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Use of Neutron Diffraction to Determine Grain-Size Effects on Strain in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ *

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Anisotropic expansion and contraction of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) grains during fabrication could lead to internal stresses that may be relieved by microcracking, resulting in low strength and critical current density. Neutron diffraction techniques have been used to examine the internal strain distribution as a function of grain size in YBCO samples. The full width at half maximum (FWHM) of the Gaussian shaped diffraction peaks is used for strain distribution analysis. The FWHM of the strain distribution for coarse-grain YBCO is found to be about 0.05%, while that for fine-grain material is significantly higher at about 0.3%. The results of this study suggest that the strains are relieved by microcracking in coarse-grain YBCO.

The discovery of superconducting materials with high transition temperatures, such as $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO), has led to a considerable effort to understand and improve the mechanical properties and current-carrying capacity of these materials and thus enhance their practical applications. The anisotropic expansion and contraction of the YBCO grains during fabrication result in internal stresses. These stresses are expected to increase with increasing grain size and to result in microcracking at a critical grain size. This microcracking results in degraded mechanical and superconducting properties ¹.

Therefore, an understanding of the effects of grain size on the magnitude of internal stresses and extent of microcracking will be very critical to the development of microstructures with improved properties. In this communication we report the results of a study to apply neutron diffraction techniques to examine the effect of grain size on strain distributions in YBCO samples.

Neutron diffraction has been applied to the study of microstrains at Argonne National Laboratory through the Intense Pulsed Neutron Source (IPNS) and General Purpose Powder Diffractometer (GPPD). In the IPNS, fast neutrons are moderated and produce beams used by various instruments such as the GPPD, which is positioned about 20 m from the neutron source. Neutrons are detected with banks of ^3He proportional counters (140 total) encircling the sample chamber on a 1.5-m scattered flight path, at 20° , 30° , $\pm 60^\circ$, $\pm 90^\circ$ and $\pm 148^\circ$ relative to the neutron forward direction. Thermal neutrons with velocities of up to 1000 m/s are of interest for neutron diffraction. At these energies, the wavelengths are on the order of the lattice spacing, and Bragg's law of diffraction can then be applied to the neutrons. In the experiments described here, Bragg's law is used first to determine the lattice spacing d for a particular hkl reflection averaged over the sample. The strain distribution is then determined from the full width at half maximum (FWHM) of the various peaks.

The FWHM of the Gaussian shaped diffraction peaks are used for the strain distribution analysis, as described in the following section. The diffraction pattern is presented in time-of-flight units (x-axis).

Time-of-flight is related to neutron wavelength by $\lambda = ht/mL$, where λ is the neutron wavelength, h is Planck's constant, t is the time-of-flight for a neutron to reach a detector after leaving the neutron source, L is the flight path for the neutron from the source to the detector, and m is the neutron mass. Bragg's law, $2d\sin\theta = \lambda$, can then be written as:

$$t = (2dmL/h)\sin\theta \quad (1)$$

where d is the lattice spacing and θ is the diffraction angle.

The widths of the Bragg peaks in the neutron diffraction spectrum were analyzed by the integral breadth method ^{2,3} which assumes that the peak shapes and strain contributions are Gaussian. The contributions to the width of the Bragg peak are from instrumentation, variation in lattice spacing (strain), and/or particle size distribution. The FWHM is corrected for instrument broadening using

$$F^2_{\text{adj}} = F^2_{\text{exp}} - F^2_{\text{standard}}, \quad (2)$$

where F_{adj} , F_{exp} , and F_{standard} are the adjusted, experimentally measured, and standard values of the FWHM acquired with a silicon standard. Particle size and strain can both contribute to broadening in the following way³:

$$F^2_{\text{adj}} = (\lambda/2D\sin\theta)^2 + (4\langle\epsilon\rangle\tan\theta)^2, \quad (3)$$

where D is the particle size and $\langle \epsilon \rangle$ is the FWHM of the strain distribution in the material.

