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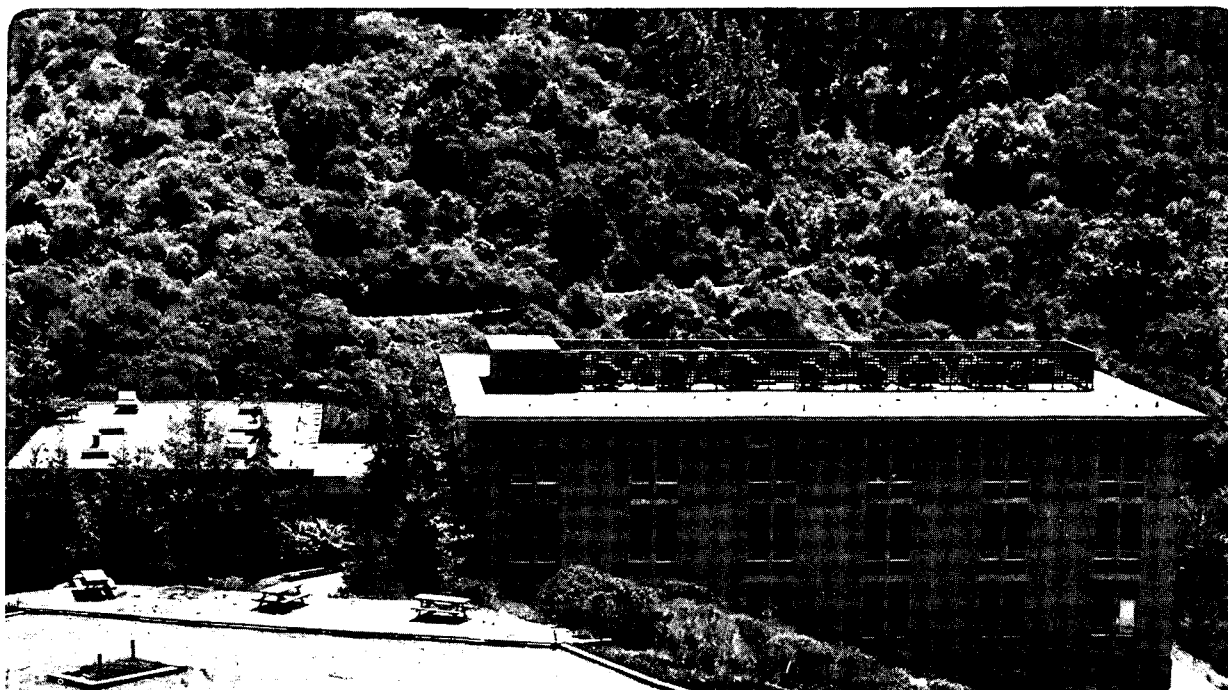
## Materials & Chemical Sciences Division

Submitted to Journal of Materials Science Letters

### Traveling Solvent Zone Texturing of Ceramic Superconductor Thick Films

T.J. Richardson and L.C. De Jonghe

June 1990



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Traveling Solvent Zone Texturing of  
Ceramic Superconductor Thick Films

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Supported thick films of superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  (YBCO) have been prepared using a modified traveling solvent zone technique. The films consist of a strongly-oriented, single superconducting phase with excellent connectivity and unusual microstructure. The technique utilizes a volatile flux which melts and evaporates as the film passes through a steep temperature gradient furnace.

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Large-scale practical applications of high-temperature ceramic superconductors await development of bulk conductors capable of carrying currents in excess of  $10^5 \text{ A cm}^{-2}$  at 77K. Attempts to produce wires and tapes with high critical current density have thus far met with considerable difficulty. Unsupported ceramic conductors are brittle; they cannot be wound in tight coils, nor will they stand up to the repulsive forces within coils and solenoids generating strong magnetic fields. Thick (25 to 1000  $\mu\text{m}$ ) superconducting films supported by ceramic substrates offer greater structural stability and can be laid down in complex patterns by screen printing and/or etching techniques.

