Use of X-Ray Imaging for Evaluation of Superconducting
Ceramics Fabricated by PIT Method

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Abstract

High-spatial-resolution microfocus-based X-ray imaging technology, both real-time and film, has been used to study the thickness variation that occurs in the oxide core of silver-sheathed BSCCO ribbons produced by the powder-in-tube method. This thickness variation (also called "sausaging") occurs during incremental rolling and degrades the critical current densities of the ribbons. Using X-ray image data we determined the onset and severity of sausaging. Onset occurred at a ribbon thickness of 203 μm , and the severity increased with subsequent rolling reductions. Scanning electron microscopy was used to confirm the X-ray image data. X-ray imaging provides several advantages over more traditional methods for characterizing the superconductors such as optical or scanning electron microscopy. The X-ray imaging provides nondestructive information about the entire width of the ribbon, with far greater speed, lower cost, and more flexibility than the traditional techniques.

Introduction

Many applications of superconductors to industrial uses will require that long ribbons be produced [1]. The powder-in-tube (PIT) method can produce long lengths of silver sheathed BSCCO ribbons. Ribbons made by this process have yielded critical current densities (J_c) values as high as 53700 A cm^{-2} in zero magnetic field [2]. The J_c of these ribbons increases as ribbon thickness decreases [3,4]. One difficulty in making thin, silver clad ribbons is that as the ribbon's thickness is reduced, the internal oxide core develops thickness variations in the longitudinal direction; this effect, called work instability or "sausaging", degrades the J_c value. The highest J_c values come from uniaxially pressed ribbons. While the deformation process is more uniform during pressing, rolling is necessary when processing long lengths. Although different J_c values between pressed and rolled ribbons can be correlated with degree of preferred texturing and densification, the main reason for this difference is attributed to the sausaging caused by rolling [5]. Because of the increase in J_c with decreasing ribbon thickness and the need to process long lengths by sequential rolling reductions, understanding the sausaging of these ribbons is essential in making long ribbons with high J_c values.

In the past, typical methods of examining a ribbon for sausaging have been optical or scanning electron microscopy. These methods are time-consuming and destructive requiring the samples to be mounted and polished. In addition, only a small part of the sample can be examined. High-spatial-resolution X-ray imaging overcomes these problems and allows full inspection of a sample. It also has the potential to monitor changes that occur during the rolling process, allowing the examination of any section of the specimen throughout the process without cutting the tape. This offers a valuable advantage for monitoring production-scale lengths.

Sample Description

The PIT process was implemented to produce silver-clad ribbons of BSCCO superconductor. The process involved the manual packing (to $\approx 30\%$ of theoretical density) of a closed-end silver tube with powders (particle size $\approx 5\text{ }\mu\text{m}$) containing a mixture of phases to give a nominal composition of $\text{Bi}_{1.8}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$. The packed tubes are 15 cm long and have outer and inner diameters of 6.35 and 4.35 mm, respectively. Typically the packed tubes are vacuum annealed in an atmosphere of flowing O_2 (3 mm Hg pressure) at 650°C for 6 hours to remove moisture and other gases that may evolve during the heat treatments. Each powder-filled tube is then sealed and swaged down to a diameter of 4.12 mm in five steps, with each reduction being about 10% per pass.

The swaged composite tube is then annealed at 350°C for 10 min to make the silver more ductile. Further reductions in diameter are carried out by swaging the ends and drawing the composite tube through a series of dies at a reduction of 10% per pass. Two intermediate annealings are usually necessary to soften the work-hardened silver sheath and to ease processing. When the tube diameter reaches 1.7 mm the rolling process can begin.

A reduction of approximately 10% per pass was maintained. A 13 cm diameter roller was used at a speed of 6 r/min. When the ribbons thickness reached $508\text{ }\mu\text{m}$ a 4 cm long section was cut for the study. A new ribbon sample was taken approximately every $50.8\text{ }\mu\text{m}$.

Equipment

The digital (filmless) radiographic detector system equipment used was designed and built at Argonne National Laboratory (see Fig.1); it consisted of an X-ray detector comprising a 14-bit CCD camera and an image intensifier (II) [6]. The X-ray source is a microfocus system (spot size approximately 10 μm) using a tungsten target and a maximum energy level of 160 kVp at .15 mA. The X-ray system is controlled by a IBM 486 computer, which also stores the raw image data. For image processing the raw image data are transferred to a Sun Sparc10 workstation that runs a shareware software package called Khoros. In addition a Macintosh II FX running a software package called NIH IMAGE is used to acquire gray-scale plots.

In addition to use of the digital (filmless) radiographic detector, ordinary film radiography (subsequently digitized by a 1024 x 1024 x 14 bit CCD camera) was explored. Ribbon samples ranging in thickness from 394 μm to 102 μm were examined using X-ray film as the detector. The resulting films showed approximately the level of sausageing detail in the ribbons as revealed by the direct digital radiography approach. Use of the CCD camera/II has the advantage of being faster and allows the images to be saved directly into the computer, where gray-scale plots and image enhancement is possible.

Procedure

To obtain the high resolution X-ray image data, the ribbon was set perpendicular to the X-ray source so the X-ray penetration was through the rolled flat section. Because the X-ray detector train incorporated an II, externally controlled lead shutters were used to reduce the detector area to that occupied by the ribbon. This also improves the signal-to-noise ratio of the image. The lead shutters allow longer exposure times by preventing saturation (over-ranging) of the detector. This gives a greater gray-scale range in the ribbon and thus better contrast.

