MICROSTRUCTURE AND MAGNETIZATION OF Y-Ba-Cu-O PREPARED BY MELT QUENCHING, PARTIAL MELTING AND DOPING

Hamid Hojaji, Shouxiang Hu and Karen A. Michael
Vitreous State Laboratory, The Catholic University of America
Washington, DC 20064

Aaron Barkatt
Department of Chemistry, The Catholic University of America
Washington, DC 20064

Arthur N. Thorpe
Department of Physics, Howard University
Washington, DC 20059

Sidney Alterescu
National Aeronautics and Space Administration
Goddard Space Flight Center
Greenbelt, MD 20770

ABSTRACT

Y-Ba-Cu-O samples prepared by means of a variety of melt-based techniques exhibit high values for their magnetic properties compared with those of samples prepared by solid-state sintering. These techniques include single-stage partial melting as well as melt quenching followed by a second heat treatment stage, and they have been applied to the stoichiometric 123 composition as well as to formulations containing excess yttrium or other dopants. The structure of these melt-based samples is highly aligned, and the magnetization readings exhibit large anisotropy. At 77 K and magnetic field intensities of about 2 kOe, diamagnetic susceptibilities as high as -14×10^{-3} emu/g have been obtained in the cases of melt-quenched samples and remanent magnetization values as high as 10 emu/g for samples prepared by partial melting.

INTRODUCTION

The attainment of high critical current (J_c) values is the key for numerous potential applications of high-T_c superconducting materials.[1] Critical currents of bulk superconducting oxides, such as $YBa_2Cu_3O_z$ with z values close to 7 (often designated 123), prepared by solid-state sintering, are generally low. Much higher values have been obtained using melt-based processing methods such as partial melting,[2-5] melt quenching and subsequent growth at intermediate temperatures,[6] and melt-textured growth using directional solidification.[7] In addition, variations from the exact 123 stoichiometry, based on increasing the proportion of one of its ingredients (in particular, yttrium) or on the introduction of other metal oxides as dopants, have yielded promising results when combined with melt-based preparative methods.[8,9]

SAMPLE PREPARATION

In the study reported here, specimens were prepared using the partial melting and the melt quench techniques. Sample compositions included stoichiometric $YBa_2Cu_3O_z$, yttrium-enriched $Y_xBa_2Cu_3O_z$ with $x > 1$, and doped $YA_{0.1}Ba_2Cu_3O_z$, with the additive A being a rare earth element or niobium. The preparation techniques are described in Refs. 5 and 9. Briefly, sample preparation involved grinding together a mixture of the oxides of Y, Cu and the additive A, whenever introduced, with $BaCO_3$, in the desired proportions. The mixture was subjected to several cycles of calcination and re-grinding. In the cases of samples prepared by partial melting, the oxide mixtures were rapidly heated to a temperature between 1000°C (the decomposition temperature of $YBa_2Cu_3O_z$[10]) and 1200°C, usually between 1030°C and 1100°C, to form a mixture of Y_2BaCuO_5 and a liquid phase[10]. The sample was then cooled down slowly under oxygen to undergo peritectic conversion to $YBa_2Cu_3O_z$ and to ensure that the material was fully oxygenated to maintain the superconducting orthorhombic structure. In cases where melt quenching was employed, the oxide mixtures were first heated to a temperature above 1400°C to yield a mixture of Y_2O_3 and a liquid phase[10], rapidly cooled down to room temperature, and then re-heated at 1030-1100°C and slowly cooled under oxygen as detailed above. The latter procedure permitted control of the growth of the Y_2BaCuO_5 phase as well as of its $YBa_2Cu_3O_z$ product. In the cases of formulations containing excess yttrium or an additive A, the final product contained a second phase based on Y_2BaCuO_5. The substrates used during the heat treatments included platinum, quartz, alumina, and alumina pre-treated with Y-Ba-Cu oxide powder. The latter, in particular, appeared to minimize the extent of sample deterioration due to interaction with the substrate.[9,11-12]

