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The study of texturing of $Bi_2Sr_2CaCu_2O_{8+\delta}$ and $Bi_{1.84}Pb_{0.34}Sr_{1.91}Ca_{2.03}Cu_{3.06}O_{10+\delta}$ superconductors as a function of pelletisation pressure

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Abstract

Crystallographic texture evolution in polycrystalline $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) and $Bi_{1.84}Pb_{0.34}Sr_{1.91}Ca_{2.03}Cu_{3.06}O_{10+\delta}$ (Bi-2223) phases under axisymmetric compression has been investigated. The degree of texturing in these samples is ascertained by X-ray diffraction. The variation of microstructure and electrical resistivity with temperature as a function of pelletization pressure of these samples are studied. $T_{c\,onset}$ changes with pressure in Bi-2212, but not in Bi-2223. The degree of texturing in Bi-2212 increases almost linearly with pressure during cold compression but in annealed samples it reaches a maximum at a pressure of 600 MPa. The optimised pressure needed for single-phase formation of Bi-2223 has been evaluated. Low-T_c phase (Bi-2212) segregation in Bi-2223 occurs at 800 MPa, higher than the pressure (400 MPa) responsible for attaining maximum texturing. Texturing is achieved presumably through two different mechanisms, i.e. by deformation texturing through slip plane movement during cold compression and by recrystallisation texturing through oriented grain growth during annealing.

1. Introduction

In the case of high-T_c superconductors with anisotropic physical properties, the vital feature of the microstructure required for obtaining a large critical current density in its polycrystalline form, is to orient grains along a certain direction, often referred to as texturing. The texturing can be achieved in various ways like melting with directional solidification [1], by changing oxygen vacancy ordering [2], or by mechanical processing [3–6]. The most convenient way of achieving texturing is the application of a uniaxial pressure before final annealing. We had earlier investigated texturing effects in $YBa_2Cu_3O_{7-\delta}$ (YBCO) [7], where we had observed a maximum in the degree of texturing at a certain pressure. In the present work, we have studied the texture evolution and its mechanism in both Bi-2212 and Bi-2223 system under axisymmetric compression through

(1) the analysis of X-ray diffraction pattern,
(2) the examination of the microstructure by scanning electron microscopy (SEM),

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(3) the measurement of electrical resistivity with temperature and
(4) the estimation of the oxygen content with the degree of densification.

2. Experimental

Bi-2212 and Bi-2223 samples were prepared from nitrates with nominal composition by the usual solid-state reaction [8,9]. After calcination at 650°C, two stage sinterings were done at 800°C and 840°C, respectively, for 50 h in powder form for Bi-2212 and in pellet form for Bi-2223 (80 MPa). Pressures employed before the final annealing at 840°C for Bi-2212 were 80, 400, 600, 800, 1000 MPa and for Bi-2223 were 240, 400, 600, 800 MPa. Annealings in air for 100 and 150 h were done for Bi-2212 and Bi-2223, respectively. The samples prepared at 80 MPa for Bi-2223 and the samples prepared from the last sintered powder for Bi-2212 were treated as random for further analysis. The phase identification and the degree of texturing were ascertained by X-ray diffraction with a Philips Diffractometer (PW1730) using Cu Kα ($\lambda = 1.547$ Å) radiation. The surfaces of the samples under investigation of SEM were polished by argon-ion sputtering followed by gold deposition. Critical temperatures of the samples were determined from measurements of the electrical resistivities ρ_{xy} (perpendicular to the application of pressure) as a function of temperature by the standard four-probe technique. The current through the samples was kept at 1 mA. The densities of the samples were measured from weights and dimensions of the pellets [10]. The oxygen contents of samples were estimated by modified iodometric titration [11].