Due to the strongly anisotropic nature of the superconducting characteristics of YBCO, it is generally accepted that highly textured material is required for efficient transport of intergranular currents.<sup>1-3</sup> Well-oriented thin films have been prepared which exhibit critical current/field behavior similar to that of a good single crystal.<sup>4-6</sup> The required vacuum equipment and low deposition rates, however, limit the use of thin film techniques to electronic device applications. Texture must therefore be developed in thick films after they have been applied to the substrate. Since undesirable non-superconducting phases precipitate at the high temperatures required to melt YBCO, melt-texturing<sup>7-12</sup> produces highly aligned but inhomogeneous regions. The length of samples textured by conventional zone melting<sup>13</sup> is limited by the accumulation of secondary phases in the melt. Here we report a modified traveling solvent zone (TSZ) technique for producing highly-oriented thick films of YBCO on a ceramic substrate.

By use of an appropriate solvent, the melting point of the superconductor/solvent mixture is reduced to the regime of thermal stability for YBCO. If the solvent is volatile, the melt solidifies as the solvent evaporates. The crystalline superconductor solute precipitates with grain alignment controlled by the preferred growth direction (normal to the c-axis in the case of YBCO) and the solvent concentration gradient. By passing the precast YBCO/solvent mixture through a steep temperature gradient furnace, dissolution and precipitation take place in a traveling solvent zone, producing a continuous, homogeneous, YBCO superconductor film.

Single-phase YBCO, prepared by the standard oxide route<sup>14</sup>, was ball milled to an average particle size of 1  $\mu\text{m}$ . An equal volume of potassium chloride was added to the superconductor powder and the mixture was ground by hand to ensure thorough mixing. A paste was made from the powder mixture by adding iso-amyl alcohol to achieve the desired consistency. A film 200  $\mu\text{m}$  thick was prepared on a 30 mm x 3mm x 1 mm thick sintered MgO plate by the doctor-blade method. After drying at 100 °C, the film and substrate were passed lengthwise through a resistively heated steep temperature gradient furnace (50°C mm<sup>-1</sup>, maximum temperature 950 °C) at a constant rate of 0.025 mm sec<sup>-1</sup>. The film melted and then re-solidified as the solvent evaporated from the hot zone, producing a porous, sponge-like solid film of essentially phase-pure YBCO.

Scanning electron micrographs of the film surface (Figs. 1 and 2) show the characteristic layered morphology of YBCO with the layers parallel to the substrate. Small amounts of an impurity phase (probably BaCuO<sub>2</sub>) are left on the surface as the last of the solvent evaporates (Fig. 1). The

interior, however appears to be free of impurities. Regions of high density are observed (Fig. 3), particularly near the substrate. The sponge-like structure, shown in cross section in Fig. 4, is due to uncontrolled evaporation of solvent from the molten film. The x-ray diffraction pattern (Fig.5) taken normal to the film surface consists primarily of intense, sharp peaks corresponding to the 00l reflections of YBCO, thus confirming both the purity and the strong c-axis orientation.

The film adheres well to the substrate, is continuous over about 20 mm, and is free of large cracks. Microcracking is not observed even after annealing in oxygen at 450 °C for 10 h to ensure complete conversion to the superconducting orthorhombic structure. Grain boundaries are not visible in the relatively dense interior of the film, indicating good electrical continuity. Efforts are under way to refine the processing parameters to achieve reduced porosity and high critical current densities. The modified TSZ method offers considerable promise for preparation of coherent cuprate superconductor thick films for large-scale applications.

This work was supported by the Electric Power Research Institute and by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division, of the U. S. Department of Energy under Contract No. DE-AC03-76SF00098.

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## FIGURE CAPTIONS

Fig. 1. Surface of traveling solvent zone (TSZ) processed YBCO thick film viewed from directly above the surface. The surface impurity phase is believed to be barium cuprate.

Fig. 2. Side view of TSZ film surface. The solvent zone traveled from right to left during processing.

Fig. 3. Close-up of dense region. Note the characteristic step structure of YBCO.

