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Conductors at Brookhaven National Laboratory

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STUDIES OF THE STRAIN-DEPENDENT PROPERTIES OF Al_5 FILAMENTARY
CONDUCTORS AT BROOKHAVEN NATIONAL LABORATORY*

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INTRODUCTION

Strain-dependent properties of Al_5 filamentary conductors can be divided into two broad categories of effects: elastic or "intrinsic" strain effects and irreversible behavior, usually associated with the formation of cracks in the Al_5 compound. The term "strain tolerance" of conductors is often used without properly distinguishing between the two types of effects with resulting confusion and conflicting claims from proponents of various types of conductors. This may be illustrated by a comparison between the behavior of Nb_3Sn and V_3Ga conductors. The intrinsic response of the "thermodynamic" superconducting properties, such as critical temperature and magnetic field, to elastic strain-produced changes in size and shape of the crystal lattice differs between the two compounds by nearly an order of magnitude, with Nb_3Sn having the larger response,¹ yet the degree to which the crystal lattice may be distorted from its equilibrium state before brittle fracture occurs is essentially the same for the two compounds. Therefore, when bronze-processed composite conductors are intentionally strained, the superconducting critical parameters of a V_3Ga conductor will be relatively insensitive to applied strain compared to a Nb_3Sn conductor. However, given equivalent initial strain states in the composite conductors, the applied strain at which irreversible behavior is observed will be essentially the same for V_3Ga and Nb_3Sn conductors, since the fracture strains of the two

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Al5 compounds are equal. Furthermore, the actual value of the applied strain at the onset of cracking is determined by the initial internal strain state of the composite conductor, and this must be properly accounted for in comparisons of the strain-tolerance of conductors.

In this paper, we will review briefly recent work at Brookhaven National Laboratory pertaining to the strain response of filamentary bronze-processed conductors. In this work we have touched upon all the aspects discussed above: the intrinsic strain dependence of the critical properties of Al5 structure compounds, the nature of the initial internal strain state of composite conductors, and the interplay between these residual strains and applied strains which governs the response of the conductor to external strain. We will also discuss briefly some factors which can enhance the strain tolerances of filamentary conductors.

INTRINSIC STRAIN DEPENDENCE OF Al5 COMPOUNDS

Filamentary bronze-processed conductors provide a unique opportunity to study the strain dependence of the superconducting properties of Al5 compounds, since the composite nature of these conductors allows much larger strains to be obtained before fracture than is possible with free-standing material. Historically the first experimental evidence of residual strains being associated with the composites took the form of measurements of the transition temperature, T_c , on monofilamentary conductors.^{2,3} The superconducting transition temperature showed significant changes when the bronze matrix was removed by etching. The data of Fig. 1 illustrate the changes in T_c observed in this type of experiment.⁴ The source of these changes in T_c is a compressive strain on the Nb_3Sn compound induced by the relatively larger thermal contraction of the outer bronze matrix, and increasing the matrix-to-core ratio increases the strain in the compound and consequently the depression of T_c , as seen in Fig. 1. A quantitative analysis of such experiments requires knowledge of the functional form of the dependence of T_c on strain and of the nature of the triaxial strain state in the composite.

The strain dependence of the critical temperature of a single crystal can be shown on rather general grounds to be expandable in powers of the strain components, and the number and form of nonzero terms of the series are determined by crystal symmetry.^{5,6} For a non-textured polycrystal in a cylindrically symmetric strain field this series, when truncated past quadratic terms and appropriately averaged with respect to orientation, reduces to¹:

$$T_c - T_c(0) = \Gamma_1 \epsilon (1 + 2\xi) + \epsilon^2 \left\{ \frac{1}{2} \Delta_v (1 + 2\xi)^2 + \frac{4}{15} \Delta_t (1 - \xi)^2 \right\} \quad (1)$$

Fig. 1. The superconducting transition temperature, inductive midpoint, as a function of the bronze-to-niobium ratio for a series of monofilamentary Nb₃Sn conductors. ■, represents as heat treated material; Δ, represents conductors whose bronze matrix has been removed by etching; o, etched material following a short anneal, 1/2 h at 700°C.