SAMPLE CHARACTERIZATION

The microstructure of the resulting specimens was characterized by means of scanning electron microscopy (SEM). The magnetic susceptibility, chi, was determined using a Faraday balance and the dc magnetization, M, was measured as a function of magnetic field

intensity using a vibrating sample magnetometer. The latter measurements, carried out at field intensities ranging up to 5 kOe, yielded values for the maximum magnetization, M_{max}, obtained at a low field intensity (H_c^*), as well as for the width of the magnetization hysteresis loop,
Delta M = M_+ - M_-. The latter quantity and its relative magnitude Delta M / M_{max} are of great importance because of the inter-relationship between Delta M and the critical current, J_c.[13,14] The results of the magnetic measurements, reported in Table 1, represent values of the magnetic susceptibility and the magnetization obtained at a temperature of 77 K. The magnetic susceptibilities in Table 1 were measured in a field of 2.25 kOe and the remanent magnetization (Delta M) in a field of 2 kOe. The orientation of each sample relative to the magnetic field was optimized prior to each magnetization measurement.

RESULTS AND DISCUSSION

The results in Table 1 cover a range of samples which include materials of stoichiometric 123 composition, materials containing excess yttrium and materials containing rare earth and Nb additives. Some of the samples were prepared by single-stage partial melting (PM), while in other cases this stage was preceded by a melt quenching (MQ) step, as detailed above. In the cases of all the materials included in Table 1, the critical temperature (T_c), based on the onset of the superconducting transition in the magnetic susceptibility measurements, was observed to be (91 +- 2) K.

The microstructure of the various melt-based samples described above, observed in the SEM studies, is generally highly dense and ordered. Unlike sintered samples, which consist of assemblages of poorly aligned small grains,[5] the melt-based samples consistently exhibit the presence of highly aligned regions which are relatively large (of the order of several millimeters). The microstructure of the last sample in Table 1 is shown in Figs. 1a and 1b.

The results shown in Table 1 indicate high values of the magnetic properties can be obtained in the cases of materials prepared using the techniques described above. The maximum magnetization reported here, which ranges up to 8 emu/g, is significantly higher than that of typical materials prepared by solid-state sintering, with values of up to about 2 emu/g.[5] More significantly, the ratio of remanent to maximum magnetization in the melt-based materials is high, ranging about 1 and, in certain cases, even higher. As a result, the remanent magnetization itself is high, reaching values up to 10 emu/g. This indicates that improvements in melt-based processing yield a higher degree of magnetic flux pinning at high fields, which is associated in its turn with higher values of J_c. The magnetization curve of the last sample in Table 1 is shown in Fig. 2. As mentioned above, the magnetization of each sample was measured at different orientations of the sample with respect to the direction of the magnetic field. In general, the magnetization is highly dependent on the orientation of the layered 123 grain structure with respect to the

TABLE 1

Magnetic Properties of Materials Prepared by the Partial Melting and Melt Quench Methods at 77 K

Composition, YxAyBa2Cu0z x	A	y	Method	Chi emu/g X1000	Delta M emu/g	Mmax emu/g	Delta M /Mmax	Hc1* Oe
1.0	Tb	0.1	MQ	-14.3	2.0	2.9	0.68	165
1.1			MQ	-12.9	2.4	4.2	0.57	205
1.0			MQ	-11.1	2.5	4.0	0.64	236
1.0			MQ	-11.7	3.1	4.3	0.71	218
1.0	Tb	0.1	MQ	-13.2	3.6	5.3	0.67	196
1.0			MQ	-10.4	3.9	5.7	0.69	237
1.0	Nb	0.1	MQ	-2.2	4.3	6.0	0.72	246
1.0	Ho	0.1	MQ	-1.9	4.4	5.0	0.89	67
1.0	Nb	0.1	MQ		4.8	5.1	0.99	105
1.0			PM		5.0	4.8	1.04	165
1.0	Tb	0.1	PM	-2.1	5.2	3.4	1.52	153
1.1			MQ	-5.9	5.4	3.9	1.38	151
1.0			PM	-3.4	5.7	4.5	1.27	219
1.0			PM	-5.1	7.3	6.1	1.19	281
1.0			PM	-5.1	7.7	8.0	0.97	297
1.0			PM		9.8	7.9	1.24	231