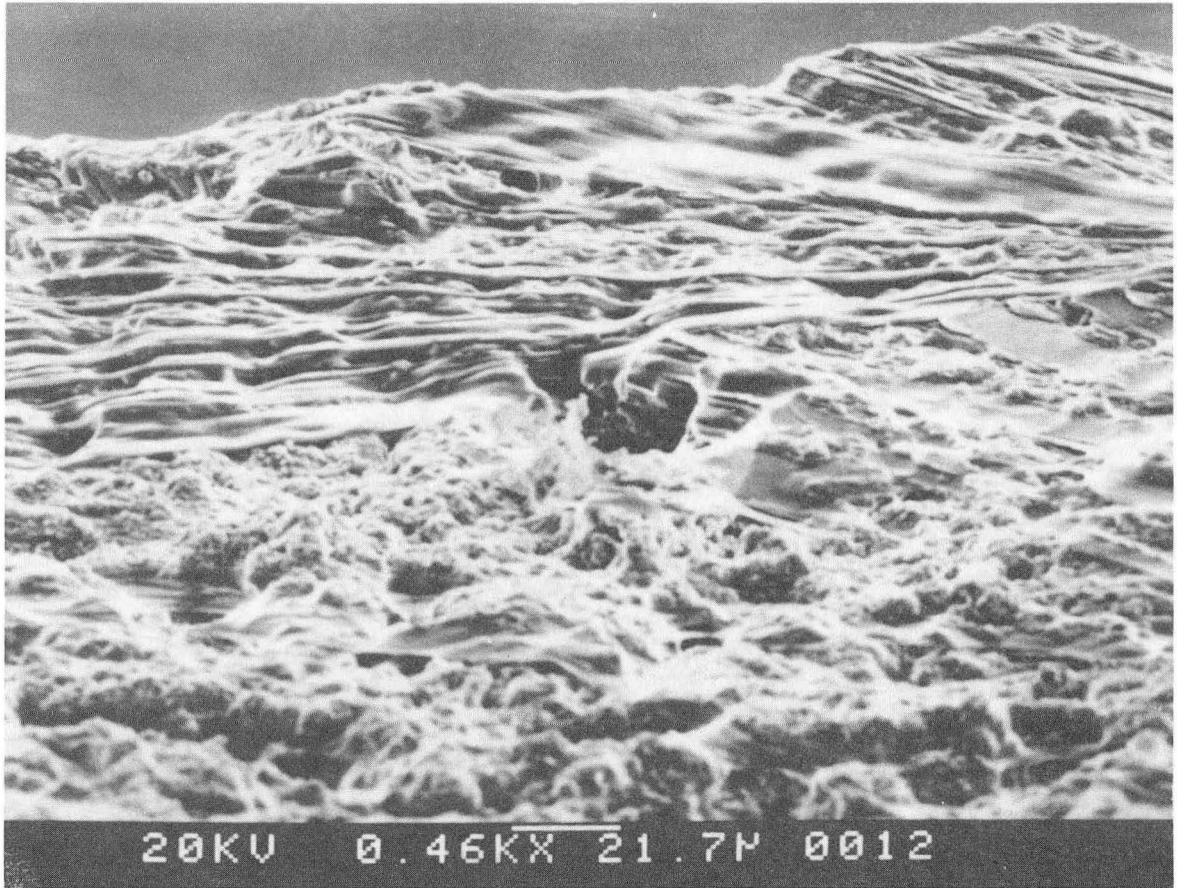
Fig. 4. Cross section of TSZ film showing high porosity and vertical expansion due to uncontrolled solvent evaporation. The dark area at the bottom is the magnesia substrate.

Fig. 5. X-ray diffraction pattern of TSZ film showing predominance of 001 reflections due to c-axis orientation normal to the film surface.