where ϵ is the axial strain in the compound, ξ is the ratio of the radial (or tangential) to the axial strain, and Γ_1 , Δ_v , and Δ_t are material constants, the first two of which describe the dependence on hydrostatic strain and the last the dependence on the non-hydrostatic or deviatoric components of strain and which must be measured experimentally or calculated from microscopic theory. A comparison of the efficacy of hydrostatic pressure and uniaxial strain in changing T_c shows that at least for Nb₃Sn and V₃Si and probably for V₃Ge and V₃Ga, the hydrostatic term, Γ_1 and Δ_v can be neglected in comparison with the uniaxial term Δ_t ¹; perhaps this behavior is generally true for Al₅ compounds. The essentially quadratic nature of the strain dependence is illustrated by the results of a series of experiments where inductive in situ T_c measurements were made while samples were strained in tension.^{7,8} The functional dependence of T_c on strain is shown in the normalized plot of Fig. 2. A parabolic strain dependence is evident over a wide range of strain, consistent with Eq. (1) when modified to account for the presence of both applied and residual strains.

Fig. 2. Normalized superconducting transition temperature data, T_c/T_{cm} , where T_{cm} is the value of T_c at its maximum, as a function of strain.

Whereas experiments on composite conductors to determine intrinsic strain properties are facilitated by the large elastic strains achievable in such composites, this is vitiated to some degree by a lack of precise knowledge of the strain state, especially in the non-axial components. (This same lack of knowledge of the strain state is a problem in developing models of the strain dependence for practical conductors but has usually been dealt with by ignoring the triaxiality. Recently progress has been made in the use of a computer to obtain numerical solutions of the triaxial strain state in conductors of complex geometry, as discussed by Scanlan et al.⁹) For the geometry of simple monofilament conductors, detailed calculations of the effects of triaxiality show that when Δ_{ϵ} is the only nonzero coefficient (i.e. only non-hydrostatic strain is important) in Eq. (1), only minor errors will be made by treating the strain state as uniaxial.¹ This is probably true for multifilamentary conductors as well; however it need not be the case when $\Delta_{\nu} \neq 0$ or for other geometries, e.g. "internal bronze" conductors.⁹

Bearing these qualifications about triaxiality in mind, the initial axial strain in the superconductor in a bronze-processed wire conductor is given approximately by¹:

$$\epsilon_0 \approx -\left|\frac{\Delta l}{l}\right| \frac{R_v C}{1+R_v C} \quad (2)$$

where $\Delta l/l$ is the fractional difference in thermal contraction, R_v is the volume ratio of bronze to core, and C is the ratio of effective "elastic" modulus of the bronze to that of the core. The latter ratio is an effective ratio of the moduli over the temperature range from the heat treatment temperature to the cryogenic testing temperature. This strain can be sufficiently large as to cause some plastic flow in the underlying unreacted niobium. Thus in Fig. 1 even after the bronze is removed T_c remains less than its unstrained value until a final stress relieving heat treatment is applied to remove the plastic set in the niobium.

Combining this estimate (Eq. (2)) for the initial strain in the compound with Eq. (1) (and neglecting the effect of hydrostatic strain and triaxiality) yields an expression for the critical temperature of the composite conductor:

$$\frac{R_v}{|T_c - T_c(o)|^{1/2}} = \frac{2}{C \left|\frac{\Delta l}{l}\right| |\Delta_t|^{1/2}} + \frac{2}{\left|\frac{\Delta l}{l}\right| |\Delta_t|^{1/2}} R_v \quad (3)$$

Replotting the experimental data of Fig. 1 indeed yields the straight line of Eq. (3), see Fig. 3. This result underscores the appropriateness of the preceding analysis.

The critical magnetic field of Al5 compounds is also a function of elastic strain. Considerations based on GLAG theory suggest that in cases where paramagnetic limitation is not a factor the dependence of the thermodynamic critical field H_c on non-hydrostatic strain should be essentially parabolic and should be stronger than the dependence of T_c because of the strain dependence of the renormalized electronic density of states and that the variation with strain of the upper critical field, H_{c2} , will be even stronger than that of H_c if the Ginsburg-Landau parameter κ_1 varies with strain. Such behavior is exhibited by Nb₃Sn, as shown in Fig. 4.