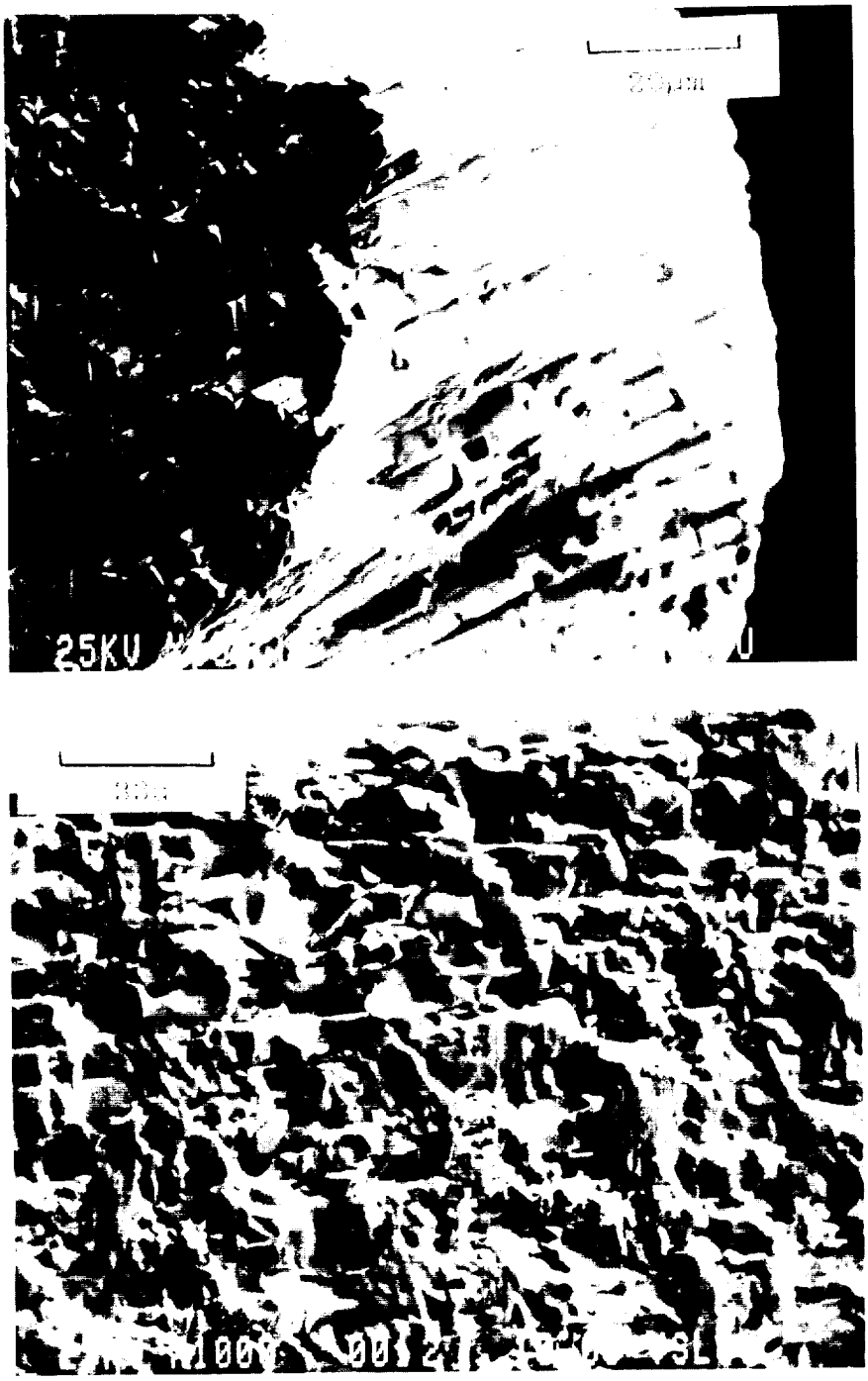

Figure 1. Scanning electron micrographs of a high-magnetization $YBa_2Cu_3O_7$ sample prepared by the partial melting technique.

