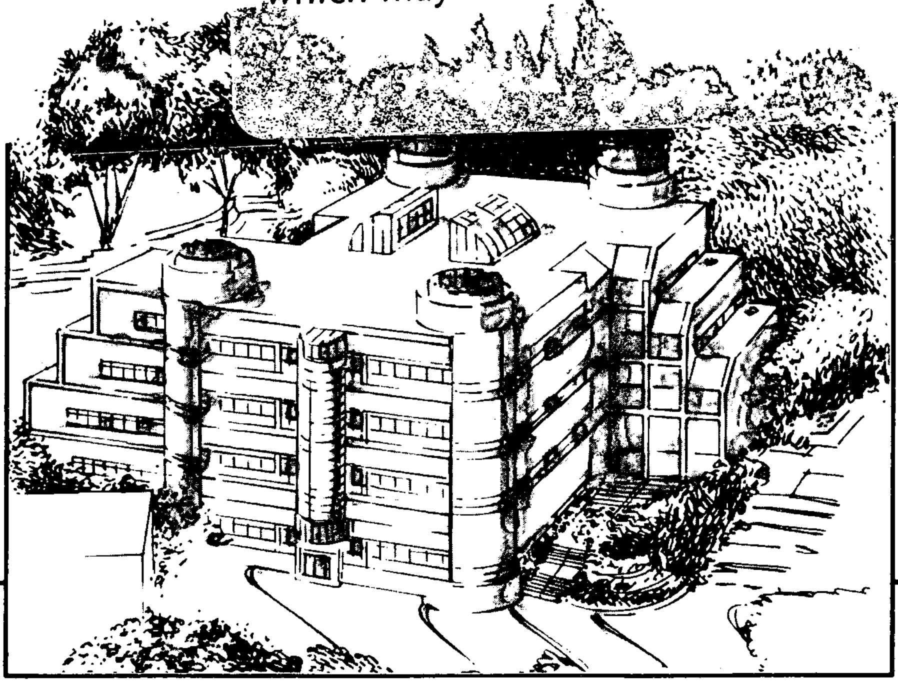

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# The $Nb_3Sn$ Grain Structure Evolution in Bronze-Processed Multifilamentary Superconducting Wire

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## SUMMARY

Two schemes have been proposed to explain the observed $Nb_3Sn$ layer morphology in bronze-processed superconducting wire. One attributes the morphology to the competition between nucleation and growth of $Nb_3Sn$ grains at the Nb interface while the other ascribes it to a break up of the columnar grains. Microstructural observations show that the latter scheme operates. Columnar grains develop very early during layer growth. With subsequent growth they retain approximately a constant length. Since the layer continues to grow this requires that the columnar grains formed earlier in the reaction break up. This process is responsible for the observed equiaxed grain morphology. The few dislocations observed in the columnar or equiaxed grain regions are present to accomodate crystallographic mismatch between grains. Recrystallization is not indicated since fine grains are not observed. The specific mechanism for the columnar grain break up remains unclear; however two classes of processes have been excluded, polygonization and recrystallization.

## INTRODUCTION

Due to the brittle nature of $Nb_3Sn$ an elaborate fabrication technique, the bronze process, has been developed to manufacture this material into wire form.[1] In its simplest form, elemental Nb rods are inserted into a Cu-Sn bronze billet and deformed to final wire size; subsequent heat treatment converts the Nb filaments to the superconducting $Nb_3Sn$. The grain structure of the $Nb_3Sn$ is critical in determining the current carrying capacity of the superconducting wire. Microstructural observations on typical $Nb_3Sn$ multifilamentary wires reveal several morphologically distinct regions within each $Nb_3Sn$ filament: a layer of small equiaxed grains located between columnar grains at the Nb interface and large coarsened grains at the bronze interface (Figure 1). The relative thickness of each of these regions varies with the particular conductor (e.g. bronze-to-niobium ratio, tin concentration in the bronze, third element additions, filament diameter) as well as heat-treatment temperature and time. Optimum current carrying capacity is expected with a uniformly fine grain structure.[2] A conductor with such an idealized microstructure is yet to be realized; first, an understanding of the morphological development of the $Nb_3Sn$ layer is essential.