The results described above suggest that for bronze-processed filamentary Al5 conductors (excluding those produced by the "internal bronze" technique⁹), and probably for so-called in-situ conductors as well, the strain state can be treated as essentially one-dimensional with little error, and that the thermodynamic critical properties are essentially dependent on the square of the

Fig. 3. A straight line is obtained when $(R_V/\Delta T_C^{1/2})$ is plotted as a function of R_V . ΔT_C is the magnitude of the T_C depression caused by the residual stress in monofilamentary Nb_3Sn conductors and was obtained by etching off the bronze from a series of samples containing different bronze-to-niobium ratios; see Fig. 1.

total axial strain, residual plus applied. Thus applied tensile strains counterbalance the thermally-induced residual strains in such a way that the superconducting properties pass through an extremum, usually but not always a maximum, (essentially characteristic of the strain-free state) as a function of applied strain. The data in Fig. 2 for Nb_3Sn conductors are consistent with such a picture.

Understanding the intrinsic (reversible) strain dependence of the critical current density, J_C , requires the addition of yet

Fig. 4. The upper critical magnetic field, H_{C2} , of Nb₃Sn normalized by T_c , as a function of R_v . (H_{C2} was obtained by extrapolating high-field critical current data.) These data show that H_{C2} varies more strongly with strain than does T_c .

another level of theory: the theory of flux flow and its dependence on strain. Because of the influence of metallurgical factors on flux pinning, this is a more difficult task than understanding the strain dependence of the thermodynamic properties and much remains to be done in this area. An illustration of the differences in behavior of T_c and J_c can be seen by comparing Figs. 2 and 5; it is clear that the J_c data do not follow a simple quadratic variation with strain about the maximum.

Scaling laws, especially the one developed by Kramer for the high-field behavior of hard superconductors,¹⁰ have played an important role in correlating the critical current behavior of filamentary conductors, and several authors^{11,12} have used Kramer's scaling law to link the strain dependence of J_c with that of H_{C2} . If Kramer's high-field scaling law holds, the critical current in the strained state, J_c , will be related to that in the strain-free state, J_{cm} , (the subscript m being used to denote the strain-free state) by:

Fig. 5. Critical current as a function of applied tensile strain. Measurements made in situ at 4.0 Tesla for a series of monofilament conductors with varying bronze-to-niobium ratios.

$$\frac{J_c}{J_{cm}} = \frac{\kappa_{1m}^2}{\kappa_1^2} \frac{(H_{c2} - H)^2}{(H_{c2m} - H)^2} \frac{(H^{1/2} - \sqrt{\phi_o \rho_m})^2}{(H^{1/2} - \sqrt{\phi_o \rho})^2} \quad (4)$$

where $\kappa_1 (\equiv H_c/H_c)$ is the Ginsburg-Landau constant, ϕ_o is the flux quantum, and ρ is the density of flux-pinning sites. There are three material parameters in the scaling law: κ_1 (or H_c), H_{c2} , and $\sqrt{\rho}$. All in principle can vary with strain. Rupp¹¹ has successfully described the strain dependence of J_c for multifilamentary Nb_3Sn conductors for $H \gtrsim 8$ T by assuming that

$$(\kappa_{1m}/\kappa_1)^2 (H^{1/2} - \sqrt{\phi_o \rho_m})^2 (H^{1/2} - \sqrt{\phi_o \rho})^{-2} \propto H_{c2m}^2/H_{c2}^2,$$

which is equivalent to H_c and $\sqrt{\rho}$ being independent of strain or depending on strain in a compensating fashion. This leads to the simple equation:

$$\frac{J_c}{J_{cm}} \approx \frac{(1 - H/H_{c2})^2}{(1 - H/H_{c2m})^2} \quad (5)$$

We find that this does not describe the residual-strain dependence of J_c of monofilament Nb_3Sn conductors for $H=4$ T, as may be seen in Fig. 6.¹³