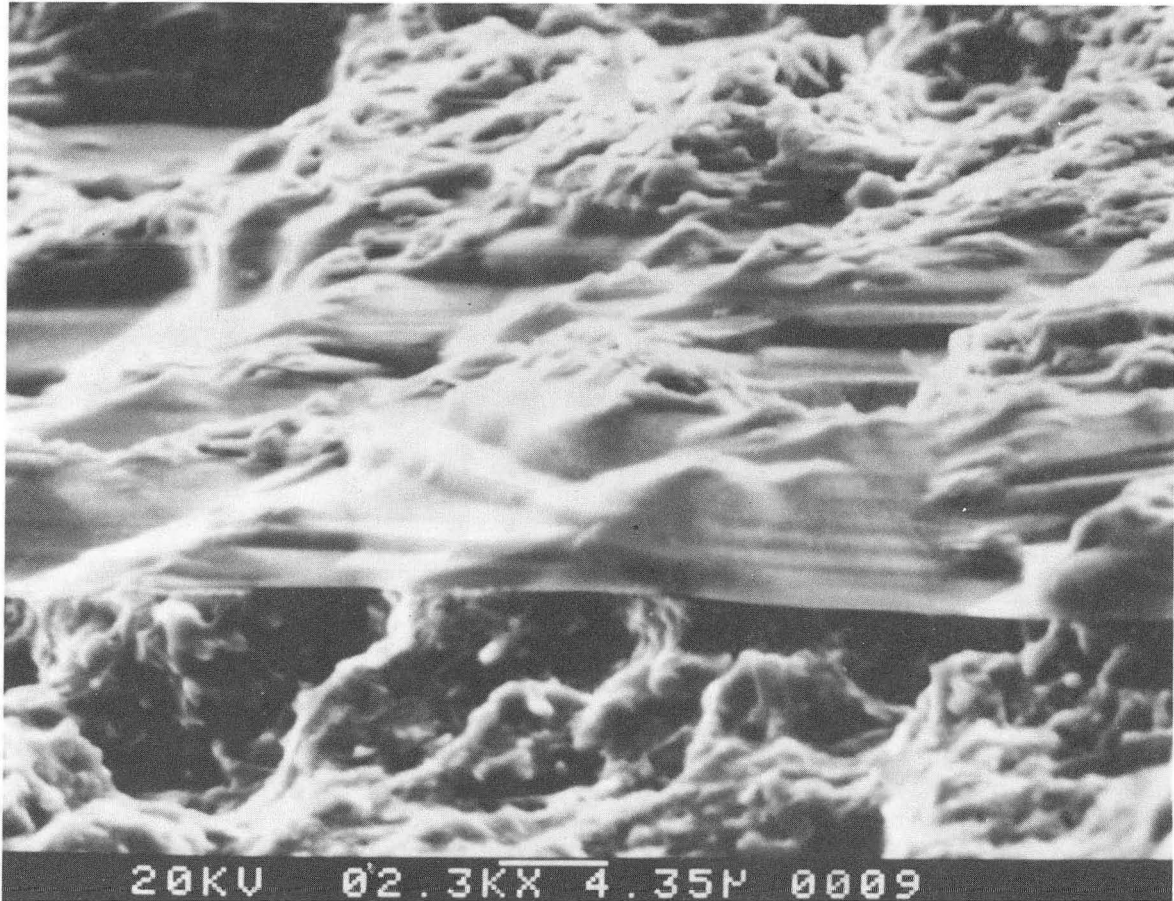
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Figure 1



