LA-UR-97- 3251

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Title:

Computational Modeling of Materials Processing and Processes

Authors:

R. J. Asaro, U. C.-San Diego
S. E. Schoenfeld, U. C.-San Diego
S. Ahzi, U. C.-San Diego
Y. Zhu, MST-STC
J. F. Bingert, MST-6
J. O. Willis, MST-STC
W. R. Blumenthal, MST-5
T. C. Lowe, MST-DO

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Computational Modeling of Materials Processing and Processes

Terry C. Lowe*, Yuntian Zhu, John F. Bingert,
Jeffrey O. Willis, and William R. Blumenthal
Materials Science and Technology Division
Los Alamos National Laboratory

Robert J. Asaro, Scott E. Schoenfeld, and Said Ahzi
Department of Applied Mechanics and Engineering Sciences
University of California, San Diego

Abstract

This is the final report of a three-year, Laboratory Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). Anisotropic mechanical properties of densified BSCCO powders are of paramount importance during thermo-mechanical processing of superconducting tapes and wires. Maximum current transport requires high relative density and a high degree of alignment of the single crystal planes of the BSCCO. Unfortunately this configuration causes high stresses that can lead to cracking, and thus reduce the density, and the conductive properties of the tape. The current work develops a micromechanical material model to help optimize the processing of anisotropic powders such as BSCCO. The model is calibrated and compared to experimental results, and then employed to analyze the effects of initial texture and confinement pressure on the densification and ultimate formability of the powder. Both pressures and shear strains in the core of oxide powder-in-tube (OPIT) processed tapes are calculated by finite-element analysis. The calculated deformations were then applied as boundary conditions to the micromechanical model. Our calculated results were used to interpret a set of prototypical rolling experiments.

Background and Research Objectives

Since the discovery of high temperature superconducting oxides, there have been great interest and research activity in the processing of superconducting films, tapes and wires. The fabrication of tapes or wires can be achieved *via* thermo-mechanical oxide powder-in-tube (OPIT) processing. In this process, the superconducting powder is inserted in a silver tube and the resulting composite is deformed by drawing and rolling. Numerous passes are required before obtaining the final shape of the composite workpiece which is usually subject to iterative thermal and mechanical processing. Oxides of primary

* Principal Investigator, Email: tlowe@lanl.gov

interest to the OPIT process are the bismuth-based oxides, $Bi_2Sr_2Ca_1Cu_2O_x$ and $Bi_2Sr_2Ca_2Cu_3O_x$, which have superconducting transition temperatures of approximately 85K and 110K respectively. This makes them attractive for use at temperatures up to 77K.

The electric properties in these materials are highly anisotropic. They preferentially superconduct (high critical current) on the basal planes of their orthorhombic crystalline structure. This implies that one of the most important conditions for obtaining high critical current in a polycrystal of these oxides is the alignment (texture) of the basal planes with the direction of the current. It was observed that the critical current of tapes decreases drastically with decreasing basal plane alignment in the plane of the tape. Other phenomena that play a role in reducing critical current in the polycrystalline oxide are: the presence of second phase particles, the presence of cracks, and low relative density (i.e. residual porosity). The shape and morphology of the grains as well as the grain boundaries also have an effect on the critical current.

ASARO et al. [1] studied the texture evolution in $Bi_2Sr_2Ca_2Cu_3O_x$ (2223 BSCCO) compacted in a cylindrical die (axisymmetric compression) and in a confined channel die (plane strain compression). For both tests, the experimental results show that basal plane normals (c-axes) rotate to align with the compression direction. For a given amount of reduction, this alignment seems to be more pronounced under axisymmetric compression. Through their results, these authors identified the limited number of shearing systems responsible for inelastic deformation in BSCCO crystals; dislocation controlled slip and sliding following microcracking on certain crystallographic planes. ASARO et al. [1] showed that these mechanisms comprise only three independent sliding systems. To simulate texture evolution in BSCCO, ASARO et al. [1] and later AHZI, ASARO and PARKS [2], proposed viscoplastic models based on crystal plasticity theory. These models neglect the effects of elasticity and porosity, and assume all shearing occurs like crystallographic slip. The predictions of the modified constrained hybrid model of PARKS and AHZI [3] showed good agreement with the experimental observations for texture evolution in 2223 BSCCO (ASARO et al. [1], AHZI et al. [2]). This model has now been extended to include elasticity (SCHOENFELD, AHZI and ASARO [4]). The elastic-viscoplastic model has also been successful in predicting textural evolution; this model, however, is also capable of predicting the complete stress state required in order to accommodate porosity evolution, and eventually the complete stress and strain behavior in 2223 BSCCO.