Empirically we find a linear relation between J_c/J_{cm} and $(H_{c2m}-H_{c2})$; see Fig. 6. A theoretical framework for discussing such a relation can be obtained from Kramer's high-field scaling law by expanding Eq. (4) to first order in ϵ^2 and replacing ϵ^2 itself by the equivalent value of $(H_{c2m}-H_c)$:

Fig. 6. The relationship between the residual strain-depressed critical current, I_{c0} , and upper critical field, H_{c20} , of as-processed Nb_3Sn monofilamentary conductors. The subscript m refers to the strain-free state. $H_{c2m} \approx 22T$. The solid curve is described by Eq. (5) and the dashed line is an empirical fit.

$$\frac{J_c}{J_{cm}} \approx + \left\{ \frac{H_{c2m}}{H_{c2m} - H} \frac{d \ln H_{c2}}{d \epsilon^2} - 2 \frac{d \ln \kappa_1}{d \epsilon^2} + \frac{\sqrt{\phi_o \rho_m}}{H^{1/2} - \sqrt{\phi_o \rho_m}} \frac{d \ln \rho}{d \epsilon^2} \right\} \frac{d \ln H_{c2}}{d \epsilon^2} \cdot H_{c2m} (H_{c2m} - H) \quad (6)$$

where the derivatives are taken at zero strain. Studies of Kramer-scaling in wires with the grain size of the specimens used to obtain the data of Fig. 6 indicate that the flux-pinning term (the last term in brackets) is probably negligible,¹⁴ while $d \ln H_{c2} / d \epsilon^2 \sim -23\%$ per $(\% \epsilon)^2$, as estimated from the data shown in Fig. 4. Thus the empirical relation shown in Fig. 6 can be interpreted with Eq. (6) to imply that $d \ln \kappa_1 / d \ln \epsilon^2 \approx -16\%$ per $(\% \epsilon)^2$, whereas the assumption of Rupp, $\kappa_{1m} / \kappa \approx H_{c2m} / H_{c2}$, leads to a value of -23% per $(\% \epsilon)^2$ for $d \ln \kappa_1 / d \epsilon^2$. Whether this discrepancy is real or a consequence of applying the Kramer scaling law at too low a field (4 T) for it to be applicable is not known. Furthermore the general applicability and literal interpretation of Kramer scaling is subject to some doubt.¹⁴ Clearly much remains to be done toward understanding the intrinsic strain dependence of J_c .

IRREVERSIBLE EFFECTS

When used in magnets, bronze-processed conductors must be limited to strains within the elastic range, an upper limit to this range is set by compound cracking and the onset of irreversible strain effects.¹⁵ Details of the association between compound cracking and irreversible strain behavior may be obtained from Ref. 15. One factor determining the cracking strain is the bronze-to-niobium ratio. Figure 7 illustrates the dependence of cracking and peak I_c strains on R_v . Both strains increase with increasing R_v values, saturating when R_v reaches ~ 10 . At this value all of the thermal contraction difference between bronze and niobium has been taken up in the compressed filament. Under the action of an applied tensile load, compression in the filament must first be counterbalanced before the filament can be strained into tension. As seen from Fig. 7 the tensile domain, that range of strain intermediate between the peak I_c strain and the cracking strain, remains essentially constant with increasing R_v values. Thus, by increasing the amount of residual compression larger R_v values enhance a conductor's overall tolerance to tensile straining.

In order to be used successfully in magnets these Al5 conductors must also possess reasonable room temperature bend-strain tolerances. In comparing critical current bend and tensile test

Fig. 7. Strains associated with peak I_c values and cracking (irreversible behavior) as a function of the bronze-to-niobium ratio for Nb_3Sn monofilamentary conductors heat treated 15 h at 725°C, 4.0 Tesla.

data it was found that tolerance to bending strains was somewhat less than expected on the bases of tensile test data.^{16,17} An example of the different responses of critical current to bending and tensile strains is presented in Fig. 8. As may be seen irreversible behavior sets in much sooner under bending strain.