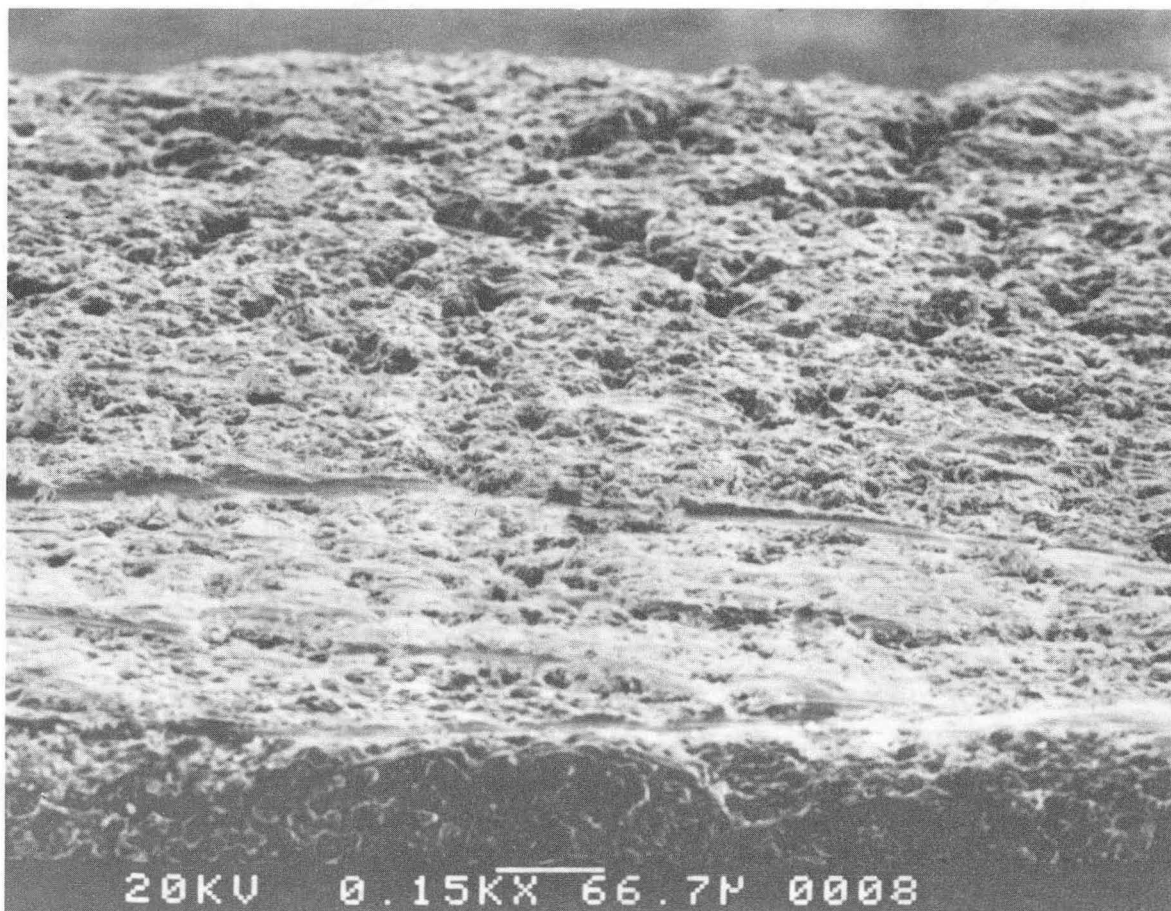
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Figure 2



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Figure 3



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Figure 4

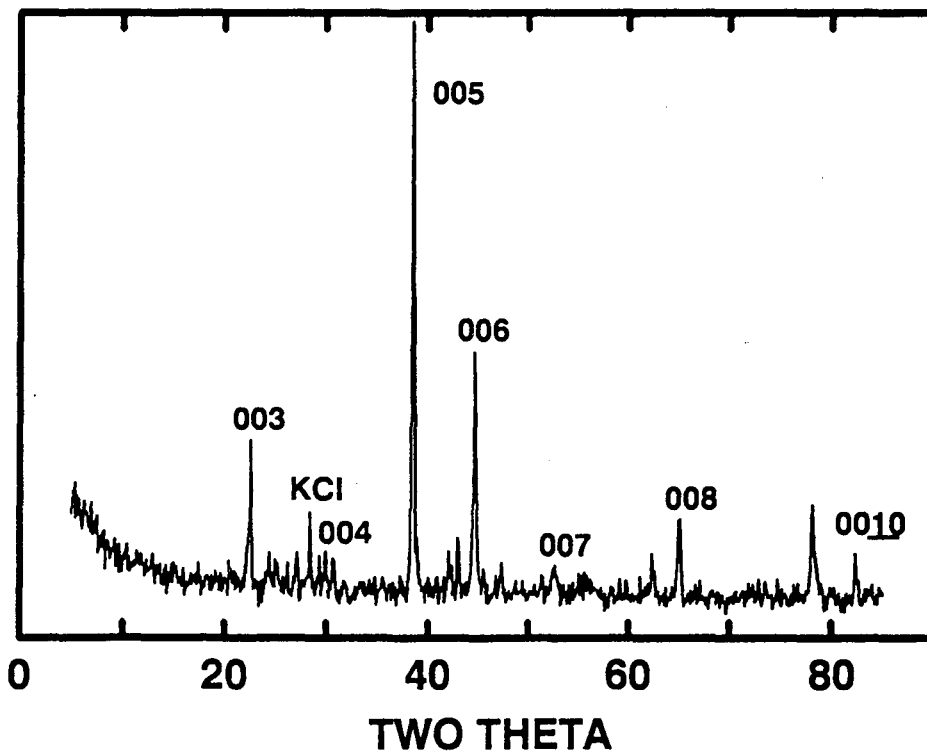


Figure 5

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