3. Results and discussions

3.1. Texturing of Bi-2212

The initial texturing in the cold-pressed samples takes place due to deformation texturing through slip plane movement. During the axial compression, the direction of major strain is parallel to the direction of application of the pressure. As a consequence, a–c planes being the predominant slip planes having highest density of atoms (8.48 atoms/nanometer square) slip and orient themselves along the direction of the major strain, i.e. parallel to the direction of the axial pressure. This reorientation may cause all $00l$ planes (parallel to the a–b plane) to become almost perpendicular to the direction of application of pressure. Hence the intensities of the diffracted X-ray from the $00l$ planes get enhanced almost linearly (Fig. 1(a)) with an increase of the pressure up to 1000 MPa. In the annealing stage of cold-pressed samples, the degree of texturing is further enhanced by recrystallisation texturing where oriented grain growth takes place through grain-boundary migration [12]. But there is a limit to the growth due to misalignment between the grains. The misalignment reduces with pressure and hence grain growth is

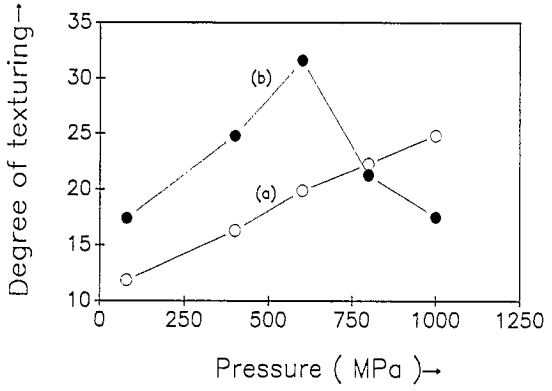

Fig. 1. Degree of texturing of Bi-2212 samples: (a) before annealing of cold-pressed samples under different pelletisation pressures, (b) after annealing of cold-pressed samples under different pelletisation pressures.

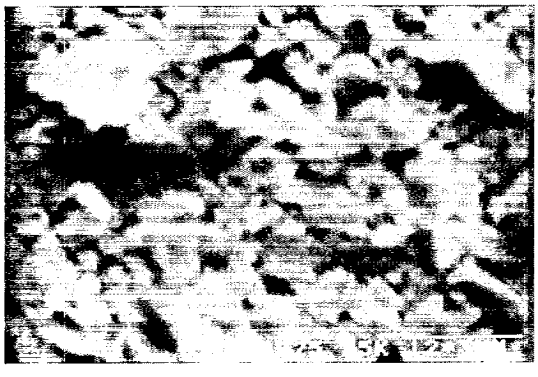

Fig. 2. Scanning electron micrograph (×2500) of an annealed Bi-2212 sample prepared at 600 MPa.

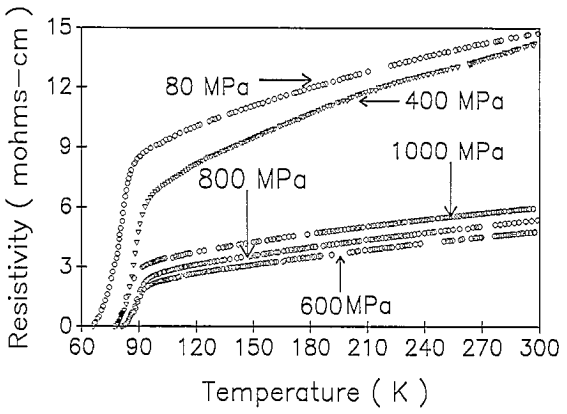

Fig. 3. Variation of the resistivity (ρ_{xy}) of an annealed Bi-2212 sample as a function of temperature and of pelletisation pressure.

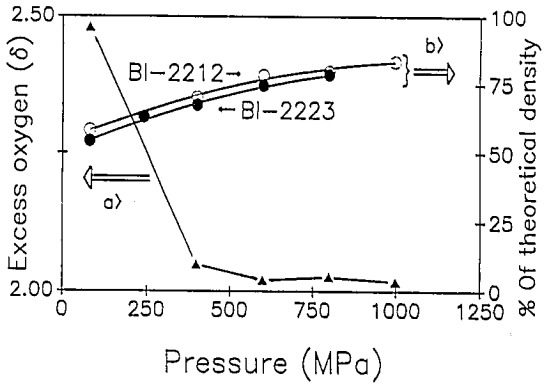

Fig. 4. (a). Excess oxygen content (δ) of annealed Bi-2212 and (b) % of theoretical density (6.566 g/cm^3) of annealed Bi-2212 and Bi-2223 as a function of pelletisation pressure.

