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Pinning mechanism in a continuous ultrafine Nb₃Al multifilamentary superconductor

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For Nb₃Al multifilamentary superconductors fabricated by a newly developed composite process using continuous ultrafine Al-based alloy cores and pure Nb matrix, J_c properties have been investigated in detail in regard to the size and morphology of the Al core. A significant enhancement in pinning force caused by reducing the Al core size and an anisotropy in J_c observed in a rolled tape have indicated that the Nb₃Al-matrix (or core) interface acts as a dominant pinning center of fluxoids.

Large-scale applications of superconducting materials require that the materials have high values of the upper critical field H_{c2} and a critical current density J_c as well as an excellent tolerance to large mechanical strains and magnetic field fluctuations. Among the advanced A 15 compounds, Nb₃Al is most promising as an alternative to Nb₃Sn multifilamentary wires, due to its excellent J_c performance at high magnetic fields and superior strain tolerance as reported in powder metallurgically processed Nb₃Al composites.¹ The short interdiffusion distance realized by a coreduction of Nb and Al powders could reduce the temperature to form Nb₃Al below 1000 °C, avoiding the drastic decrease in J. caused by grain coarsening. However, the powder process seems to encounter difficulties in the development of a conductor of low ac losses because of the discontinuity of Nb₂Al filaments. Recently, the authors have succeeded in fabricating a continuous ultrafine Nb₃Al multifilamentary conductor by a new composite process.2 Alloying the Al core with Mg, Ag, Cu, and Zn diminishes the difference in hardness between the Nb matrix and the Al core, and improves the workability of the Nb/Al multifilamentary composite, resulting in a successful fabrication of continuous ultrafine Al filaments embedded in the Nb matrix.3 Reacted wires at 700-950 °C show excellent critical properties involving J_c over 1.5×10° A/m² at 4.2 K and 10 T. Nb/Al-Cu composites showed the best workability, although its critical properties were slightly inferior to the Nb/Al-Mg or the Nb/ Al-Ag composites. Therefore, the present study has been carried out for the Nb/Al-Cu composite to study in detail the effect of the filament size on J_c , which seems to be a key to clarifying the pinning mechanism of Nb3Al with continuous ultrafine filaments.

The details of the sample preparation have been already reported elsewhere.³ An Al-2 at. % Cu alloy rod of 6.9 mm Ø was encased into a Nb tube of 7 mm i.d. and 14 mm o.d., where the atomic fraction of Al to Nb is 26.4%, if the Al alloy rod is replaced by the pure Al one. The resulting single-core Nb/Al composite was cold drawn by cassete-roller dies into a wire of 1.14 mm Ø and cut into short pieces. The 121 short, single-core wires were bundled in a Nb tube of 14 mm i.d. and 20 mm o.d., and the resulting 121-core Nb/Al com-

posite was cold drawn into a wire and cut into short pieces again. These procedures were repeated two times more, and finally a $1.8 \times 10^6 (121 \times 121 \times 121)$ -core Nb/Al composite with continuous ultrafine Al-2 at. % Cu alloy cores was fabricated. Scanning electron microscopy (SEM) revealed that ultrafine single cores less than 100 nm in diameter were clearly separated, and the cylindrical shape of the Al core remained unchanged drastically in contrast with ribbon-like filaments¹ in the powder-processed Nb/Al composite. The final composites with filaments 25 nm to 2.3 μ m in diameter were heat treated at 750 °C to form Nb₃Al through the diffusion reaction between the Nb matrix and Al alloy cores.

Critical temperature T_c and critical current I_c were measured by a four-probe resistive method. T_c was defined as the midpoint of the transition. I_c was defined as the current at which the second deviation of $d^2V(I)/dI^2$ (giving the distribution of critical current in the multifilamentary composite⁴) in the voltage-current characteristic reached a maximum. J_c was defined as I_c /S. S was the cross-sectional area of the 1.8×10^6 Nb/Al single-core composites (i.e., exclud-

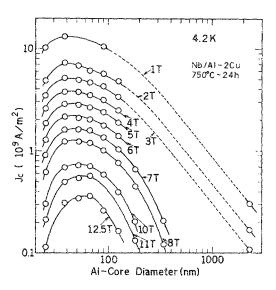


FIG. 1. Dependence of J_c on the calculated Al core diameter for composite wires reacted at 750 °C for 24 h.

