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Possible solution of the grain-boundary problem for applications of high- T_c superconductors

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It is shown that the critical current density of high- T_c wires can be greatly enhanced by using a three-fold approach, which consists of grain alignment, doping, and optimization of the grain architecture. According to model calculations, current densities of 4×10^6 A/cm² can be achieved for an average grain alignment of 10° at 77 K. Based on this approach, a road to competitive high- T_c cables is proposed. © 2002 American Institute of Physics. [DOI: 10.1063/1.1516831]

Vital for large scale applications of high- T_c superconductors^{1,2} is the solution of the grain-boundary problem, which manifests itself by the exponential decrease of the grain boundary critical current density J_c of the high- T_c cuprates as a function of the grain-boundary angle.^{3,4}

We propose to solve this problem using a three-fold approach: Through (a) grain alignment,³ (b) grain-boundary doping,⁵ and (c) optimization of the microstructure to maximize the effective grain-boundary area.⁶ In contrast to the powder-in-tube technology where large grain-boundary areas and grain alignment are used to enhance J_c ,⁷⁻⁹ today's coated conductor technologies¹⁰⁻¹² focus on grain alignment only. As we have shown, however, simple ways exist to also preferentially dope the grain boundaries⁵ and engineer large effective grain-boundary areas^{13,14} to further enhance the performance of coated conductors.

As pointed out in 1987, large effective grain-boundary areas can be realized by engineering the microstructure of the superconductor to obtain grains with big aspect ratios, for example, by stacking in a brickwall-type manner plateletlike grains on top of each other.^{6,7} The enhancement of the critical currents hereby gained is responsible for the large J_c of the Bi-based high- T_c superconductors fabricated with the powder-in-tube technology.⁶⁻⁹ Recently, we found ways to use large effective grain-boundary areas to enhance J_c of coated conductors that consist of two- or three-dimensional grain-boundary networks, as illustrated in Fig. 1. Although it is clear that each one of the three techniques described substantially enhances J_c , the increase that can be gained by utilizing all three, for example as shown by Fig. 2, is unknown.

Therefore, we have calculated the performance, which can be achieved by combining grain orientation, doping, and large effective grain-boundary areas. Based on these calculations, optimized sets of parameters for the fabrication of coated conductors are derived.

The calculation of current percolation through disordered networks of weak links, some of which may be Josephson junctions, is a complex problem,⁶ and several algorithms have been developed for its solution (see, e.g., Refs. 6

and 15-19). As the fast algorithms are limited to two-dimensional networks, for the present work an alternative had to be devised. Like in several of the existing algorithms, to achieve the required speed, phase effects and self fields were neglected.

These calculations are based on Dijkstra's shortest path algorithm for undirected graphs,²⁰ the grains acting as vertices, the grain boundaries, weighted by their critical currents, as edges. To initialize a calculation, a polycrystalline superconductor, typically containing 10^3-10^4 grains is modeled first. To this superconductor, an intragrain $J_{c,\text{grain}}$ and misorientation angle dependent grain-boundary critical current densities $J_c = J_c(\theta)$ are ascribed. The calculations are performed in steps, in each step, i the algorithm finds in Dijkstra's sense the shortest path through the network. The critical current I_i of this path and the respective current densities are calculated and compared to the current densities of the grains and boundaries that form this path. At the end of step i , the critical currents of the grains and grain boundaries involved are reduced by I_i . After the final step m , all possible current paths have been cancelled by this procedure and the critical current I is given by $I = \sum_{i=1}^m I_i$. These calculations are repeated N times to calculate J_c for different networks. The final result is obtained by averaging the intermediate results.

Because the algorithm uses an undirected graph model, it is fast and can determine the critical current of three-dimensional grain-boundary networks. Its accuracy is a func-

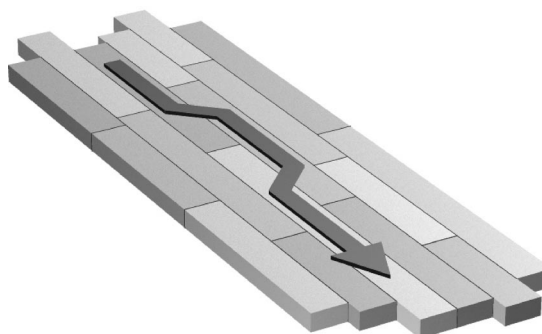


FIG. 1. Sketch of a coated conductor containing grains with big aspect ratios. Large currents are supported by the conductor, because bypasses around the standard, small area grain boundaries are provided.