Although the previous crystallographic slip models were able to capture many of the fundamental mechanical properties of BSCCO aggregates, strain hardening and porosity have yet to be incorporated into the modeling effort. In order to develop a complete

predictive material model for the compaction of the oxide powder, we propose to couple a crystal plasticity model with a porosity model. The crystal plasticity model is based on the elastic-viscoplastic formulation of the modified Taylor model by SCHOENFELD et al. [4]. This model was specifically formulated for crystals which lack five independent slip systems necessary for the accommodation of an arbitrary plastic deformation. In the present work, this elastic-viscoplastic theory is used to model large deformations in polycrystalline 2223 BSCCO. The porosity model that is adopted in the present work is based on the suggestion by FLECK, KUHN and McMEEKING [5] for the use of their quadratic yield function at low relative density, and the use of Gurson's yield function (GURSON, [6]) near full density. At low relative density, FLECK et al. [5] assume the yielding to be controlled by limited contact zone yielding between particles, whereas in the present work, we use their quadratic yield function with the approximation of uniform yielding within each crystal of the powder. In the coupling of these two models, the plastic part of the macroscopic velocity gradient is decomposed into a sum of the deviatoric contribution from the crystals forming the solid (matrix) of the powder, and the volumetric contribution due to the compaction of the voids. In this way, the initial orientation distribution of the crystals can be either random or textured while the evolution of anisotropy associated with large deformations can now be calculated.

To calibrate the proposed model for the compaction of 2223 BSCCO, we conducted a series of experimental tests that consist of channel die compression with a low level of confinement in the channel direction. These tests allowed us to measure the confinement stress, the compressive stress and the relative density versus the compressive and/or channel flow strain. The measured confinement stress serves as an input boundary condition to the model. The hardening parameters are chosen in order to fit predicted stress-strain behavior to that of the experiment. The predictive capabilities of the model are validated by direct comparison to the experimental results. Once calibrated and verified, the model is used to help resolve the macroscopic mechanical behavior of 2223 BSCCO and to understand the complex interaction between bulk anisotropy and density evolution.

In order to illustrate how the controlled processing of such superconducting oxides can be investigated, we have conducted a detailed analysis of the deformation modes that may be observed during the late stages, i.e. tape rolling portion, of the OPIT process. In addition to the rolling of the standard bimaterial workpiece (superconductor core with silver sheath), we have also examined the OPIT workpiece either clad in an additional packing material, pack rolling, or surrounding an additional silver rod, *rod-in-tube* processing. It is the intent here to use a finite-element analysis of the rolling process in order to calculate the

roll gap displacement field. These calculated displacements will then be applied as imposed compression and shearing boundary conditions to an advanced, micromechanically based model of a porous aggregate of low-symmetry BSCCO crystals.

The material model used here will be the porous, elastic-viscoplastic model of SCHOENFELD et al. [7,8]. The model approximates both the lattice-based sliding following microcracking and bulk dislocation motion as crystallographic glide in a polycrystal of low symmetry constituent single crystals. This model has been coupled to a macroscopic flow theory for the compaction of voids within the aggregate, and thus constitutes an ideal model for our purposes. Once the deformations in the center of the core have been calculated from the finite-element model, they will be applied as macroscopic, homogeneous compressive and shear deformations to the porous polycrystal model for BSCCO. It is our goal that such a procedure will allow us to resolve how the rolling process affects the microstructure of the BSCCO core, and how different process parameters may be incorporated so as to mitigate textural locking until the desired thickness reduction and densification are obtained.