Because the diffraction angle is fixed and the wavelength varies, Eq. (3) can be shown to reduce to

$$(\Delta t/t)^2 = (h/2mLD \sin\theta)^2 t^2 + 4\langle \epsilon \rangle^2, \quad (4)$$

where it can be shown that $(\Delta t/t)^2 = (\Delta\lambda/\lambda)^2$, and a plot of $(\Delta t/t)^2$ vs. $(t)^2$ results in a line with intercept $4\langle \epsilon \rangle^2$ and a slope related to D , the particle size.

YBCO samples were fabricated with different grain sizes. YBCO powder (3-5 μm) was produced by vacuum-calcination and then pressed at 140 MPa into bars measuring 3 x 6 x 10 mm. The bars were then sintered in flowing oxygen. The fine-grained material was fabricated by sintering at 910°C for 10 h to reach densities of 80-85% of theoretical; Coarse-grain samples were made by sintering at 950°C for 20 h, with resulting densities of 85-90% of theoretical. The dominant grain size was about 100 μm for the coarse-grain material and about 5 μm for the fine-grain. Twinning was visible in the coarse grain material.

The fine-grain size samples should have had more twinning present than the coarse-grain samples; this additional twinning could lead to broader lines in the spectra of the fine-grain YBCO spectra ⁴.

Therefore, observed line broadening could be the result of twinning and/or strain. Evaluation of the coarse-grain material indicates that despite the presence of twinning line broadening was minimal. Thus, it is likely that the contribution to the line broadening due to twinning is small and that the line broadening in the fine-grain YBCO is primarily an indication of strain distribution. Some shifts in the position of the diffraction peaks were observed and are attributed to variation in stoichiometry between fine- and coarse-grain material.

Figure 1 is a plot of corrected $(\Delta t/t)^2$ vs. $(t)^2$ for the coarse-grain YBCO. A linear fit to the data is shown. Figure 2 shows the results for the fine-grained sample. Data scatter for the fine-grained samples is much larger and, as a result, the intercept is estimated by eye in this case.

The strain distribution ($\langle \epsilon \rangle$) in the coarse-grain material is very small, as can be seen in Fig. 1. Calculated from Eq. 1 with a linear fit to the data in Fig. 1, strain distribution is about 0.05%. The essentially zero slope implies that particle size effects are not significant. On the other hand, the strain distribution for the fine-grained material calculated from the intercept of about 10^{-5} (Fig. 2) was 0.3%. This is significantly higher than the value for the coarse-grain material.

The anisotropic thermal expansion coefficients could lead to significant residual microstresses in the large-grain YBCO, yet the strains here are lower than those in the fine-grain material. It is likely that the strains are relieved by much more microcracking in the

coarse-grain YBCO although another possibility is that residual stresses result in dislocation arrays that relieve the strain. The microcracking scenario is consistent with the discussion of Richardson and De Jonghe¹ who studied acoustic emissions from YBCO during processing. They present evidence indicating that smaller-grain-size YBCO would have less microcracking (they also point out that alignment of grains may be the most effective way to reduce microcracking). Strains in the smaller-grained samples and resultant stresses do not exceed the fracture strength of the material and thus are not relieved by cracking.

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Fig. 1. Representation of FWHM vs. lattice spacing for coarse-grain YBCO material for several diffraction peaks. Data are presented as a plot of $(\Delta t/t)^2$ vs. $(t)^2$; linear fit to the data is shown. Intercept is $4\langle\epsilon\rangle^2$, where $\langle\epsilon\rangle$ is the FWHM of strain distribution in the material.

Fig. 2. Representation of FWHM vs. lattice spacing for fine-grain YBCO material for several diffraction peaks. Data are presented as a plot of $(\Delta t/t)^2$ vs. $(t)^2$; linear fit to the data is shown. Intercept is $4\langle\epsilon\rangle^2$, where $\langle\epsilon\rangle$ is the FWHM of strain distribution in the material (intercept is estimated in this case because of the data scatter).

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