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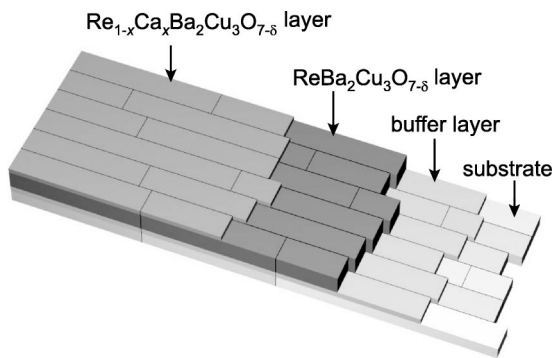


FIG. 2. Simplified sketch of a tape, fabricated by the rolling assisted biaxially textured substrate (RABiTS) technology, with a modified grain architecture which is based on grains with aspect ratios $\rho \gg 1$. Preferentially doping of the grain boundaries is achieved by utilizing a doped cap layer and grain-boundary diffusion.

tion of the grain number and of N . The numerical accuracy of the data presented, typically obtained with 10^3 grains and $N=20$, is better than 5%.

This algorithm was used to assess possible approaches for the optimization of coated conductors at 77 K. For the present calculations, the intragrain J_c was taken to be 5×10^6 A/cm², and the grain orientations and lengths were chosen using Gaussian distributions with parameterized widths (see also Ref. 13). To determine the effects of doping on I_c the $J_c(\theta)$ dependence in the simulation was modified based on the experimental data,⁵ which show that at least in the range of $24^\circ < \theta < 36^\circ$, the critical current density can be doubled by doping. For lower and higher angles, J_c is taken to be exponentially reduced to the undoped values.

To consider the effects of stacking coated conductors in multilayer configurations, the intergrain critical currents flowing in the c direction were modeled by reducing $J_c(\theta)$ by an additional c -axis coupling factor f_c . For bilayers, the transverse misalignment of the grains in the top and bottom layer was taken to be 30% of the grain width. These calculations provide a clear assessment of the possibilities to optimize coated conductors, as shown in the following.

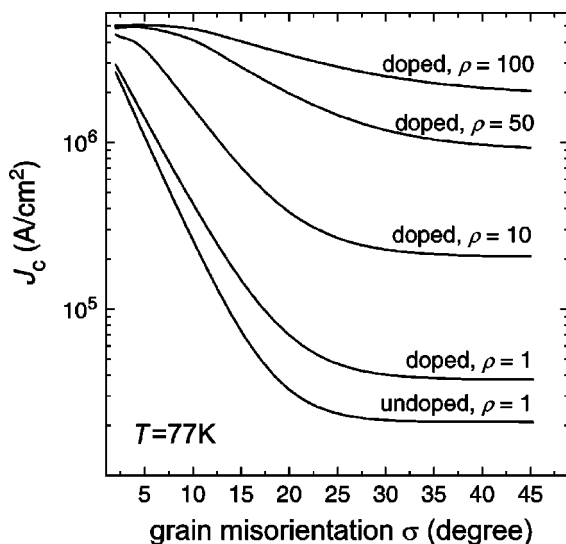


FIG. 3. Calculated critical current densities of various RABiTS tapes with different grain aspect ratios ρ as a function of the grain misorientation σ .

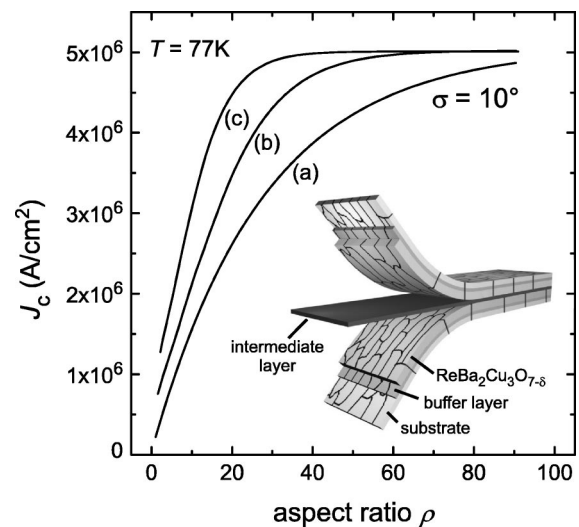


FIG. 4. Calculated dependence of the critical current density as a function of the aspect ratio ρ of two RABiTS tapes with misorientation $\sigma = 10^\circ$ stacked on top of each other with different coupling factors $f_c = 0$ (a), 10^{-4} (b), and 10^{-3} (c). The inset shows a sketch of such a tape, the intermediate layer is used to weld the two tapes together.

