

## Macroscopic and Microscopic Modeling of the Growth of YBaCuO Bulk Material

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**Abstract**—Melt processing of YBaCuO is widely accepted to yield the best superconducting properties in bulk specimens of this material. Modeling of melt processing is a promising tool to optimize the production of bulk YBaCuO material.

In a macroscopic simulation, temperature fields in YBaCuO specimen of arbitrary geometry in e. g. Bridgman or VGF furnaces are calculated during solidification. This allows the visualization of the shape of the solidification isotherm and its dependence on cooling rate and the geometries of furnaces and samples.

Microscopic modeling of microstructural evolution during YBaCuO growth is of great interest to deepen the understanding of growth kinetics. For this purpose, YBaCuO growth can be simulated numerically using the multiphase field model.

### I. INTRODUCTION

Melt processing of small samples of YBaCuO material in principle is accepted to yield superconducting properties exceeding the requirements for several technical applications such as magnetic bearings and fault current limiters. However, industrial applications of this high  $T_c$  superconducting material require improved manufacturing processes. Scaling up the sample size of single domain bulk material is as necessary as a reduction of the processing time. Macroscopic and microscopic simulations yield a deeper insight in the melt texturing process and help to overcome problems related with it. In this paper results of macroscopic and microscopic simulations are presented.

Macroscopic thermal field calculations allow the prediction of the temperature distribution even in large specimen of complex geometry and accordingly enable process optimization. Thermal field calculations are carried out using the finite element code CASTS (=Computer Aided Solidification Technologies) [1].

The prediction of the microstructure resulting from a specific process also is most important with respect to the control of good superconductive resp. magnetic properties of the resulting specimen. Simulations of the grain growth and the resulting microstructures are performed with the multiphase field method [2].

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### II. MACROSCOPIC SIMULATIONS OF MELT GROWTH OF YBaCuO

In process optimization of the melt growth process, both minimization of the processing time as well as an improved texture of the bulk material is sought. This demands an optimal choice for the temperature program of the heaters of the furnace being used for the melt growth process. For this purpose information about the process is required which can hardly be measured experimentally, e. g. the point of time and the spot where the isotherm of solidification enters the YBaCuO specimen. The quality of the melt growth process also depends on the curvature of this isotherm. Therefore, the numerical simulation of the temperature field during the process is a valuable tool to recover this data. The software CASTS [1] is designed to perform such calculations. Besides solving the heat conduction equation this software also takes into account the transport of energy from the heaters to the specimen (and vice versa) via infrared radiation as well as the release of latent heat.

With this kind of process simulation the time required for the melt growth process can be reduced significantly. This can be done by choosing an appropriate temperature program for the heaters of the furnace ensuring that the isotherm of solidification enters the specimen exactly after a specified time. Furthermore, the cooling rate can be increased after the retirement of the solidification front from the specimen, Fig. 1. Also the distortion of the temperature field in the YBaCuO specimen by the holding devices becomes visible.

The temperature fields in a Bridgman furnace are visualized in Fig. 2. The temperatures of the heating zone and the water-cooled cooling zone were assumed to be 1150° C and 20° C respectively; the height of the baffle (i. e. the thermal insulation zone) was 3.4 cm. The temperature field was calculated for cylindrical YBaCuO specimen with two different diameters (10 mm and 32 mm). The withdrawal velocity was set to 1 mm/h. As shown in Fig. 2, the deviations of the isotherm of solidification from a planar surface are negligible. The temperature gradient parallel to the furnace axis at the isotherm of solidification is 80 K/cm. The magnitude of this gradient can be increased by decreasing the height of the baffle.