The results from these simulations will be used to help interpret experiments that have been designed not around obtaining high critical current density, J_c , but designed specifically to investigate the effects of processing variables on core uniformity. The detailed analysis of the rolling process will provide insight into the core deformation environment, the micromechanical modeling will build the knowledge base on how such process parameters affect the material state within the core, and finally, correlation with these experimental results will offer insight as to how such material states affect the core morphology.

Importance to LANL's Science and Technology Base and National R&D Needs

Materials process modeling has become an integral element of materials design, development, and certification to the extent that it is an enabling technology for many materials applications. At Los Alamos, materials process modeling is an essential element of the Accelerated Strategic Computing Initiative and of other programmatic components of the nuclear weapons program relevant to weapon performance, materials aging, and nuclear weapons materials processing. It is also a prominent part of our industrial, energy research, energy technology, and defense program activities. Recent consensus within the

national materials community is that theoretical and computational modeling developments are changing the very nature of materials science and engineering.

Though this work focusses on the specific application of the materials process model to the processing of BSCCO superconducting powder, the main contribution is the development of the model itself, advancing our ability to analyze processing of anisotropic materials that are not fully dense.

Scientific Approach and Accomplishments

As previously discussed, anisotropy of both mechanical and electrical properties is paramount to the performance of these dense compacts. In an effort to understand how the initial grain alignment will affect mechanical properties in the channel die, we have conducted simulations of the channel die experiment using different pre-textured states to characterize the initial aggregate. As previously discussed, uniaxial compression tends to align the **c**-axis with the compression direction; likewise (according to our simulations), uniaxial tension tends to align the **c**-axis perpendicular to the compression direction. Note that this is not a state that would be simple to realize in a powder compact such as the one used in our experimental studies, but this state may be possible to obtain during the wire drawing portion of an OPIT process. Since the orientation with the compression axes in the compression direction, **CCD**, contains the most grains with their **c**-axes initially aligned with the compression direction, it also produces the highest stresses at the earliest compressive strain. Since the channel die compression will eventually texture all samples in a similar manner, one may wish to begin with an initial texture so that enough strain can be introduced to the sample to provide the desired densification before textural alignment creates high stresses and a profusion of macroscopic cracks. With this in mind, the compression axes in the transverse direction, **CTD**, simulation shows that if the **c**-axes are initially aligned in the transverse direction, they have the largest rotation to follow in order to align themselves with the compression direction. Further, the transverse direction of the channel die sees the smallest amount of deformation, and very large strains will be required to re-orient the **c**-axes to the compression direction. This particular texture may not be relevant for current bulk processing methods, but the compression axes in the rolling direction, **CRD**, simulation also produced a very formable compact. This texture may be possible by high pressure extrusion of an OPIT workpiece prior to rolling.

Analyses of compressive strain verses relative density curves for the above calculations show that the more formable initial textures also produce higher densities for a given amount of compressive strain. Further, since high stresses due to textural alignment have been mitigated by more diffuse textures, these samples will be able to sustain larger compressive strains and become more dense than their more aligned counterparts.

In our experimental study, we compacted the oxide powder in the channel die and used copper plugs to provide powder containment. After approximately 30 to 40% compressive strain, densification in the powder ceased. This could have been due to a lack of confinement stress, or it could have been due to the oxide reaching its maximum strength. In either case, it is obvious that increased confinement stress would cause increased densification in the compact. Such conditions could be achieved by increasing the initial dimension of the copper plugs in the rolling direction.

Our calculations show that although a highly aligned c-axes texture is desirable for large current transport, such grain alignments are not conducive to continued deformation by the same modes that produce them. The calculations also provide strong evidence that the most successful deformation process will be one that accounts for, and may to a certain degree mitigate, the development of a highly aligned texture until the oxide also becomes highly dense. Such processes need to be *designed*; processing should subject the powder to a variety of strain paths in order to keep the oxide intact, and to arrive simultaneously at the desired texture and density.

