Project Title:	Nucleation and Growth of YBCO by the BaF_2 Process and AC Losses of YBCO Films
Organization(s):	**Brookhaven National Laboratory**
Presenters:	M. Suenaga and D.O. Welch
FY 2004 Funding:	$500 K

Project Purpose and FY 2004 Objectives: The purpose of this project is 1) to develop synthesis methods/protocols, which are suitable for the fabrication of $YBa_2Cu_3O_x$, YBCO, coated conductors, through the development of an understanding of the mechanisms, kinetics, and thermodynamics of a- and c-axis nucleation in the BaF_2 process for YBCO thick films. 2) Another purpose of this project is to perform small-scale ac loss measurements which assist in the development of understanding of the currently known or expected ac losses in electric utility devices using HTS conductors. Our objectives for FY 2004 were to continue the study of the nucleation mechanisms in thick YBCO films by investigating the effects of variations in the growth conditions on the c-axis nuclei densities in the films. We also planned to emphasize our nucleation study on the films on buffered metallic substrates. These substrates were to be provided by American Superconductor Corp., AMSC. Also, we began a component of this program devoted to the theory and modeling of the thermodynamics and kinetics of nucleation and growth in the BaF_2 process, focusing especially on factors which control a- and c-axis nucleation.

FY 2004 Performance and FY 2005 Plans: YBCO thick film processing: a) We have developed a new and very simple optical microscopy technique to measure directly areal densities of YBCO nuclei in fluorinated precursor films on a substrate. We used this technique to study the relationship between the nuclei densities and critical current densities of YBCO films, primarily on buffered metallic substrates from AMSC. A simple thermodynamic model for the nucleation was also developed to describe some of the experimental observations. b) Theory and modeling efforts focused on the use of semiempirical atomistic and statistical thermodynamic methods to estimate thermophysical properties pertinent to the epitaxial nucleation of YBCO in the BaF_2 process. Also, in collaboration with W. Wong-Ng (NIST) and H. Su (Caltech), the systematic variation of the temperature of the orthorhombic-to-tetragonal transition in the family of RE-123 compounds was investigated. c) A collaboration was initiated with M. Cima (MIT) to investigate the roles which fluorine plays in the nucleation and growth of YBCO in the BaF_2 process. d) A simple method for the measurements of ac losses of YBCO in perpendicular magnetic fields was developed with J. R. Clem (Ames Laboratory). This technique was employed to measure the losses of YBCO films on buffered metallic substrates from AMSC and Superpower/IGC.

In FY2005: 1) The growth of thick (> 2 μm) YBCO films on buffered metallic substrates will be continued and will emphasize the control of the growth conditions to achieve the consistency of the properties of the films. In this effort, our new technique to determine the YBCO nuclei densities and our theoretical understanding of the factors controlling c- and a-axis nucleation will play important roles for the development of the clear relationship between the processing conditions and the properties of YBCO thick films. 2) Theory and modeling will be focused on delineating processing conditions which result in appreciable a- and randomly oriented axis nucleation. 3) ac losses of YBCO films on metallic substrates will be continued emphasizing the effects of stacked films and non-uniform macroscopic flux penetration on the losses. 4)The collaboration will also be continued with M. Cima at MIT on the modeling of nucleation and growth kinetics and nanoscale structure evolution in the case of MOD-derived precursors for comparison with the behavior for the precursors by an e-beam deposition method used at BNL.

FY 2004 Results: Key results from the FY 2004 program are summarized below.

1) YBCO nucleation and growth

a) YBCO nuclei densities and growth conditions

We developed an experimental method for direct measurements of areal densities of YBCO nuclei in fluorinated precursor films which are on CeO_2 buffered metallic and single-crystalline

$SrTiO_3$ substrates. The aerial density of YBCO nuclei was measured by polarized light microscopy after the nuclei have grown to the surface of the film. The density was found to depend strongly, not only on processing conditions, but also on the type of the substrate. We have also established the correlation between the areal density of the nuclei and the nucleation of the randomly and/or a-axis oriented YBCO grains in the films. A model, which is based on classical nucleation theory, correctly predicts some aspects of the functional dependence of the nuclei density with processing parameters. It is shown that the c-axis-oriented nucleation dominates if the average distance between the nuclei is a few times or greater than the film thickness of the precursor. The formation of randomly oriented grains is interpreted to be a result of collective interactions among the c-axis-oriented nuclei which are initially formed. By controlling the nuclei densities, J_c (self fields) of ~/ > 1 MA/cm^2 was achieved for 2 μm thick YBCO films on buffered metallic substrates from AMSC.

b) Modeling of Thermodynamics, Kinetics, and Mechanics

A combination of atomistic modeling and the use of scaling-relations based on ionic-crystal theory was used to estimate the lattice energy, surface energy, and elastic moduli of the $(Y,Ba)(O,F)_2$ precursor in the BaF_2 process and to estimate the interfacial energies between the precursor, YBCO, and a CeO_2 substrate. These results were used to estimate the dependence on the degree of oxygenation of the nucleation barriers for c-axis and a-axis nucleation. As part of an exploration of the comparative processing behavior of YBCO and family of REBCOs, the systematic variation of the thermodynamics of oxygen content and order was studied by atomistic simulation and the results compared with experimental data obtained by W. Wong-Ng and L. Cook (NIST) for the temperature of the orthorhombic-tetragonal oxygen ordering transition, thus yielding an understanding of the systematics of oxygenation in these superconductors.

2) AC losses

An ac loss measurement method, which used a planar pick-up coil, was developed using a YBCO film from S. Foltyn/LANL. This method does not require calibration for the losses and it was shown that the correct losses were measured as long as the diameter of the coil was greater than ~3 times of a specimen diameter. Also a specimen can be as much as 1 mm above the coil for correct loss measurements. The latter allows the use of this technique to study the effect of stacking the films and this is important for applications of YBCO coated tapes in electric devices.

Research Integration: Our studies in FY04 were performed as informal collaborations with the staffs at LANL, MIT, NIST, AMSC, and Superpower. We would like to point out that the results of the work in this program can take some "incubation period" before their value becomes apparent to industry. For example, the concept of subatmospheric pressure processing in the BaF_2 process, which we introduced a few years ago, is now an indispensable modification in the fabrication of YBCO coated conductors by this process.