Also the visualization of the shape of the isotherm of solidification is helpful to improve the melt growth process. As an example the isotherms were calculated in cylindri-

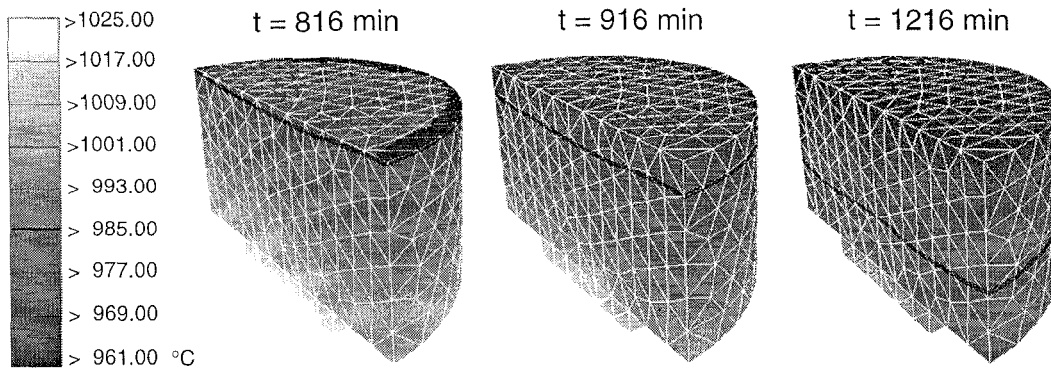


Fig. 1: Simulations of the temperature field in a conventional furnace with vertical temperature gradient. Sections through the specimen (diameter 30 mm, height 15 mm) are shown. The cooling rate is  $\dot{T} = -2$  K/h. The isotherm of solidification is marked with black color. Experimental set-up is from IPHT Jena, Germany.

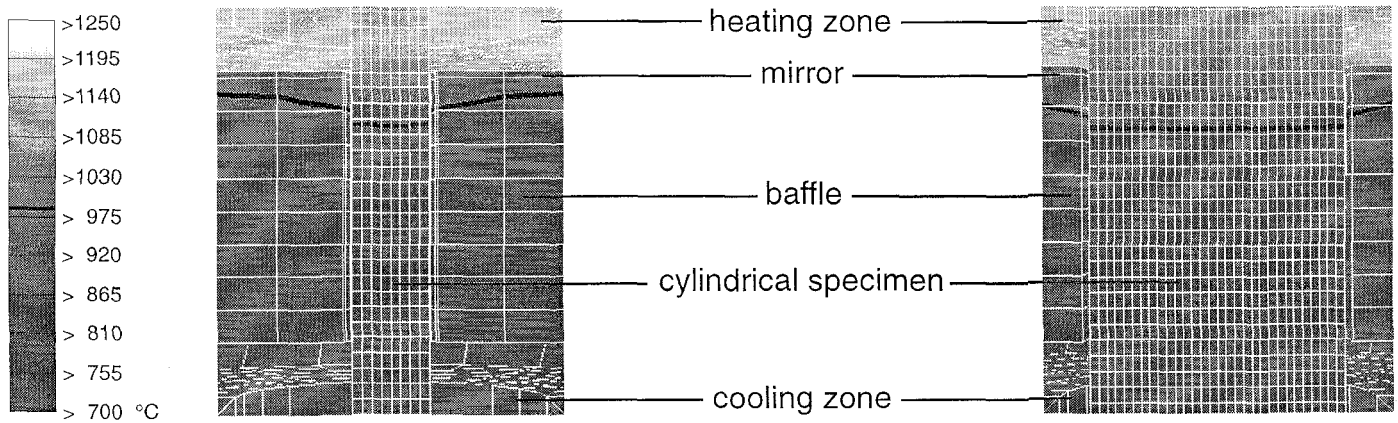


Fig. 2: Simulations of the temperature field in a Bridgman furnace. Sections through the furnace are shown. The specimens have a diameter of 10 mm (left diagram) and 32 mm (right diagram). Withdrawal velocity is 1 mm/h. A mirror surface attached to the baffle ensures that the baffle does not become too hot. The black drawn isotherm of solidification is planar.