The purposes for our finite-element simulations have been to investigate the deformation environment during the late stages of processing OPIT wire to tape, to understand the effects of the various deformation paths that are available on the microstructural state of the core, to correlate these material states with experimental core morphologies, and to suggest improvements to the OPIT process based on this new understanding.

We investigated the deformation states and confinement pressures that arise in the oxide core of a rolled composite tape. The mechanics of rolling were discussed with reference to the non-dimensional roll gap parameter, ζ , which controls how the deformation fields scale with continued reduction of the workpiece thickness. For clarity, and maximum effect of any particular deformation field, we restricted our discussion to rolling with a constant ζ . This is very different from rolling by reductions of constant h or Δh , where h is the sample thickness. A workpiece that is processed by fixed ζ will undergo fixed proportions of shear to compressive strain during processing. A workpiece that is reduced by a fixed Δh , or $\Delta h/h$, would by contrast undergo rapidly decreasing amounts of

shear strain relative to compressive strain during subsequent passes. Rolling schedules that continuously impose large shear strains may be difficult or uneconomical to conduct; large shear strains during a single pass however, may not be so difficult to achieve. We have shown that large shear strains imposed on highly aligned cores can offer the means to weaken such textures if the shearing is done in a cyclic, bi-directional manner. The advantage would be that late strain, textural locking could be mitigated until the desired reductions have been achieved.

SCHOENFELD and ASARO [9] discussed the mechanics of rolling in terms of ζ and found that there are actually two distinct types of through-thickness-deformation gradients; large draught rolling ($\zeta \geq 5.0$) and small draught rolling ($\zeta \leq 0.5$). Due to the relatively small thickness of the OPIT processed tapes, it is unlikely that we will encounter the small draught scenario using the standard rolling mills available today. The mechanics of large draught rolling, as they pertain to the rolling of a composite workpiece, are now established. The basic deformation fields that exist will generally provide for proportionally greater shear strain relative to compressive strain in the core when reductions are achieved by relatively small ζ . Further, core pressures will generally be lower when reductions are made under such circumstances. The relative benefits of increased shearing and decreased core pressures remain a topic of debate. Though we have shown that shear strains can be used to control the grain alignment leading to high stresses, depending on the hardening properties of the particular BSCCO powder, the extra deformation induced by such shearing may also drastically increase the resulting compressive stresses. Experimental data concerning such properties are, to date, not definitive.

Figure 1 shows micrographs of longitudinal sections from the four processing conditions for silver-clad tape after rolling to true compressive strains on the order of 250%. A strong inverse correlation between reduction per pass and core dimensional uniformity is observed. The same effect was observed for the lower-strength aluminum-clad tapes, with core instabilities developing at somewhat smaller strains and being more pronounced at final reductions. Although the rolling schedules were not ideal for analysis as previously discussed, it is obvious from our calculations that the tape rolled by smaller reductions is, pass for pass, of a significantly lower ζ . As such, it will generally be subjected to a higher shear/lower pressure environment than the tape rolled by relatively larger reductions. As seen from the micrographs, the tape rolled by small reductions has a more uniform core-sheath interface and is more likely to maintain a smooth interface when subjected to further rolling. The tape rolled by large reductions has a much more pronounced "sausage" shaped core. Geometrical non-uniformities have been linked to

lower current densities (PARRELL et al. [10]) and could be initiated from cracked states. Even though subsequent rolling has healed the cracks, the non-uniformity left at the bimaterial interface could set the stage for non-uniform cores such as the ones in Figure 1. This scenario becomes even more plausible when one considers that such bimaterial tapes are already susceptible to the non-uniformities that arise as a natural consequence of the material interface (STEIF, [11]). With this as a consideration, cracked states far less severe could easily degrade transport performance and provide a catalyst for drastic core non-uniformities.