Two schemes have been proposed to explain the grain morphology that develops in the $Nb_3Sn$ layer of a bronze-processed wire. One scheme relates the grain structure to the tin supply in the bronze.[1,3] A high tin concentration in the bronze, present early in the reaction, is thought to promote nucleation of new grains at the Nb-$Nb_3Sn$ reaction interface thus producing the equiaxed-grain region. Low tin concentrations, found late in the reaction, are expected to favor the growth of existing grains, resulting in a columnar grain structure. The second scheme attributes the equiaxed grains to a break up of columnar grains.[4,5,6,7,8] All grain growth at the reaction interface is columnar; those grains formed early in the layer growth process eventually break up. Through transmission electron microscopy (TEM) on sections of multifilamentary wires at various stages of heat treatment, i.e. reaction to $Nb_3Sn$, the probable operative scheme is determined. Additionally, this work proposes the driving forces and mechanisms responsible.

## EXPERIMENTAL PROCEDURE

The bronze-processed wire used in this investigation was produced by the Hitachi Corporation. The wire contains 10,261 filaments (31 bundles of 331 filaments) in a bronze matrix of 7.5at%Sn-0.4at%Ti. Each bundle is isolated from the copper stabilizer by a Nb diffusion barrier.

The overall diameter of the wire is 1.2mm. Individual sections of the wire were heat treated in quartz tubes that had been evacuated and backfilled with argon. The heat treatment temperature was fixed at 700$^{o}$C and the time varied from one hour up to eight days. TEM observations of the $Nb_3Sn$ layer were obtained from longitudinal sections of the wire. The wire was mechanically thinned to 0.05mm and ion milled to produce electron transparent regions.

## RESULTS AND DISCUSSION

Very early in the reaction (one to two hours), when the Sn concentration at the growth interface is near maximum, a well developed columnar grain morphology is present (Figure 2). The absence of small grains at the growth front indicates that nucleation of new grains has ceased (Figure 3). With further heat treatment to seven hours columnar grains of about the same length (as those observed at two hours) are still found at the Nb interface. The remaining layer is composed solely of equiaxed grains: the columnar structure present at two hours has reconfigured into a more equiaxed structure (Figure 4). Furthermore, the width of the equiaxed grains, specifically those nearest the columnar grains, is comparable to the short transverse diameter of the columnar grains. The layer growth rate decreases considerably after one day at 700$^{o}$C, probably due to a depleted Sn supply. Throughout the entire reaction process columnar grains remain at the growth front. Layer growth occurs through a widening of the equiaxed layer. Measurements are in progress to quantify the grain width change with distance from the growth interface.

Closer inspection of the microstructure shown in Figure 4 reveals a columnar grain apparently pinching off (marked). Additionaly, the equiaxed grains located directly above the columnar grain appear aligned with this columnar grain. Such a microstructure may result if the tails of the columnar grains pinch off thus forming an adjacent equiaxed layer.

Three possible sources for grain reconfiguration are: polygonization of dislocations, recrystallization, and columnar grain instability. Based on microstructural observations neither polygonization nor recrystallization appear viable mechanisms. Very few dislocations are observed; the few present probably accommodate crystallographic mismatch between adjacent grains (Figure 5). Typically, recrystallization and polygonization are associated with a very high dislocation density. The required dislocation density is not observed. Loss of dislocation density during specimen preparation is unlikely due to limited dislocation mobility in the very small grained $Nb_3Sn$. Additionally, fine grains, that would suggest recrystallization at the tails of the columnar grains, are not present.