enhanced, as is evident from the SEM photograph (Fig. 2) of the sample at 600 MPa. Beyond the pressure of 600 MPa, the degree of texturing deteriorates (Fig. 1(b)) due to the onset of fine grains at the stage of annealing. These fine grains result from the generation of a large number of defects at the stage of cold compression, which causes hindrance to grain-boundary migration.

It is noted that on going from 400 MPa to 600 MPa (Fig. 3), there is a sharp decrease in resistivity which is much larger than the changes observed from 80 MPa to 400 MPa. This large decrease of resistivity is attributed to the predominant effect of grain alignment of texturing rather than intergrain linking as it is reflected in smaller changes in density corresponding to 400–600 MPa as compared to 80–400 MPa (Fig. 4(b)). Densification is supposed to cause better intergrain linking. Beyond 600 MPa, there is a slight increase in the resistivity due to onset of fine grains. Moreover, $T_{c\ onset}$ depends on the intragranular oxygen content (δ) which controls the carrier concentration in the conducting CuO_2 layer and follows a typical bell-shaped curve with δ [13] in Bi-2212. The increase in $T_{c\ onset}$ reflects the lower intragranular oxygen content (δ) which falls in the right-hand side of the bell-shaped curve. We observe that $T_{c\ onset}$ increases with pressure from 80 MPa to 400 MPa and then gets saturated as shown in Table 1. With increasing compression, samples become more densified due to compaction causing less oxygen absorption as shown in Fig.

Table 1
Degree of texturing, resistivity, $T_{c\ onset}$ and T_c ($R = 0$) of annealed Bi-2212 and Bi-2223 samples as a function of pressure of pelletization.

Sample	Pressure (MPa)	Degree of texturing $\Sigma 00l/115$ $l = 6, 8, 10, 12$ for Bi-2212 $l = 8, 10, 12, 14$ for Bi-2223	Resistivity (300 K)	$T_{c\ onset}$ (K)	T_c ($R = 0$) (K)
Bi-2212	0 (random)	3.3			
	80	17.4	14.7	88	66
	400	24.8	14.2	93	79
	600	31.6	4.6	94	83
	800	25.0	5.3	94	81
	1000	21.3	5.9	93	77
Bi-2223	80 (random)	7.6	34.3	115	99
	240	9.2	5.1	115	101
	400	14.7	3.4	115	105
	600	10.6	7.5	115	104
	800	3.4	9.8	115	102

4(a). Beyond the pelletisation pressure of 600 MPa, the deterioration in texturing is due to the onset of fine grains which is also reflected in the increasing ΔT_c, resistivity and decreasing $T_c(R=0)$ as shown in Table 1.

3.2. Texturing of Bi-2223

The single-phase formation in Bi-2223 is more complex than Bi-2212. Bi-2223 phase formation is based on an intergrowth mechanism under diffusion control, where extra Ca–Cu–O admixed particles diffuse to the matrix of Bi-2212 [13]. In contrast to Bi-2212, a minimum pelletisation pressure of 80 MPa is required in Bi-2223 during the sintering stage. This is needed to improve the kinetics of diffusion through compaction and generation of defects in the lattice during cold compression. The kinetics of diffusion also depends on the shape, size and the quantity of the admixed phase particles [10]. At a high pressure, with an increasing number of defects, the concentration of finely dispersed admixed particles also increases which slows the kinetics of diffusion needed for complete phase formation. As a result, a low-T_c phase of Bi-2212 with low degree of texturing is observed at a high pressure of 800 MPa (Fig. 5).