2444

ing all Nb used for cladding). Therefore, S is the maximum cross-sectional area of Nb₃Al under the assumption that no other phase than Nb₃Al is formed by the diffusion reaction. H_{c2} was determined by the extrapolation of a Kramer plot.

Figure 1 shows the dependence of J_c at 1–12.5 T on the Al core diameter. With decreasing Al core diameter, J_c initially increases, reaches a maximum, and then decreases. The Al core diameter at which J_c reaches a maximum is shifted towards a smaller value by reducing the magnetic field. The improvement of J_c by reducing the Al filament size was also reported in the powder-processed Nb3Al, where the Al filament size was controlled by varying the initial Al powder size or the reduction ratio R of the composite. $^{1}J_{c}$ increased with increasing R and did not saturate at least up to R = 1400 for the composite with an initial Al powder size of 9 μ m. The final filament size is evaluated to 240 nm using the formula: (initial powder size)/ $R^{1/2}$, which is consistent with the results in Fig. 1. For the large-diameter Al core sample, the relative amount of Nb₂Al seems to be very small, because the Al supply is infinite and NbAl₃ forms preferentially. 5 However, the decrease in the Al filament size reduces the amount of intermediate compounds of NbAl, and Nb, Al due to the finite Al supply, and eventually increases the relative extent of Nb3Al, resulting in an improvement of J_c .

Similar Al filament size dependences are also observed for T_c and H_{c2} , respectively, as shown in Fig. 2(a). Maximum T_c and H_{c2} values are obtained at the same filament size. The Al core diameter at which T_c and H_{c2} reach maxima is apparently larger than those of the J_c peak for 1-8 T (Fig. 1). As shown in Fig. 2(b), T_c optimized by varying reaction time at a given Al core diameter is also depressed by the excess cold working. Degradation of T_c at small filament

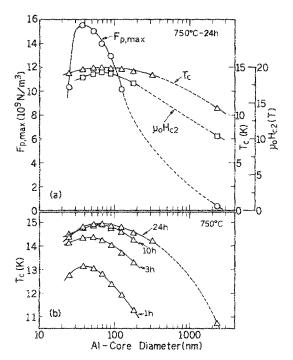


FIG. 2. (a) Variations in T_c , $\mu_0 H_{c2}$, and $F_{\rho, \max}$ as a function of calculated Al core diameter for composite wires reacted at 750 °C for 24 h. (b) Same variations in T_c for samples reacted for different periods.

sizes may be explained by the proximity effect. If the filament size is less than about $16\xi_s(\xi_s)$ is coherence length), the critical parameters of the superconductor filaments decrease rapidly with decreasing the filament size by the proximity effect. For the 38 nm Al core sample, ξ_s is estimated at 4.2 nm using the formula of $(h/4\pi eB_{c2})^{1/2}$, where h is the Planck's constant, e is the electric charge of electron, and B_{c2} is 18.6 T (Fig. 2). In this case, the diameter of Nb₃Al formed around the core is at most 76 nm, which is comparable to $16\xi_s$. On the other hand, the degradation of T_c at large filament sizes may be explained by the nonstoichiometry of Nb₃Al which forms simultaneously with the other intermetallic phases.

In the pinning force density F_p versus magnetic field curves at 4.2 K, a maximum F_p which is denoted by $F_{p,\max}$ was observed at 3–4 T for the Nb/Al-2 at. % Cu wire with H_{c2} of 19.3 T. As shown in Fig. 3, there is a clear difference in the field dependence of pinning force density between the present Nb₃Al composite and a commercially available multifilamentary Nb₃Sn wire, where the normalized peak field of Nb₃Al is 0.16, being somewhat smaller than 0.23 for Nb₃Sn. These differences in the respective f_p (normalized pinning force density) vs h (normalized field) curves suggest the different pinning mechanism between them.