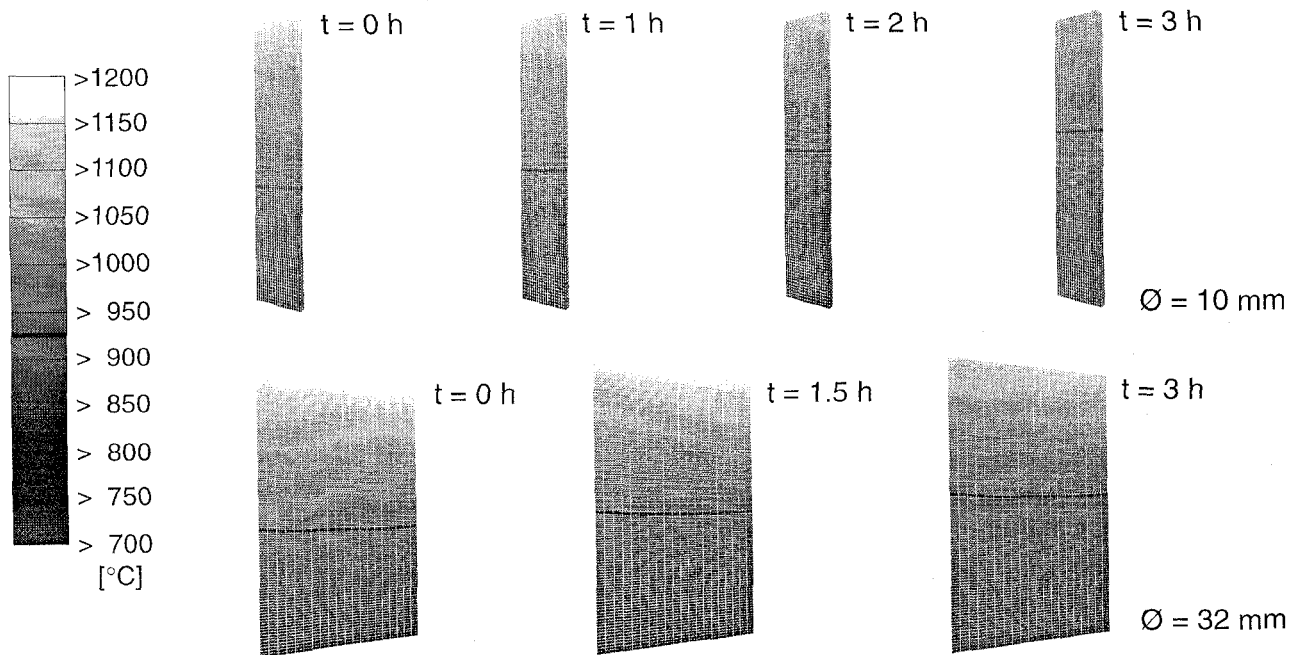


Fig. 3: Simulations of the temperature field in a VGF furnace. Sections through the specimen with height 5.4 cm and diameter 10 mm (above diagrams) and 32 mm (below diagrams) are shown. Cooling rate is  $\dot{T} = -10$  K/h. The isotherms of solidification are marked with black color. The curvature of the isotherms increases slightly with the diameter of the specimen.