High current capacities have also been linked to high relative densities. Therefore high triaxial stress states during OPIT processing should be considered desirable. SCHOENFELD et al. [4] modeled the confined channel die compression of the 2223 BSCCO powder and showed that the powder responds to higher pressures by increased densification. Figure 2 presents microhardness data taken at 25 to 50% reduction increments from the BSCCO cores of silver-clad tapes shown in Figure 1c (5%/pass) and Figure 1d (25%/pass). The microhardness measurements shown in Figure 2 suggest that the core resulting from larger reductions per pass is denser than that resulting from smaller reductions per pass. Finally, our finite-element calculations (Figure 3) show that higher pressures are achieved with increasing ζ . This final calculation directly connects the experimental observations with the calculated phenomenology concerning the effects of increased pressure. Unfortunately, this increased pressure is not enough to confine the core uniformly along the material interface.

The effect of relative core thickness of the composite tape was modeled and the results summarized in Figure 4 and Figure 5. Both the shear strains and pressures in the core center decrease as a result of decreasing this ratio. As the thickness ratio decreases, we approach the large-draught, single-component workpiece analysis of SCHOENFELD and ASARO [9]. This analysis shows no significant shear strain in the center of the rolled workpiece. The pressure decreases due to relatively smaller amounts of the higher strength BSCCO material surrounding the core center. As such, lower pressures are to be expected. From a mechanical processing point of view, there is little to be gained from processing low thickness ratios; since maximum current capacity will depend on relatively high amounts of BSCCO relative to Ag, this type of processing should be avoided.

Processing with stronger sheath material has been shown to create higher pressures in the core without a significant difference in the amount of shear that the core sees. The results from our material modeling suggest that this should always be considered an asset. The positive effect of stronger clad material on core uniformity can be observed

experimentally by comparing silver-clad to aluminum-clad tapes. Work-hardened yield strengths for 99.99 silver and 1100 aluminum are approximately 300 Mpa and 180 Mpa, respectively. The stronger silver clad results in more uniform core geometries at large reductions. In fact, the small relative core, aluminum-clad tape is nearly discontinuous. The improved uniformity exhibited by the annealed tape suggests the work-hardening rate of the clad material may also play a significant role in delaying core mechanical instabilities. These observations are consistent with the analysis of STEIF [11], which suggests that both higher yield strength and higher work hardening rates should serve to postpone the onset of non-uniformities even in the face of drastic increases in the core strength and work hardening rate (textural locking). In this case it is not the weakening of texture due to shearing that helps preserve the uniformity, but the increased mechanical performance of the clad material.

As to the effects of the shear strain, we have very mixed results. Our initial analysis modeled an aggregate without strain hardening; i.e., we neglected microstructural effects due to increased resistance to slip through increased dislocation densities or increased resistance due to increasing the frictional area for microcrack movement. All hardening effects were due purely to the reorientation of anisotropic grains with increasing deformation. As compressive strains are introduced to the polycrystal, the plastically inextensible c-axis direction of the BSCCO single crystal aligns with the compression direction. The result is a drastic increase in compressive stresses that will rapidly lead to the formation of macroscopic cracking modes. Our analysis of the rolling process was conducted with the specific aim of finding deformation paths that would delay this inevitable (and highly desirable from the point of view of obtaining high J_c) alignment until maximum densification could be achieved. When such lattice based hardening mechanisms are neglected our modeling shows that the introduction of cyclic shearing (resulting from bi-directional rolling) will increase the amount of compressive strain that we can apply to the oxide.