There are at least two possible driving forces causing the columnar grain break up. One arises from the inherent instability of a long rod to decompose into spheres to reduce its surface energy. This phenomenon is observed frequently with fluid columns [9] and filaments of one phase in a matrix of another.[10,11] Similar phenomena can occur in single-phase materials but additional constraints must be considered. Another driving force arises from reaction stresses that are incurred on reaction to $Nb_3Sn$ at the $Nb/Nb_3Sn$ interface. $Nb_3Sn$ layer growth is the result of Sn diffusion along the $Nb_3Sn$ grain boundaries, and reaction with Nb at the $Nb/Nb_3Sn$ interface. This reaction produces ~35% volume increase (unconstrained), and the accompanying strain energy must somehow be accomodated.

A calculation of these stresses using a plane strain elastic model (assuming the bronze has a yield strength of zero and is perfectly plastic) yields the results in Figure 6. A state of triaxial tension is obtained in the Nb core while the $Nb_3Sn$ shell is in biaxial compression in the axial and circumferential directions, and in tension in the radial direction. The functional form of these results differs from those of Wallach and Evetts [7] and Pugh et al.[8] The stress magnitude in both models is unrealistically large. Unfortunately, a distinct mechanism can not be inferred from these stress results.

However, a global argument can be invoked based on Le Chatelier's principle. To lower the overall energy (i.e. strain energy) the microstructure will evolve in such a way as to relieve the stress. Since an equiaxed grain morphology most easily facilitates mass flow, the microstructure evolves toward that morphology from the columnar morphology formed initially.

## CONCLUSIONS

$Nb_3Sn$ grain nucleation stops early in the layer growth process. All subsequent layer growth occurs by growth of existing grains at the $Nb/Nb_3Sn$ interface resulting in columnar grains. A columnar grain break up accounts for the equiaxed $Nb_3Sn$ grains adjacent to the columnar layer. Microstructural observations indicate that the tails of the columnar grains pinch off thus resulting in the equiaxed grain layer. The details of the break up mechanism remain unclear; however two classes of processes have been excluded, yielding with polygonization and recrystallization. The probable driving forces for the break up process are reduction of surface energy and reduction of strain energy. Future experiments to ascertain the mechanism of columnar grain break up are planned.

## ACKNOWLEDGMENT

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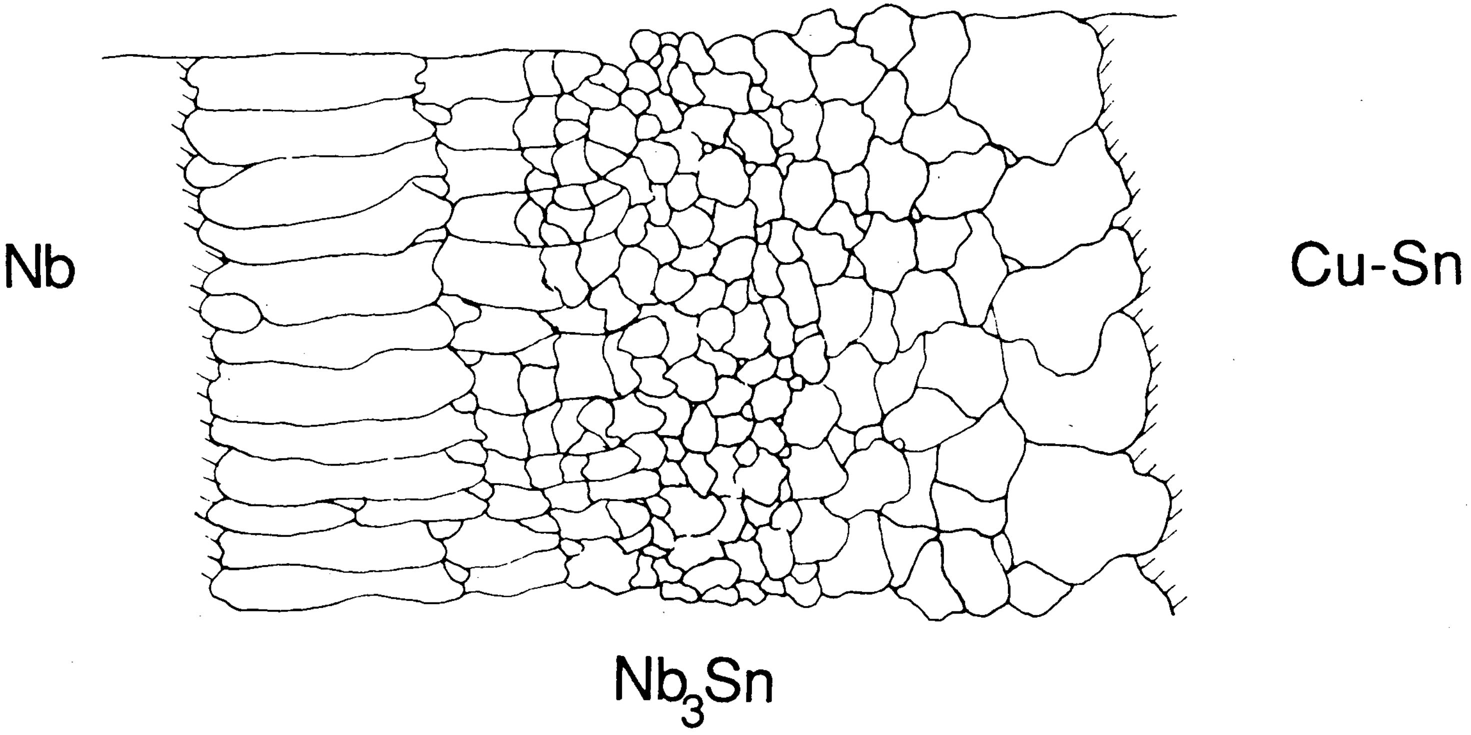