In Fig. 3, the critical current densities of various coated conductors are plotted as a function of the average grain misorientation σ and aspect ratio ρ of the grains. As seen, the current density of conventional tapes ($\rho = 1$, $\sigma \geq 15^\circ$) is approximately doubled by doping the grain boundaries, in agreement with experimental results. An enhancement of the aspect ratio significantly increases I_c further, and a tape with $\sigma = 45^\circ$ and $\rho = 50$ has the same J_c as conventional tape with an alignment of 6° . This graph suggests combining moderate grain alignment ($\sigma = 10^\circ$), large aspect ratios ($\rho = 20$ to 30), and doping to achieve critical current densities of $3\text{--}4 \times 10^6$ A/cm².

As revealed by Fig. 4, in which J_c is plotted as a function of aspect ratio ρ and coupling f_c for two undoped rolling

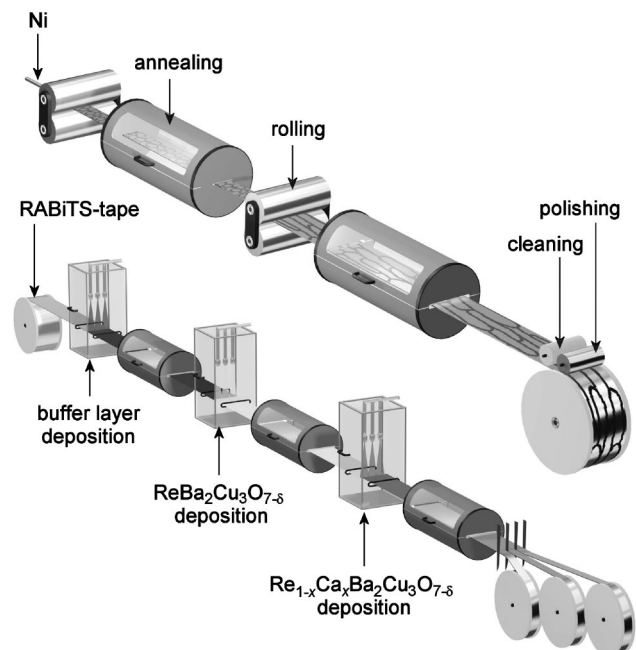


FIG. 5. Sketch illustrating large scale production of $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$ -based RABiTS tapes as motivated by the results shown in Figs. 3 and 4.

assisted biaxially textured substrate (RABiTS) tapes with 10° texturing stacked on top of each other, the use of bilayers increases J_c roughly by 10^6 A/cm². Bilayers therefore only provide a significant advantage for conventional tapes with $\rho \approx 1$. The benefits are much smaller for tapes with larger aspect ratios and it seems reasonable to apply only either stacking or elongation of the grains for a given tape.

The perspectives opened by the data shown in Figs. 3 and Fig. 4 suggest the mass production of high- T_c tapes, as illustrated by Fig. 5. This process is based on the standard RABiTS technology, and can generate tapes with high critical currents by utilizing grain-boundary doping and tapes containing grains with big aspect ratios. Here, it is envisaged that Ni tapes with elongated grains and proper texture can be fabricated, using processes as reported in Ref. 21. A RABiTS tape is rolled and annealed to obtain an average grain misorientation of $\approx 10^\circ$ and aspect ratios in the range of 20–50. The buffer layer system and the $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film are deposited with nonvacuum techniques,²² before the tape is covered with a cap layer of $\text{Re}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$, annealed and cut. According to Fig. 3, such tapes support critical current densities of 4×10^6 A/cm², corresponding to critical currents of 400 A/cm for 1 μm thick, single-sided superconducting films. Although significant technological problems associated with the growth of thick superconducting films, ac losses, and quench protection remain to be solved, the process outlined appears to be suited for large scale production of coated conductors with large critical currents.

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