The time required to obtain an X-ray image data set ranged from 2 to 4 sec depending on ribbon thickness and the kVp of the x-ray source. The optimal X-ray energy level for imaging varies with the thickness of the

ribbon [7]. An approximate optimal energy level was determined by theoretical calculations and experimental results [8].

Image artifacts may occur due to unevenness in the detector system and dust on the optical components. To remove these artifacts an image correction can be used. Two different methods of image correction were used. The first method was to subtract a blank screen image from the image of the ribbon and then add an offset value to make all the gray-scale values positive. Subtracting a blank screen helped to minimize the artifacts but was not successful in removing them. The second image correction method used flat field correction. It employed the following formula to correct the image:

$$\text{Corrected Image} = \frac{(\text{Sample Image} - \text{Reference}) \times (\text{Mean Pixel Value})}{(\text{Flat Field} - \text{Reference})}$$

This was found to be very effective in correcting for equipment-induced artifacts without damaging the integrity of the image.

Discussion and Results

X-ray image data reveal changes in sample thickness or changes in density of an imaged part. Each ribbon is actually a composite i.e., a silver sheath around a BSCCO-2223 powder core. The silver - with a density of 10.5 g/cm^3 - will absorb more X-rays for a given unit thickness than will BSCCO powder with a density of $5.1 \pm 0.7 \text{ g/cm}^3$. When sausaging occurs during the rolling reduction, the ratio of silver thickness to BSCCO in a given cross section changes, and this change is detected by differences in X-ray attenuation that show on the images. Using digital image processing, one can obtain plots of changes in the gray-scale that allow comparisons of local relative changes throughout the powder core. The amount of relative changes in the gray-scale values in the image determines if sausaging has occurred and its severity. These changes are determined by examining where the larger changes in gray-scale values (higher peaks) occur. The greater the ratio of gray-scale values, the greater the extent of sausaging.

The distance between the peaks in the gray-scale plots allows the spacing of the sausaging to be determined.

An example of the spatial-scale detection sensitivity of X-ray imaging to sausaging can be seen in Fig. 2, which shows an SEM photo and a gray-scale plot from an X-ray image of the same ribbon sample. The peaks on the gray-scale plots represent areas where the BSCCO powder core is thickest. Decreases in the gray-scale values correspond to decreases in the thickness of the BSCCO powder core. It should be noted that Fig. 2 does not show a 1:1 correlation between the X-ray image and SEM photo; because of the small spatial scale, it was difficult to exactly match the location of the sausaging seen with the SEM and the corresponding gray-scale peaking in the X-ray image.

Figure 3 shows X-ray images of four ribbons of different thicknesses: 394, 254, 203 and 102 μm . Figure 4 shows gray-scale plots from the center of the X-ray images shown in Fig. 3. The X-ray image of the 394 μm ribbon shows a uniform powder core with gray-scale peak changes of 250 gray-scale values or less. The X-ray image data for the 254 μm thick ribbon shows a small amount of sausaging. Changes in gray-scale peak values were typically 300, with a few of the largest peaks reaching 450. There is a strong transition from no sausaging to significant sausaging between the 254 and 203 μm ribbons. This seems to be the thickness range during rolling where onset of sausaging occurs. X-ray image data for the 203 μm ribbon shows significant gray-scale changes, with peaks in the 500 range. The X-ray image data for the 102 μm ribbon shows that sausaging increases as the rolling reduction continues. Changes in gray-scale peak values for the 102 μm ribbons commonly reached 1000, with a maximum of 1300. SEM photos (Fig. 5) of cross sections of each ribbon confirm the X-ray image data in Fig 3 and 4.

A total of five sets of ribbons, each from different starting tubes, were examined. All ribbon samples were examined with both SEM and X-ray imaging methods, and the SEM results confirmed the X-ray image data in all cases.

It was also shown during these studies that X-ray imaging can be used to find and examine other types of processing defects. Figure 6 shows a 254 μm thick ribbon with such an example. The series of low-density regions along the left side of the ribbon, at the edge of the powder core, are most likely processing voids. Large second-phase particles would also be detected in the image. The X-ray image would allow sizing and location of voids. This is information that would be difficult to obtain with any other method.

Conclusions

X-ray imaging has been shown to be a reliable and useful nondestructive evaluation technique to evaluate process control of silver-clad BSCCO ribbons produced by the powder-in-tube method. X-ray image data have been shown to allow determination of the onset and severity of thickness variations (sausaging) in the superconducting core as well as detection of processing induced voids. X-ray imaging is faster and provides more information than destructive optical and scanning electron microscopy methods traditionally used. With X-ray imaging, sausaging effects can be established over the entire width of the ribbon, not just at one cross section. The greatest strength of this technique is that it is nondestructive, allowing the same ribbon to be imaged in different stages of processing so that the final properties can then be related back to the X-ray image information.

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Figures

Figure 1: Schematic diagram of Argonne's microfocus X-ray image system.

Figure 2: Correlation of X-ray image data and SEM data to detect onset of sausaging. Gray-scale peaks correspond to regions where powder core is thickest.

Figure 3: Digital X-ray images of BSCCO PIT ribbons of different thicknesses. (a) 394 μm , (b) 254 μm , (c) 203 μm , and (d) 102 μm . Lightest regions have the highest density

Figure 4: Axial gray-scale plots from X-ray image data of BSCCO PIT ribbons of different thicknesses.

Figure 5: SEM photo of axial sections of BSCCO PIT ribbons.

Figure 6: Digital X-ray image of 254 μm thick ribbon showing voids and sausaging. Lighter Areas have lower density.

Figure 1

Figure 2

Figure 3

(A)

(B)

(C)

(D)

Figure 4

Figure 5.

Figure 6