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Fig. 1 Schematic representation of the $Nb_3Sn$ grain morphology that forms in a bronze-processed wire.

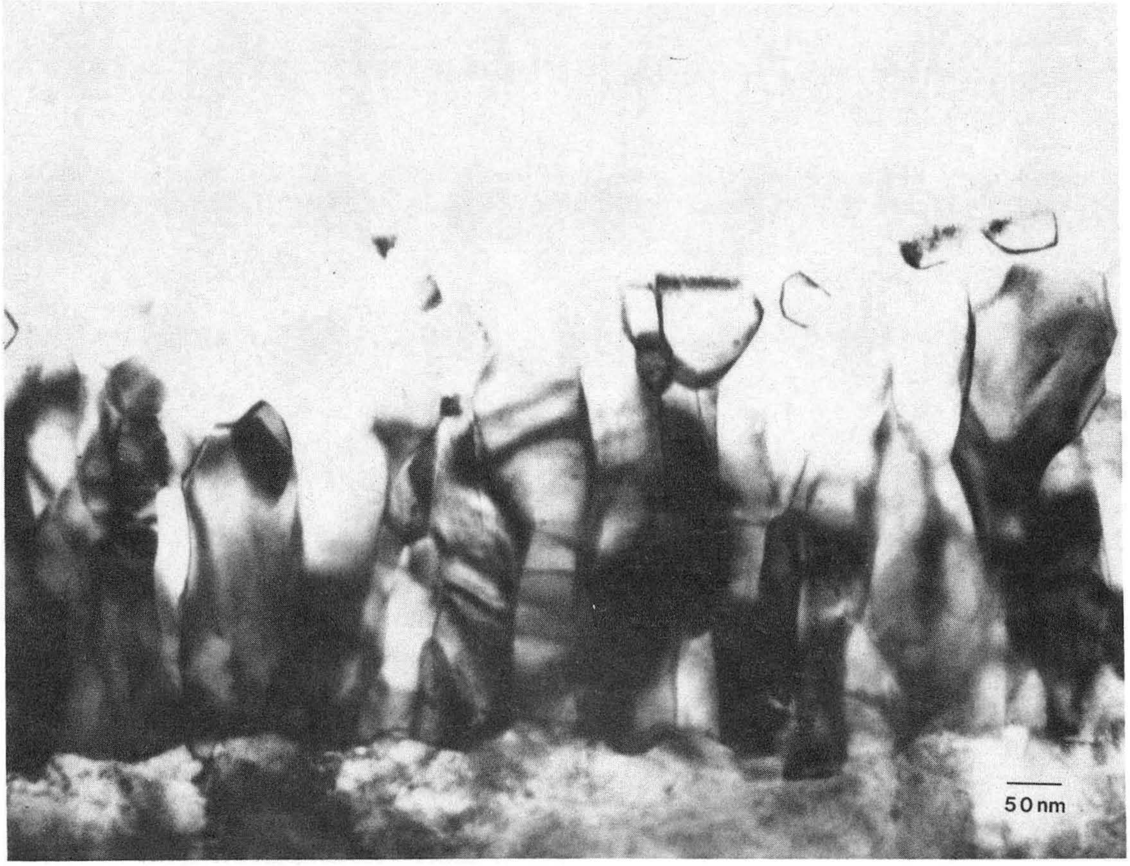

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Fig. 2 TEM micrograph of a