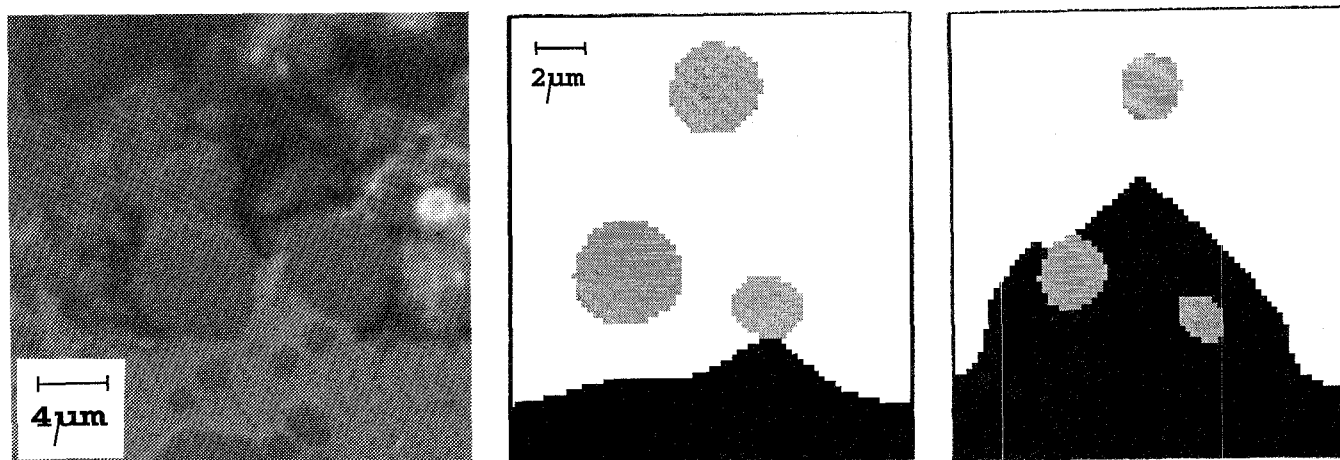


Fig. 4: Left side: Micrograph of a quenched YBaCuO specimen. Dark (medium, light) grey corresponds to the liquid (211, 123) phases respectively. Engulfed 211 particles are clearly seen. Also bridge formation is observed. Middle and right side: Microscopic simulation also reveals engulfment and bridge formation. Black (grey, white) color indicates the 123 matrix (211 particles, liquid), respectively.

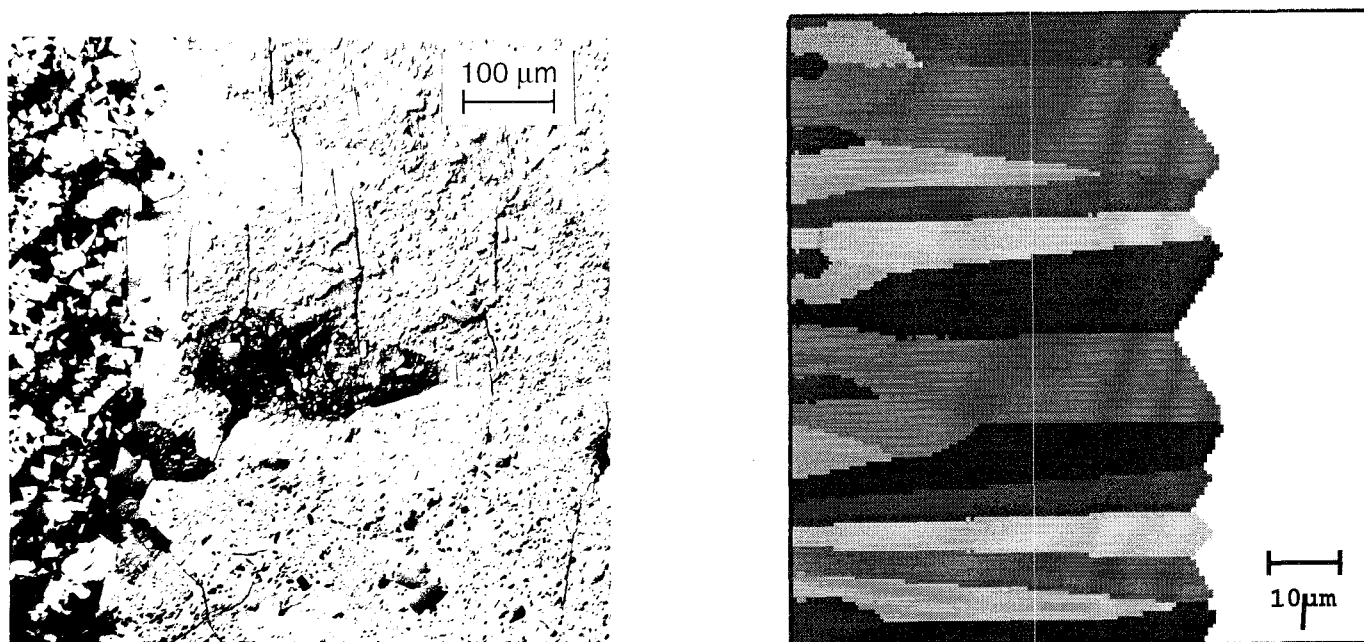


Fig. 5: Left side: Experimental observation of grain selection 123 grains grow from the  $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$  powder on the left side. A short distance ( $75 \mu\text{m}$ ) is sufficient for grain coarsening. Closer examinations reveal that the selected grains have c-axis orientation. Right side: The effect of grain selection was also studied by numerical simulations using the phase field method. Different 123 grains are visualized by different grey levels.