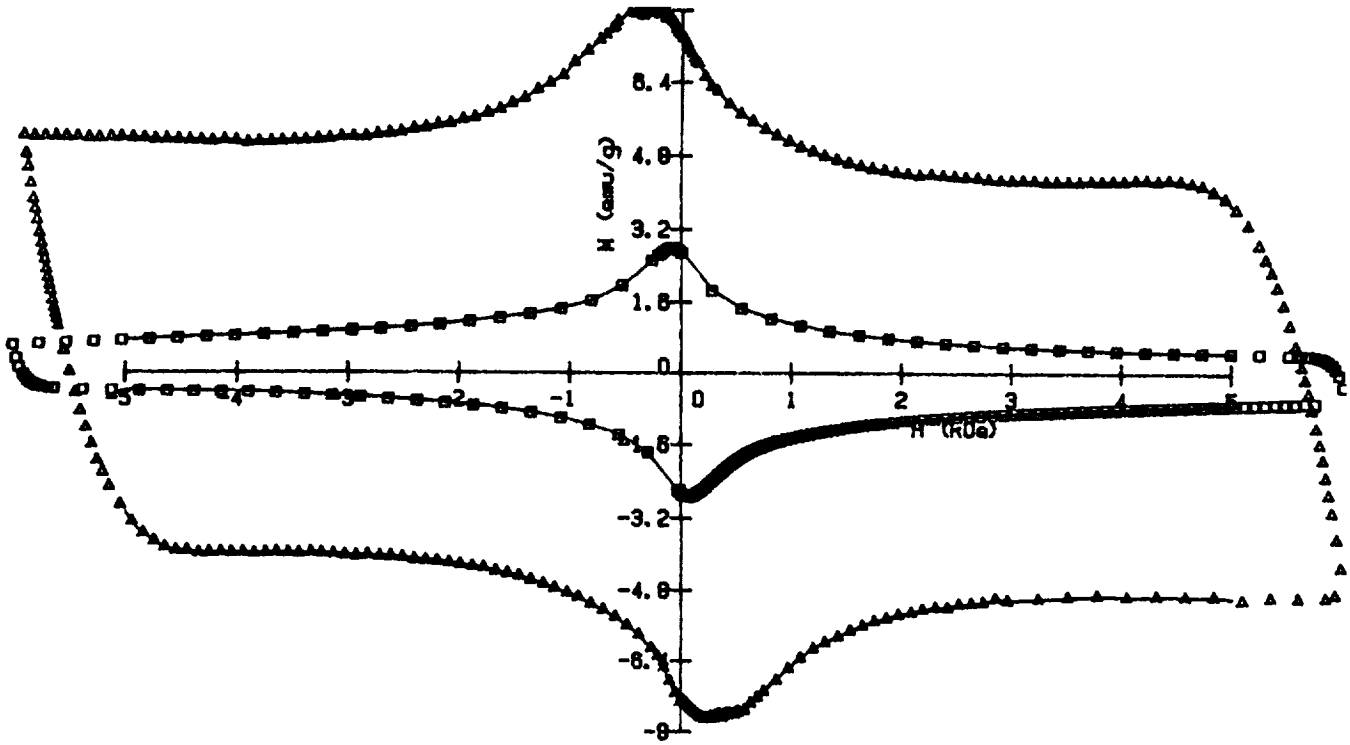

Figure 2. Magnetization hysteresis loop at 77 K for the sample shown in Figure 1. Triangles - field parallel to c axis; squares - field perpendicular to c axis.

applied field. This large anisotropy is reflected in Fig. 2, which shows that the magnetization obtained with the magnetic field perpendicular to the layered structure is much higher than the magnetization obtained when the field is parallel to the 123 layers. These findings are consistent with the notion that the layers are grown in a direction parallel to the ab plane, and that the maximum magnetization is obtained when the applied field is perpendicular to this plane, while the current density is highest parallel to the ab plane.[15] Similar findings were previously reported in magnetization measurements on single crystals.[16]

The formation of more quantitative conclusions on the basis of the data shown in Table 1 is hampered by the irreproducibility among readings obtained with samples prepared using an identical, or near-identical, procedure. This irreproducibility may be due to several factors, including the varying extent of completion of the oxygenation of the samples in the course of the final cooling stage, which is highly dependent on sample dimensions, and the presence of cracks, resulting from the evolution of excessive stresses at grain boundaries during the growth of the 123 phase from the combination of Y_2BaCuO_5 and liquid. The remaining differences in magnetic properties among the various melt-based samples may reflect differences in the extent of mutual alignment among the regions of highly aligned grains, which are shown in Figs. 1a and 1b, in different samples. The degree of domain alignment depends in its turn on the number and size of the domains of parallel 123 crystals and is highly sensitive to the thermal history of each sample.