sample heat treated at 700°C for 2hrs. Columnar grains are seen at the Nb/$Nb_3Sn$ interface.

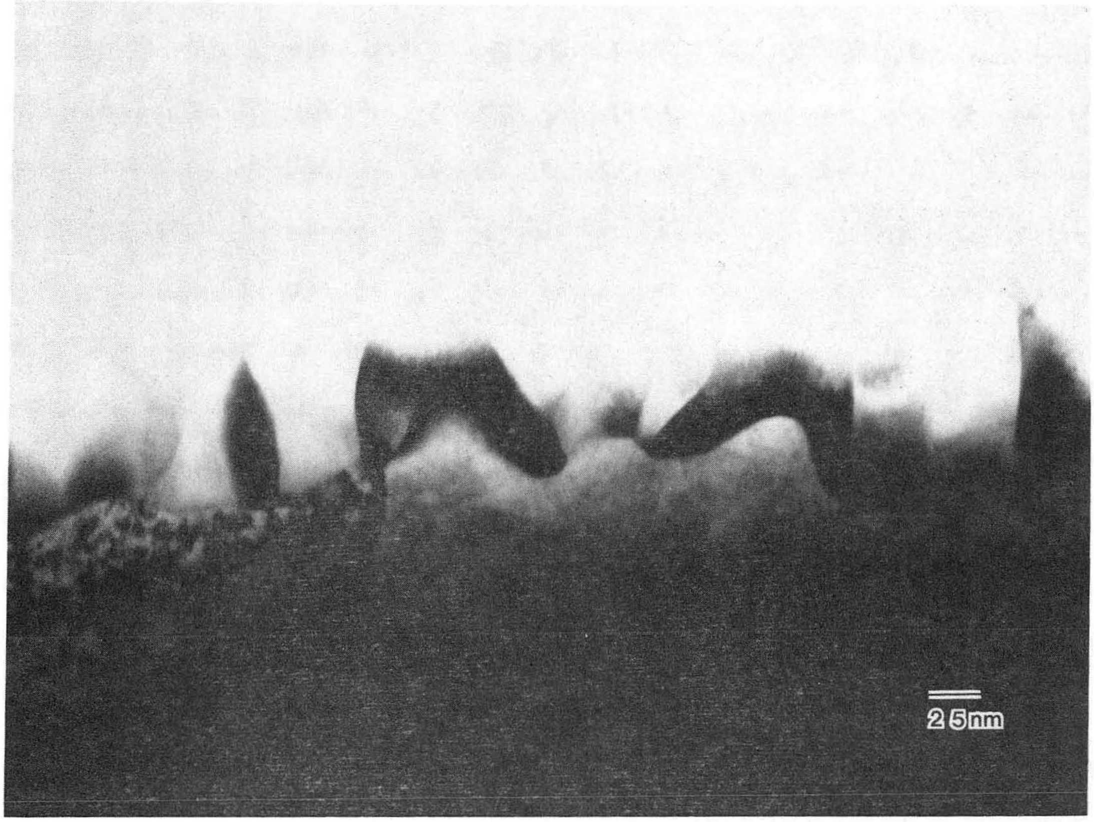

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Fig. 3 TEM micrograph of Nb/$Nb_3Sn$ interface. Columnar grains (top) growing into Nb (bottom).