cal specimen of different radii in a Vertical Gradient Freeze (VGF) furnace with 8 ring-shaped heaters with a diameter of 8.9 cm [3], Fig. 3. In this example the cooling rate was  $\dot{T} = -10 \text{ K/h}$ . In the specimen with 10 mm diameter the curvature of the isotherm is almost planar. A small curvature of the isotherm of solidification is observed in the specimen with a diameter of 32 mm. For values of the cooling rate below  $-10 \text{ K/h}$  the curvature of the isotherms does not depend on the cooling rate. The temperature gradient parallel to the furnace axis is  $40 \text{ K/cm}$  ( $45 \text{ K/cm}$ ) in the specimen with diameter 10 mm (32 mm). In further investigations, the temperature program of the heaters in the

VGF furnace could be optimized to minimize the curvature of the isotherms.

### III. MICROSCOPIC SIMULATIONS OF YBaCuO GRAIN GROWTH

Modeling of microstructure formation is a relative new topic in materials' sciences. Three different approaches can be distinguished: Monte-Carlo-simulations [4], cellular automata models [5] and the phase field method [6], [7]. Recently, the phase field concept has been extended to multiphase systems [2]. This model allows simulations of the

peritectic solidification process of YBaCuO from the melt.

In the multi phase field concept each phase field variable represents either the liquid or a single grain of the phases  $Y_1Ba_2Cu_3O_{7-x}$  or  $Y_2Ba_1Cu_1O_{5-x}$  respectively. Their evolution in time is described by a set of nonlinear parabolic differential equations being coupled with a diffusion equation determining the local concentration of Yttrium at any position and time. These equations are discretized by a finite difference technique using explicit algorithms on a workstation.

The first example deals with bridge formation and engulfment of  $Y_2Ba_1Cu_1O_{5-x}$  particles into the solidification front of the  $Y_1Ba_2Cu_3O_{7-x}$  phase [8]. Below the peritectic temperature, 211 particles are dissolving in the presence of growing 123 grains. As a result, a gradient of the Y-concentration occurs and growth of 123 grains towards the dissolving 211 grains is favoured (bridge formation). If the 211 particles are too big there is not enough time to dissolve before the arrival of the 123 solidification front. As a result, they are trapped in the 123 matrix and may serve as pinning centers (engulfment). These processes are experimentally observed and are also amenable to numerical simulations, Fig. 4.

Another example is a recently proposed geometrically organized grain growth process [9], [10]. This process is supposed to have the potential for an economic isothermal production of e. g. superconducting tapes. Essentially for this process is grain selection of 123 grains caused by anisotropic growth conditions. Of particular interest is the grain selection distance required for the well oriented grains to overgrow the misaligned ones. The phase field method is used to study this process, Fig. 5. For example, the effect of variations of the diameter of the heterogeneous seeds and their mean distance can be investigated.

#### IV. CONCLUSIONS AND OUTLOOK

Modeling of YBaCuO melt processing is a valuable tool to optimize the production of bulk YBaCuO material. Macroscopic simulations of the thermal fields in the specimen during the melt texturing growth process provide information about position, curvature and velocity of the isotherms of solidification. This allows for a substantial shortening of the processing time as well as a minimization of the curvature of the isotherms during solidification of large samples. A quasi isothermal seeded melt growth process with a cold finger at the site of the seed is most promising for an economic large scale production of YBaCuO bulk material. In contrast to the directional solidification processes discussed above, the temperature gradients occurring in these processes are very small. Thus, the temperature field in the specimen is dominated by the release of latent heat. Simulation of the temperature fields makes an important contribution to exploit the potential of process optimization.

Microscopic simulations assist to deepen the understanding of the evolution of the microstructure in YBaCuO bulk

material having a major influence especially on the critical current density. Since standard procedures for the solution of the phase field need extensive computer power, a much more efficient algorithm was developed to decrease storage requirements and CPU time. This will enable three dimensional simulations soon. Future work will focus on quantitative simulations using thermodynamical data sets of the various phases in the YBaCuO system in order to investigate other effects controlling the growth like e. g. oxygen pressure.

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