We have observed a sharp decrease in resistivity from 80 MPa to 240 MPa (Fig. 6). This is due to combined effect of compaction (Fig. 4(b)) and grain alignment in contrast to Bi-2212. The decreasing trend persists till the pressure of 400 MPa, where the

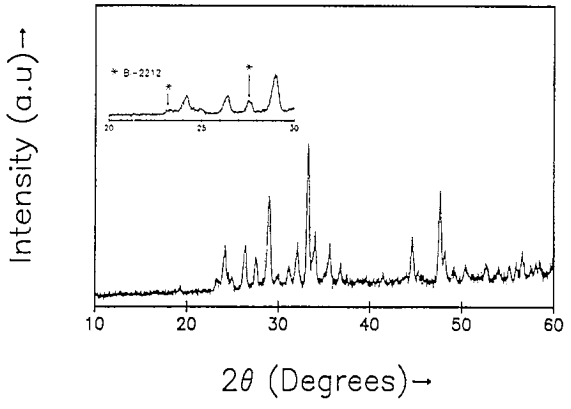

Fig. 5. X-ray diffraction patterns of an annealed Bi-2223 sample compressed under pelletisation pressures of 800 MPa.

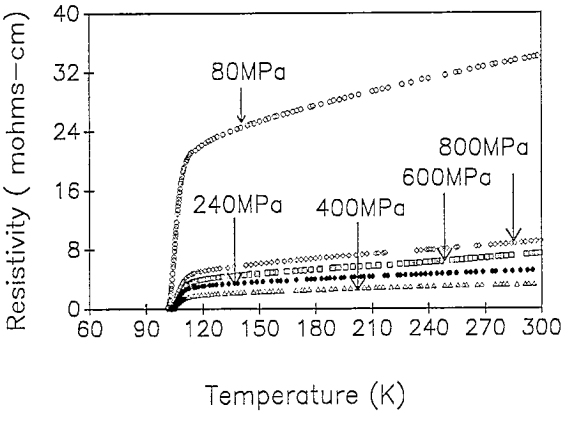

Fig. 6. Variation of the resistivity (ρ_{xy}) of annealed Bi-2223 samples as a function of temperature and of pelletisation pressure.

degree of texturing attains maxima as evident from the XRD analysis and SEM photograph (Fig. 7). Beyond 400 MPa, the resistivity also increases due to weakening of the intergrain links.

The structural stability in these cuprates is governed by a tolerance factor t [14], which is defined as

$$t = (R_{\mathrm{Bi-O}})/(1.4R_{\mathrm{Cu-O}})$$

where $R_{\mathrm{Bi-O}}$ and $R_{\mathrm{Cu-O}}$ are the bond length of Bi–O in the rock-salt block and Cu–O in the perovskite block, respectively. In undoped Bi-2212 t turns out to be 0.78 and is less than the value needed for structural stability ($0.8 < t < 0.9$). Since the Cu–O bond is rigid, the structural stability is attained by accommodating excess oxygen in the Bi–O layer. Hence Bi-2212 is vulnerable to absorb excess oxygen depending on the condition of synthesis and

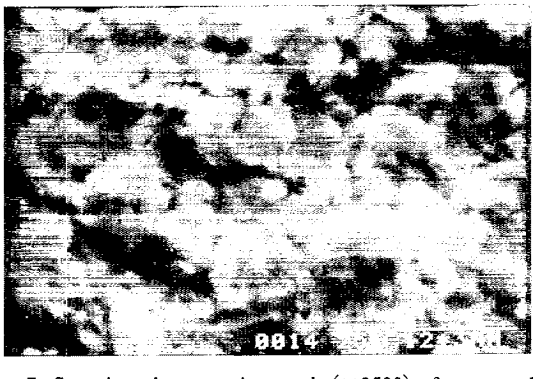

Fig. 7. Scanning electron micrograph (×2500) of an annealed Bi-2223 sample prepared at 400 MPa.

thereby $T_{c\ onset}$ changes with pressure, as discussed earlier. But in Pb doped Bi-2223, due to partial substitution of Bi^{+3} (0.93 Å) by Pb^{+2} (1.2 Å) with a larger ionic radius, the tolerance factor t comes to lie in the range of stability. Hence Bi-2223 does not absorb much excess oxygen ($\delta = 0.14$) unlike Bi-2212 where the oxygen content depends on the condition of synthesis. Hence $T_{c\ onset}$ of the Pb doped material remains unchanged (115 K) with increasing pelletisation pressure.