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Fig. 4 TEM micrograph for a sample heat treated at 700°C for 7hrs. Columnar grains are seen at the Nb/$Nb_3Sn$ interface. The equiaxed grains have replaced the columnar grains present at two hours into the reaction.

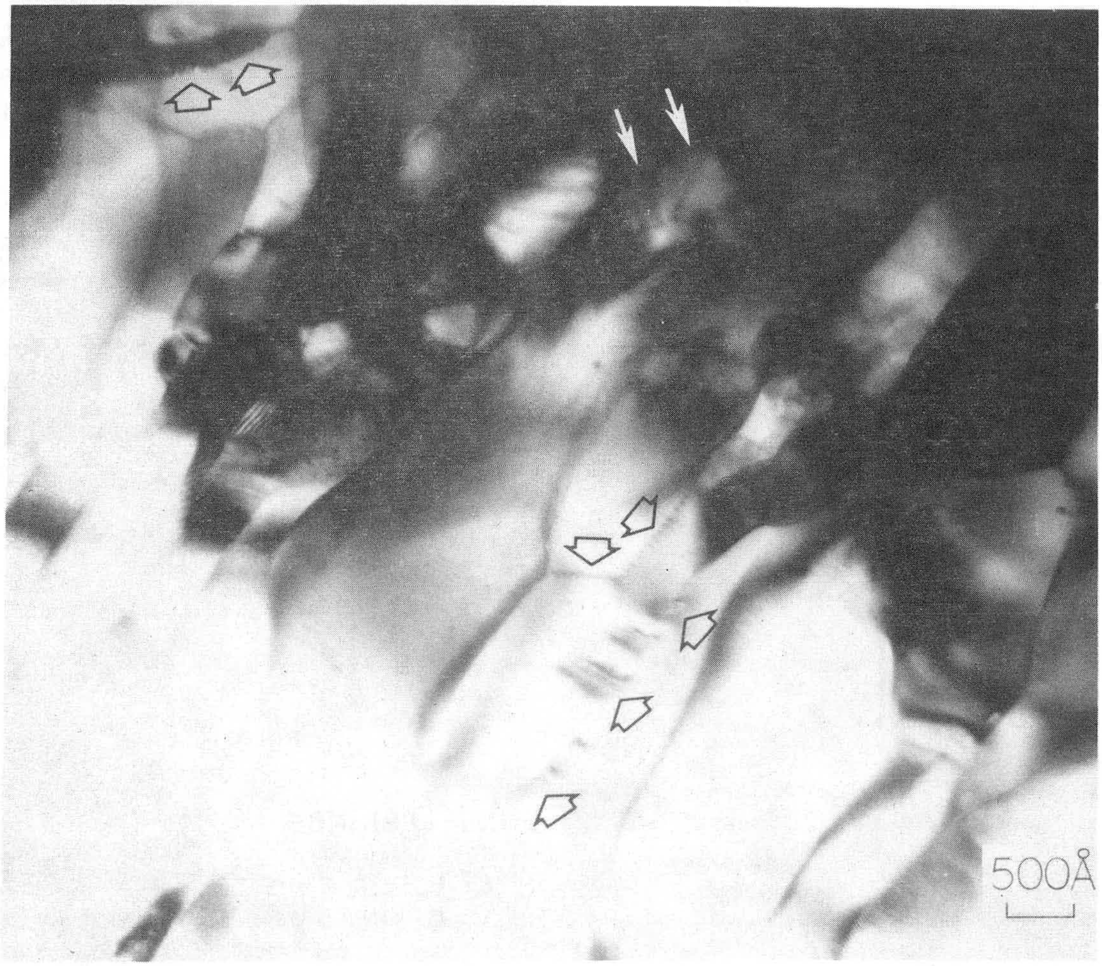

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Fig. 5 TEM micrograph showing dislocations at grain boundaries.