The dependence of $F_{p,\max}$ on the Al filament size is also shown in Fig. 2(a). $F_{p,\max}$ is much more sensitive to the Al filament size than T_c and H_{c2} . The highest $F_{p,\max}$ of $1.6\times 10^{10}\,\mathrm{N/m^3}$ is obtained at an Al core diameter of 38 nm, which is much smaller than that for T_c and H_{c2} (70 nm). For the ultrafine multifilamentary Nb-Ti conductor, Hläsnik reported that T_c began to fall with decreasing filament sizes from 135 nm due to the proximity effect, while J_c continued to increase until 46 nm because of an increase of the surface current contribution. The shift of J_c and $F_{p,\max}$ peaks toward smaller Al filament sizes is probably also due to the enhancement of Nb₃Al-matrix(core) interface pinning which is caused by reducing the composite.

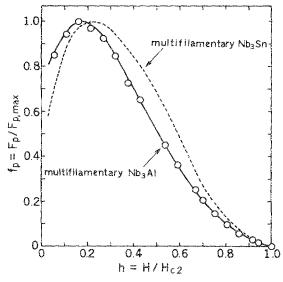


FIG. 3. Normalized pinning force density vs normalized magnetic field curves for a Nb/Al-2 at. % Cu composite with an Al core diameter of 38 nm reacted at 750 °C for 24 h and a commercially available multifilamentary Nb₃Sn wire (see Ref. 7).

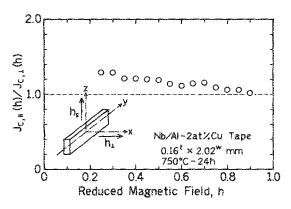


FIG. 4. Anisotropy in J_c , defined as $J_{c,\parallel}$ / $J_{c,\downarrow}$, as a function of reduced magnetic field for a composite tape reacted at 750 °C for 24 h.

To verify the role of interface pinning, a tape conductor 0.16 mm in thickness and 2.0 mm in width was prepared by rolling a 1.8×10^6 core composite wire of 0.7 mm \varnothing , because it could induce a morphology and configuration change of the interface on which J_c was expected to depend strongly. SEM observation revealed ribbon-like filaments aligned parallel to the rolling plane. If the Nb₃Al-matrix (or core) interface acts as a dominant pinning center, strongest pinning should occur when the fluxoids are parallel to the tape surface, just like for an in situ Nb₃Sn(V₃Ga) tape conductor. As shown in Fig. 4, J. for magnetic fields parallel to the tape surface, $J_{c,\parallel}$, is larger than that for fields perpendicular to the surface, $\hat{J}_{c,1}$, supporting the above speculation. A small contribution of the grain boundary pinning can be explained by the following: (1) the grain size of Nb₃Al is estimated to be comparable to the filament size, ^{5,10} (2) the elementary pinning force at the interface is in general much stronger than that at the grain boundary.

Since H_{c2} is slightly higher by about 1 T when the field is parallel to the tape surface, the anisotropy factor of J_c , defined as $J_{c,\parallel}/J_{c,1}$, is plotted against reduced magnetic fields. The origin of anisotropy in H_{c2} might be clarified if the preferred orientation of the Nb₃Al crystal could be measured. It is noted that the anisotropy in J_c and H_{c2} reported in the present letter should be advantageous when the multifilamentary Nb₃Al conductor with a large aspect ratio would be wound into the double-pancake coil.

In the present study, J_c has been found to depend strongly on Al core diameters which play important roles in varying both the Nb₃Al formation rate and the surface current contribution. However, J_c is also known to depend on various fabrication parameters³ such as additive elements, mechanical strain, heat treatment conditions, etc. Therefore, it is not so easy to optimize J_c , and the highest J_c of 1×10^8 A/m² at 18.5 T is obtained, at least hitherto, for the Nb/Al-5 at. % Mg composite wire with 90 nm Al core reacted by a two-stage reaction11 consisting of a first reaction at 950 °C and a subsequent second reaction at 700 °C.3 To understand the pinning mechanism in more detail, microstructural studies must be carried out in the future by using transmission electron microscopy and x-ray diffraction, which can reveal the kinetics of the Nb3Al formation and its nature such as the composition and atomic long-range order parameter.

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