New Gas Flow System for Long-Tape Reaction Tube of TFA-MOD Process

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Abstract. A multi-turning Reel-to-Reel tape conveyance system of the long YBCO coated conductors using TFA-MOD process is desired to be high production rate without large degradation between tape lines. The numerical simulation, which calculated the YBCO growth kinetics, gas element diffusion and gas flow, shown a possibility that the vertical gas flow onto the tape surface realized high production rate of YBCO conductor even on the wider tape. This possibility was also proved by experimental measurements.

1. Introduction

The MOD process using precursor solution containing metal trifluoroacetates (TFA) is a low cost process for YBa2Cu3O7-x (YBCO) coated conductors because of its non-vacuum process. The epitaxial growth of YBCO crystals are accompanied by both consuming H2O gas and releasing HF gas at the crystal growth interface. Thus, it is required for manufacturing long YBCO coated conductors at a high production to design an appropriate gas flow system for the efficient above mentioned conversion kinetics. A numerical method for simulating the YBCO growth, which contains the multi-dimensional gas flow effect, has been proposed[1,2]. This numerical method is based on the YBCO reaction model as,

\[ R = k_+ \cdot X_{H_2O}^p - k_- \cdot X_{HF}^p = k_+ \left( X_{H_2O}^p - K^{-1} \cdot X_{HF}^p \right), \]

where R is the YBCO growth rate, \( k_+ \) and \( k_- \) are rate constants of the reaction and the reverse one, and \( X_{H_2O}^p \) and \( X_{HF}^p \) are molar fractions of H2O and HF at the interface in the precursor, respectively[3]. A one-dimensional growth model of the YBCO was proposed in consideration of the one-dimensional growth kinetics at the interface between the precursor and the YBCO crystalline layers together with the conservation of the gas components, H2O and HF, in the precursor layer[3]. This growth model was treated as a boundary condition for the convective multi-component diffusion equations in the gas region. Subsequently, the convective multi-component diffusion equations and Navier-Stokes equation in the gas region were solved in multi-dimensional manner by the finite difference method. It was found that this numerical model calculation could make a good estimation for the growth rate distribution in the tape and the molar fractions of the components in the gas region. Furthermore, it was confirmed that the supplied water vapor molar fraction dependence, the positional dependence
and the inlet gas velocity dependence of the calculated YBCO growth rate were in good agreement with the experimental results[1]. And, it was revealed that this numerical method was an effective tool to design a suitable configuration of the gas flow in the reaction tube for uniform reaction as well as realizing higher production rates[2].

It was confirmed by the experiments that the conventional gas flow mode, in which the gas flew in parallel with the tape surface, the Jc value monotonously decreased toward the leeward direction, as shown in figure 1[4]. In this mode, the YBCO growth rate also resulted in inclined growth rate distribution in figure 2[2]. Thus, it is considered that the evident degradation of the Jc value is related to the large difference of the growth rate in a wider tape. On the other hand, a multi-turning Reel-to-Reel tape conveyance system for a high production rate of the long YBCO coated conductors is desired keeping a small reaction tube size. In this system, however, the YBCO growth reaction will act on a wide surface, of which the width is the total of multi-put tapes on the holder in the reaction tube. Assuming three 1-cm width tapes placed on the holder in the reaction tube of the conventional parallel gas flow mode, the difference of the growth rate values between the upwind tape and the leeward tape is more than three times, as seen in figure 2. This difference may result in decreasing the total mass of YBCO layer. Thus, the conventional parallel gas flow mode is not considered to be suitable for the multi-turning Reel-to-Reel tape conveyance system. By the numerical simulations, however, it is shown a possibility that the vertical gas flow onto the tape surface realized highly uniformed YBCO growth rate distribution on the wider reaction area of three parallel 1-cm wide tapes. The reliability of this vertical gas flow mode will be proved by comparing with the growth rate distributions of the experimental measurement and numerical simulation.

2. Vertically Supplied Gas Flow System

The schematic illustration of the cross-sectional reaction tube for the multi-turning Reel-to-Reel tape is shown in figure 3. The longitudinally uniform temperature length, which is a reacting length, in the tube is designed as 30cm. Three 1-cm wide tapes are placed on the holder. The three nozzle lines of the supplied gas pipe are vertically faced the three tapes, respectively. Two nozzle lines of the discharging gas pipe are placed in the both sides of the tape holder as the same height of the holder surface. Thus, the supplied gas, which is Ar gas containing H₂O vapor, flows perpendicularly to the tape surface and the reacted gas, which contains HF gas, is discharged from the both side of the holder.

3. Experimental Conditions

The tube was heated up at the constant rate of 2°C/min and held at 778°C for the formation of the YBCO phase. The water vapor partial pressure in the supplied gas was 13.6%. The gas flow rate was...
7.5 liter/min. Three 15 cm long and 1 cm wide tapes, of which the final YBCO thickness was 0.67 μm, were placed on the holder in parallel. The YBCO growth rate values in the center of the three tapes were measured by using the electrical resistance value which changes as the YBCO layer grows epitaxially changing during YBCO growth period. Details were reported by R. Teranishi et al. [5].

Figure 3. Schematic illustration of the cross-section of the reaction tube for the vertical supplied gas flow mode.

4. Numerical Simulation
Two-dimensional calculation was performed in the above mentioned conditions for the configuration in figure 3. The kinetic rate constant of YBCO growth $K$ in equation (1), which is usually selected as $10^{-8}$ order value for the TFA-MOD process, was used as $0.7 \times 10^{-8}$ in all present two dimensional calculations, which was a reasonable value in this process[3]. The calculated steady state distribution of the HF molar fractions around the tapes and nozzles are shown in figure 4, in which the symmetrical half region is drawn from the vertical center-line. It is schematically confirmed that the boundary thickness on the tapes seems to be almost constant. This numerical result gives a prospect that there are no large difference of the growth rates between the three tapes, since the growth rate value is influenced mainly by the HF concentration boundary layer thickness[3].

5. YBCO Growth Rate
The numerically obtained growth rate value distributions are shown in figure 5 with the experimental results. These are good agreement with the measured growth rate values. Comparing with figure 2, the total YBCO layer productions seems to be similar. In figure 5, however, there is no evident degradation of the growth rate as figure 2 along the width direction. When we consider a wider reaction width, the higher production rate with high efficiency could be expected by the vertical gas flow system. Consequently, it is proved for the vertical gas flow mode to realize high production of the YBCO layer for the wider reaction tape.

6. Conclusion
It is confirmed that the vertical gas flow mode, which is applicable to a wider tape to increase the production rate of the long YBCO coated conductors with keeping the reaction tube small, is suitable for the multi-turning Reel-to-Reel tape conveyance system by numerical and experimental studies.
Figure 4. Steady state distribution of HF molar fraction.

Figure 5. YBCO growth rate distributions in the tapes.

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