Based on our earlier work [7] and present studies, we have noticed that the pressures responsible for attaining the highest degree of texturing decreases from YBCO to Bi-2223 namely 800 MPa for YBCO, 600 MPa for Bi-2212 and 400 MPa for Bi-2223. Correspondingly, there is increase in c parameters (11.8 Å in YBCO, 30.7 Å in Bi-2212 and 37.1 Å in Bi-2223). The unit cell with large c parameter is vulnerable to contain a large number of defects responsible for saturation of texturing at low pressure.

4. Conclusion

The highest degree of texturing is achieved at different pressures for two systems (viz. 600 MPa for Bi-2212, 400 MPa for Bi-2223) due to deformation texturing through slip plane movement during cold compression followed by recrystallisation texturing through grain growth at the annealing stage. In contrast to Bi-2212, a minimum pressure of 80 MPa is required in Bi-2223 during the sintering stage to improve the kinetics of diffusion needed for complete phase formation. Moreover in Bi-2223, a high pressure of pelletisation at 800 MPa also causes inhibition of intergrowth of Ca–Cu–O admixed phase to the matrix of Bi-2212. In the case of Bi-2212, $T_{c\ onset}$ increases from 80 MPa to 400 MPa due to a decrease in excess oxygen content. But in Pb doped Bi-2223, there is no appreciable change in excess oxygen content and hence $T_{c\ onset}$. The pressure responsible for the highest degree of texturing is different in three systems (viz. YBCO-123, Bi-2212, Bi-2223) perhaps due to the difference in c parameters.

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References

[1] H. Schluter and H.J. Guntherodt, Physica C 153–155 (1988) 383.
[2] P.S. Mukherjee, A. Simon, M.S. Sharma and A.D. Damodaran, Solid State Commun. 81 (1992) 253.
[3] D.N. Matthews, A. Bailey. S. Town, G. Alvarez, G.J. Russel, K.N.R. Taylor, M. McGirr and D.J.H. Corderoy, in: Adv. Supercond. Proc. 1st Int. Symp. Supercond. (ISS'88), eds. K. Kitazawa and T. Ishiguro (Springer, Berlin, 1988) p. 253.
[4] T. Asano, Y. Tanaka, M. Fukutomo, K. Jikihara and H. Maeda, Jpn. J. Appl. Phys. 28 (1989) L595.
[5] R.J. Asaro, S. Ahzi, W. Blumenthal and A. DiGiovanni, Philos. Mag. A 66 (1992) 571.
[6] J.G. Noudem, J. Beille, D. Bourgault, A. Sulpice and R. Tournier, Physica C 230 (1994) 42.
[7] P. Barat, S.K. Bandyopadhyay, P. Dasgupta, P. Sen, S.K. Kar, P.K. Mukhopadhay and C.K. Majumdar, Physica C 218 (1993) 63.
[8] N. Knauf, J. Hernischmacher, R. Muller, R. Borowski, B. Roden and D. Wohlleben, Physica C 173 (1991) 414.
[9] U. Endo, S. Koyama and T. Kawai, Jpn. J. Appl. Phys. 28 (1989) L190.
[10] V. Sima, K. Knizek, J. Chval, E. Pollert, P. Svoboda and P. Vasek, Physica C 203 (1992) 59.
[11] J.T.S. Irvine and C. Namgung, J. Solid State Chem. 87 (1990) 29.
[12] G.E. Dieter, Mechanical Metallurgy, sec. ed. (McGraw-Hill, Kogakusha) p. 239.
[13] C. Allgeier and J.S. Schilling, Physica C 168 (1990) 499.
[14] H. Zhang and H. Sato, Physica C 214 (1993) 265.