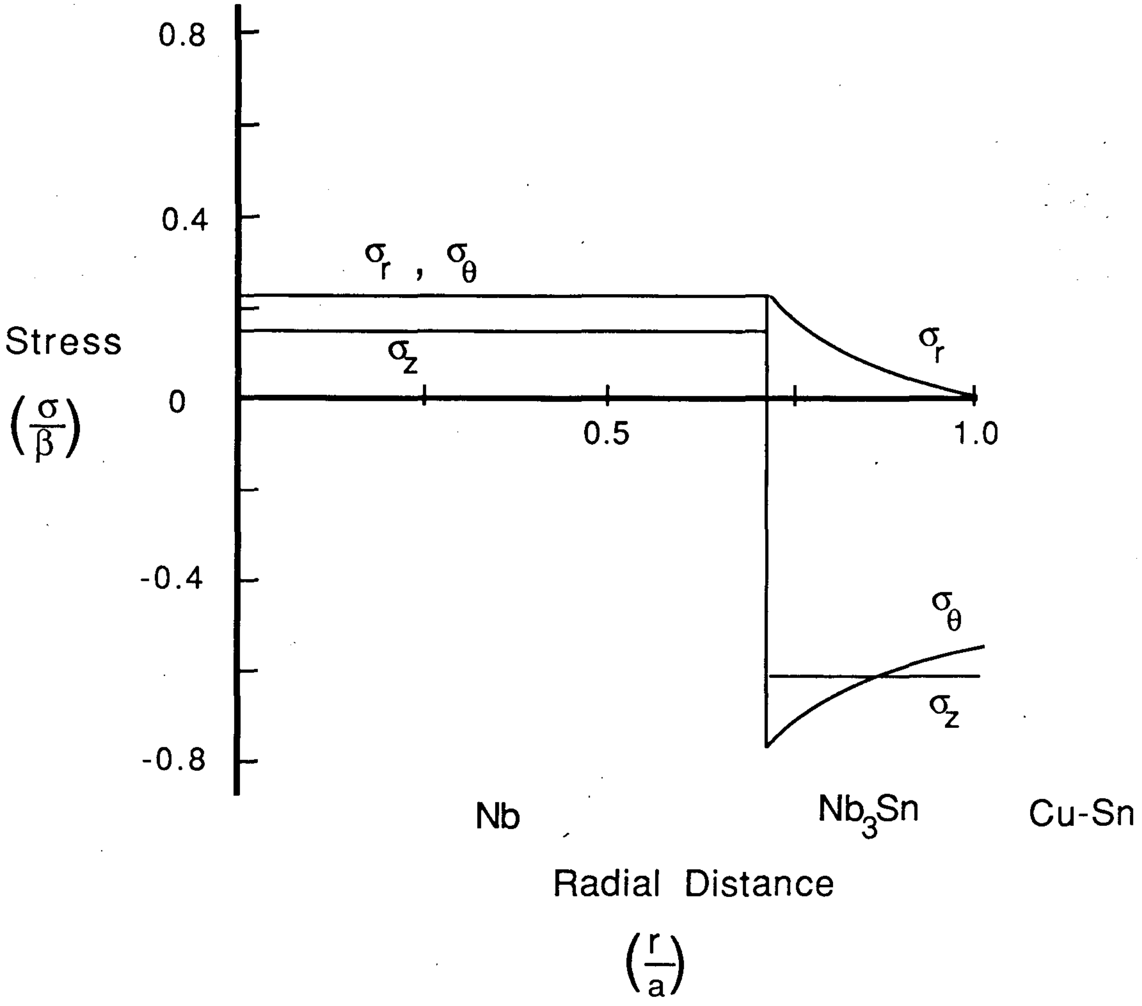

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Fig. 6 Normalized stresses versus normalized radial distance across a filament. The filament has a Nb core with a $Nb_3Sn$ shell. (Both regions are assumed to have the same elastic constants). $\beta$=12GPa, a=3μm.

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