We believe that the primary origin of the difference in strain tolerances is a shift in the neutral axis of the conductor during bending. Unless the neutral axis shift is taken into account, the calculated strain in the compound during bending is less than the strain actually applied. The shift occurs as a result of interaction between applied and residual strains. Because the matrix is in tension in the as heat treated condition, tensile strains associated with bending result in plastic flow of the tensioned

Fig. 8. Comparison of critical currents as a function of bending and tensile strains for a monofilamentary Nb_3Sn conductor heat treated 15.5 h at 725°C , 4.0 Tesla, $R_V=13$. The dot-dash line shows the tensile-strain dependence.

bronze before plastic flow occurs in the compression side of the conductor. The neutral axis is established by a force balance between the compression and tensile zones and thus with plastic flow in the tension zone, and consequently a lower effective modulus, the neutral axis will shift toward the center of bend curvature. This shift of the neutral axis means that the Nb_3Sn compound will be under a larger tensile strain than calculated by geometric considerations alone, and cracking under bending conditions will occur at seemingly lower applied bending strains than those associated with tensile tests.

A test of these ideas was made by constructing a model in which the critical current density in a filament is assumed to be given by an average of the strain-dependent critical-current density integrated over the cross section of the filament which is still uncracked, the fracture strain in the compound is assumed to be

the same as that found in tension, and the plasticity of bronze, i.e. the neutral axis shift, is accounted for. The dependence of critical current on bending strains was calculated and is compared with experimental data¹⁶ in Fig. 9. The agreement is reasonably good and basically confirms these ideas, although somewhat too large values are predicted at the peak in critical current. This is believed to be a result of an assumption in the calculation where the critical current is represented by two linear functions for the tensile and compressive domains rather than one continuous quadratic function.

ON IMPROVING THE STRAIN TOLERANCE AND OTHER MATTERS

As pointed out above, the strain tolerances of Al5 conductors may be increased by increasing the bronze-to-niobium ratio.

Fig. 9. Normalized critical current behavior as a function of bending strain. The solid curve represents calculated behavior including a shift in the conductor's neutral axis. The dashed curve is expected behavior should the compound not crack.

Unfortunately adding more bronze dilutes the conductor's overall current density. Some efforts have been made to metallurgically enhance the strain tolerances without diluting the overall current density. The most promising appears to be the alloy addition of Be to the Cu/Sn bronze.¹⁸ The high temperature strength of the bronze is increased through this approach thus eliminating any accommodating plastic flow in the bronze during cool down from the reaction heat treatment temperature. The net effect of plastic flow in the bronze is to minimize the amount of residual compressive strain induced in the Al₅ compound. Thus the Be addition, by virtue of the fact that plastic flow in the bronze is eliminated, can increase the amount of residual compressive strain without the necessity of increasing the bronze-to-niobium ratio. Figure 10 illustrates the effect of a 0.18 wt% Be addition to a Cu+11.2 wt% Sn bronze matrix. With this alloy addition all of the available difference in thermal contractions, ~ 1%, is found as a

Fig. 10. Superconducting critical current, normalized to the maximum critical current value, as a function of applied tensile strain at 4.2 K and 4.0 T. Sample A represents a Cu+11.2 wt% Sn bronze, sample B a Cu+11.2 wt% Sn+0.18 wt% Be bronze. Samples were heat treated for 15 h at 725°C.

compressive strain in the compound.

It is evident from the preceding discussions that the composite nature of bronze-processed Al5 conductors, in particular their internal residual strains, play an important role in determining strain tolerances. This is also true for mechanical properties. The existence of internal residual strains prohibits application of a simple law of mixtures. For each conductor with a different R_v value the contribution from the bronze constituent to the overall mechanical strength is different because of the different plastic-strain history and possibly different work hardening rate in each case. This results in anomalous stress-strain relationships.¹⁹ Figure 11 illustrates how the bronze component varies in its contribution to the conductor's overall mechanical strength. The bronze has a different stress-strain relationship depending on the bronze-to-niobium ratio, that is, the tensile prestrain in the bronze. This is an important consideration when developing models

Fig. 11. Derived²⁰ stress-strain relationships for the bronze constituents of a series of monofilament conductors. Measurements made at 4.2 K.¹⁹

of the mechanical properties of these composite conductors for use in predicting their behavior under straining conditions.

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