In order to calibrate our material model to the experimental data of SCHOENFELD et al [7,8], Taylor rule hardening was introduced to the model. Possible microstructural mechanisms for this type of hardening were discussed, but no experimental justification for such mechanisms was ever found. As such, the simplest strain hardening assumptions were made; we assumed that each slip system began with an equal resistance to slip and evolved equally with the accumulated sum of slips on all the systems. This is an overly simplistic assumption when one considers the mechanisms responsible for slip in BSCCO single crystals. When compressive and shear strains were applied simultaneously to the

model, any reductions in the compressive stress states or locking strains that we saw previously were rapidly overcome by increased macroscopic hardening. The macroscopic hardening is now directly proportional to the accumulation of slip in the single crystals. Thus any added shear on the aggregate will be immediately reflected by increased flow stress in the aggregate. The current rolling experiments suggest that there is a benefit to increased shear strain in the core; more experimentation is required to resolve the micromechanisms involved.

Our final set of rolling calculations suggests that both the shear strains and the pressure that is seen by the core can be increased either by pack rolling, or rod-in-tube processing; therefore both processes should be considered. Shear strains in both cases can be controlled with different materials used as the packing or core respectively. Pack rolling has the additional convenience that it can be employed intermittently during the rolling schedule. Pack rolling with strong (relative to the BSCCO core), thick (relative to tape dimensions) packing can be used to provide single passes that induce high shear strains in the core. Any shearing done to the core should also be reversed by a subsequent pass. Rod-in-tube rolling may be easier and more economical from a processing point of view; there will be less waste material and the process itself may be simpler to implement than pack rolling, but the rod-in-tube process has the additional difficulty of establishing a suitable core material. The material must be relatively strong (to provide maximum shear and pressure) but must also be chemically compatible with the entire process. Further, the rod-in-tube process will produce relatively small core to tape thickness ratios and thus reduced current carrying capacities.

In conclusion, we find that core non-uniformities are a natural outcome of deforming the composite system (STEIF, [11]); and that these instabilities are a combination of all the process parameters and material strength and work hardening properties in both the cladding and the core. The successful tape rolling operation must consider all these factors and interactions in order to produce long lengths of highly dense, highly uniform, superconducting tapes.

Publications

1. SCHOENFELD, S. E., AHZI, S., ASARO, R. J, and BLUMENTHAL, W. R., "The Bulk Processing of 2223 BSCCO Powders: I. Densification and Mechanical Response," *Philosophical Magazine A*, **A73**, pp. 1565-1590, (1996).
2. SCHOENFELD, S. E., ASARO, R. J, BINGERT, J. F., and WILLIS, J. O., "The Bulk Processing of 2223 BSCCO Powders: II. Tape Rolling," *Philosophical Magazine A*, **A73**, pp. 1591-1620, (1996).

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Figure 1. Light micrographs of longitudinal sections from silver-clad tapes showing inverse relationship between reduction per pass and core uniformity for both thin and thick cores: a) 5%/pass, relative core thickness (rct) = 0.2; b) 25%/pass, rct = 0.2; c) 5%/pass, rct = 0.6; and d) 25%/pass, rct = 0.6.

Figure 2. Diamond pyramid microhardness of BSCCO core as a function of rolling strain for silver-clad tapes from Figure 1c (5%/pass) and 1d (25%/pass).

Figure 3. Pressure profiles along the length of the oxide core for the two phase composite with a BSCCO to silver ratio of 0.60. The workpiece is rolled through three different rolling geometries ($\zeta \approx 5.04, 7.13$ and 11.3). The pressures are normalized by the flow stress in BSCCO at the final compressive strains, while distances along the length of the oxide core are normalized by the contact length for each geometry.

Figure 4. Shear strain profiles along the length of the oxide core for the two phase composite with a BSCCO to silver ratio of 0.20. The workpiece is rolled through three different rolling geometries ($\zeta \approx 5.04, 7.13$ and 11.3). The shear strains are normalized by the final compressive strains, while distances along the length of the oxide core are normalized by the contact length for each geometry.

Figure 5. Pressure profiles along the length of the oxide core for the two phase composite with a BSCCO to silver ratio of 0.20. The workpiece is rolled through three different rolling geometries ($\zeta \approx 5.04, 7.13$ and 11.3). The pressures are normalized by the flow stress in BSCCO at the final compressive strains, while distances along the length of the oxide core are normalized by the contact length for each geometry.