High critical current density and improved irreversibility field in bulk MgB2 made by a scaleable, nanoparticle addition route

J. Wang, Y. Bugoslavsky, A. Berenov, L. Cowey, A. D. Caplin, L. F. Cohen, and J. L. MacManus Driscoll

Centre for High Temperature Superconductivity, Imperial College, Prince Consort Road, London SW7 2AZ, United Kingdom

L. D. Cooley, X. Song, and D. C. Larbalestier

Applied Superconductivity Center, University of Wisconsin–Madison, 1500 Engineering Drive, Madison, Wisconsin 53706-1687

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Bulk samples of MgB2 were prepared with 5, 10, and 15 wt % Y2O3 nanoparticles, added using a simple solid-state reaction route. Transmission electron microscopy showed a fine nanostructure consisting of ~3–5 nm YB4 nanoparticles embedded within MgB2 grains of ~400 nm size. Compared to an undoped control sample, an improvement in the in-field critical current density Jc was observed, most notably for 10% doping. At 4.2 K, the lower bound Jc value was ~2 × 106 A cm−2 at 2 T. At 20 K, the corresponding value was ~8 × 104 A cm−2. Irreversibility fields were 11.5 T at 4.2 K and 5.5 T at 20 K. © 2002 American Institute of Physics. [DOI: 10.1063/1.1506184]

In slightly more than one year after the discovery of superconductivity in magnesium diboride, there is now a wide body of evidence indicating that MgB2 does not contain intrinsic obstacles to current flow between grains, unlike the high-temperature superconducting cuprates. Evidence for strongly coupled grains has been found even in randomly aligned, porous, and impure samples, suggesting that dense, strongly coupled grains has been found even in randomly high-temperature superconducting cuprates. Evidence for intrinsic obstacles to current flow between grains, unlike the wide body of evidence indicating that MgB2 does not contain superconductivity in magnesium diboride, there is now a...
Figure 1(a) shows the results of x-ray diffraction (XRD) analyses for the series of doped samples compared to an undoped sample. In addition to MgB$_2$, small quantities of MgO and, in the doped samples, YB$_4$, are indicated; there are no peaks corresponding to either pure Mg or Y$_2$O$_3$. Hence, in the doped samples, it can be concluded that the Y$_2$O$_3$ reacted with B to form YB$_4$. Since this decreased the amount of B available for reaction to MgB$_2$, excess Mg either was transported away from the pellet or became oxidized. Indeed, an increase of the MgO peak intensities correlates with increasing Y$_2$O$_3$ fraction in Fig. 1(a). The volume of the MgB$_2$ unit cell was calculated as a function of Y$_2$O$_3$ fraction, which is shown in Fig. 1(b).

Figure 2(a) shows a transmission electron microscopy (TEM) diffraction contrast image of the 10% Y$_2$O$_3$ sample. The grain size of the MgB$_2$ is 400 nm and precipitates at two different levels are seen. Precipitates with 10 nm size occur at the MgB$_2$ grain boundaries (region 1), while inside the MgB$_2$ grain interior (region 2), evenly distributed, 3–5 nm precipitates are seen. A magnification of region 2 is shown in the inset of Fig. 2(a). Selected area diffraction patterns taken from both regions were very similar. Figure 2(b) shows the diffraction pattern along the MgB$_2$ [120] direction. YB$_4$ ring pattern is outlined.

Critical temperature $T_c$ values were obtained by measuring the magnetic moment versus temperature $m(T)$ using a vibrating sample magnetometer (VSM), shown in Fig. 3. Samples were zero-field cooled and then warmed from 10 K in an applied field of 5 mT. Similar transitions, with an onset at $\sim 39$ K and an endpoint at $\sim 38$ K, are seen for the control, the 5% and 10% Y$_2$O$_3$ samples, but these values are reduced by $\sim 1$ K for the 15% sample.

Figure 4 shows $J_c(H)$ at 20 K for the series of doped MgB$_2$ samples, as well as the undoped sample, a fragment of a sample from a commercial source, the 10 at. % Zr-doped sample of Feng et al. and high-pressure synthesized MgB$_2$. The inset of Fig. 4 shows $J_c(H)$ at 4.2 K for the 10 wt. % Y$_2$O$_3$ doped sample. All samples were measured in a perpendicular magnetic field of 10 T.
VSM and the Bean model was used to deduce the critical current density from the magnetization hysteresis. For our measurements, in fields of <1 T the apparent plateau in $J_c$ is artificial, due to saturation of the magnetometer. Therefore, the actual values are higher than shown. Our $J_c$ values are based on full sample connectivity and are multiplied by 2 to allow for the 50% porosity. In fact, associated with the porosity is a reduction in the grain-to-grain contact area, and so a restriction of the cross section available for current flow.

Also, the grain boundaries in the doped samples are partially obstructed by precipitates associated with the porosity. In addition, the macroscopic crack through the undoped sample was also reduced by the 50% porosity. In fact, associated with the porosity is a reduction in the grain-to-grain contact area, and so a restriction of the cross section available for current flow.

FIG. 4. $J_c(H)$ at 20 K for the series of Y$_2$O$_3$-doped MgB$_2$ pellet samples, as well as an undoped pellet, a fragment of a sample from a commercial source (Alfa Aesar). For comparison, the 10 at. % Zr-doped sample of Feng et al. (Ref. 15) and high-pressure synthesized MgB$_2$ (Ref. 19) are included. Inset shows $J_c(H)$ at 4.2 K for the 10 wt % Y$_2$O$_3$-doped sample.

The rapid formation technique used in this experiment apparently produced different superconducting properties, relative to those of high-pressure synthesized bulk, in all of the samples. However, at <4 T a higher irreversibility field and higher critical current density are seen for the doped samples, suggesting the additional effects of the nanoparticles. At >5 T, the undoped sample outperforms the doped samples, but this is most likely related to the greater connectivity of grain boundaries in the undoped sample, the doped samples having additional phases at the grain boundaries.

Interestingly, the samples show similar $J_c$ and $\mu_0 H^*$ values to a recent report describing heavily ball milled, nanocrystalline powders of Gumbel et al. However, ball milling also reduced $T_c$ to 34.5 K, suggesting disorder or possibly alloying (from the milling process). Reduced $T_c$ in the Zr-doped samples is also indicative of alloying. In the present work, the critical temperature remains near 39 K, and there is a weak increase (if any) in the unit cell volume. We believe, therefore, that the nanoparticle additions neither alloyed the surrounding MgB$_2$, nor produced significant disorder. The observed increase in $\mu_0 H^*$ may be due to increasing the number of point scattering sites, since the observed precipitates are 3–5 nm in size and uniformly distributed within the grains.

In summary, we have shown that incorporating Y$_2$O$_3$ nanoparticles together with Mg and B powders results in the formation of MgB$_2$ with a uniform dispersion of YB$_4$ nanoparticles. This nanostructure was achieved using a reaction at 900 °C for 15 min. The precipitates have 3–5 nm size, with larger ~10 nm precipitates occurring at some grain boundaries. At 20 K, the critical current density deduced by magnetization is $>10^5$ A/cm$^2$ in low fields, comparable to that of high-pressure synthesized bulk. Significant increases in the irreversibility field were also observed.

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