With these limitations in mind, the data in Table 1 suggest that all three techniques used in the present study (partial melting, melt quenching, use of non-stoichiometric formulations) produce improved magnetic properties compared with those of sintered samples (see above). This may reflect the effectiveness of melt-based techniques and the introduction of dopants, accompanied by slow cooling during the growth of 123 from the melt and its oxygenation, in minimizing the formation of pores, cracks and insulating phases such as $BaCuO_2$[9]. The data obtained so far are not sufficient to determine conclusively which of these techniques could give rise to the highest magnetization. It should be noted, however, that increased magnetization does not appear, in general, to be associated with high diamagnetic susceptibility (see Table 1). The highest values of the diamagnetic susceptibility are encountered in the cases of samples prepared by means of the melt quench technique, but the samples which have the highest susceptibility do not exhibit a very high magnetization. On the other hand, among the samples characterized in the present study, those prepared by partial melting have so far yielded the highest magnetization values, as well as somewhat higher ratios of remanent to maximum magnetization, indicative of highly effective pinning of the magnetic flux.

ACKNOWLEDGEMENT

The authors are very grateful to Dr. A.C. Buechele for his assistance in performing the SEM work. This study was supported by the National Aeronautics and Space Administration under Contract no. NAG 5-1017.

REFERENCES

1. D.U. Gubser, S.A. Wolf and R.J. Soulen, Science, 245, 111-112 (1989).
2. M.E. McHenry, J. McKittrick, S. Sasayama, V. Kwapong, R.C. O'Handley and G. Kalonji, Phys. Rev. B, 37, 623-626 (1988).
3. E. L. Venturini, D.S. Ginley, R.J. Baughman, B. Morosin and J.F. Kwak, in High-Temperature Superconductors, eds. M.B. Brodsky, R.C. Dynes, K. Kitazawa and H.L. Tuller, Mater. Res. Soc. Symp. Proc., Vol. 99, Materials Research Society, Pittsburgh, PA, 1988, pp. 639-642.
4. K. Salama, V. Selvamanickam, L. Gao and K. Sun, Appl. Phys. Lett., 54, 2352-2357 (1989).
5. H. Hojaji, K.A. Michael, A. Barkatt, A.N. Thorpe, M.F. Ware, I.G. Talmy, D.A. Haught and S. Alterescu, J. Mater. Res., 4, 28-32 (1989).
6. M. Murakami, M. Morita and N. Koyama. Jpn. J. Appl. Phys., 28, L1125-L1127 (1989).
7. S. Jin, T.H. Tiefel, R.C. Sherwood, M.E. Davis, R.B. van Dover, G.W. Kammlott, R.A. Fastnacht and H.D. Keith, Appl. Phys. Lett., 52, 2074-2076 (1988).
8. Y. Wadayama, K. Kudo, A. Nagata, K. Ikeda, S. Hanada and O. Izumi, Jpn. J. Appl. Phys., 27, L1441-L1224 (1988).
9. H. Hojaji, A. Barkatt, K.A. Michael, S. Hu, A.N. Thorpe, M.F. Ware, I.G. Talmy, D.A. Haught and S. Alterescu, J. Mater. Res., 5, xxx-xxx (April 1990).
10. M. Murakami, M.Morita, K.Doi, K. Miyamoto and H. Hamada, Jpn. J. Appl. Phys., 28, L399-L401 (1989).
11. W.N. Wang, H.B. Lu, W.J. Lin, P.C. Yao, H.E. Hsu, M.F. Tai and H.C. Ku, Jpn. J. Appl. Phys., 273, L1268-L1270 (1988).
12. K. Yoshiara, K. Kagata, S. Yokoyama, T. Hiroki, H. Higuma, T. Yamazaki and K. Nakahigashi, Jpn. J. Appl. Phys., 27, L1492-L1494 (1988).
13. C.P. Bean, Phys. Rev. Lett., 8, 250-253 (1962).
14. W.A. Feitz, M.R. Beasley, J. Silcox and W.W. Webb, Phys. Rev., 136, A335-A345 (1964).
15. Y. Nakagawa, H. Yamasaki, H. Obara and Y. Kimura, Jpn. J. Appl. Phys., 28, L547-L550 (1989).
16. T.R. Dinger, T.K. Worthington, W.J. Gallagher and R.L. Sandstrom, Phys. Rev. Lett., 58, 2687-2690 (1987).