Enhancement of Electrical and Mechanical Properties in Nanosize MgO Added Bi$_2$Sr$_2$CaCu$_2$O$_8$ Superconductor Ceramics

N. A. Hamid, N. F. Shamsudin, and K. W. See

Department of Engineering Sciences and Mathematics,
College of Engineering, Universiti Tenaga Nasional, 43009 Kajang, Malaysia
(Tel.: 603-89287252; e-mail: Nasri@uniten.edu.my)

Abstract: In this study, 3 % to 8 % weight percentage of nanosize MgO particles was added to Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi-2212) high-temperature superconductor in view of fabricating Bi-2212 superconductor elements with superior electrical and mechanical properties. The Bi-2212/MgO compounds were palletized and heat treated, followed by partial melting and slow-cooling. X-ray diffraction (XRD) was used to study the phases present in the samples. Scanning electron microscopy (SEM) with energy dispersive x-ray (EDAX) analysis was performed to investigate the microstructure, and for identifying the elemental composition of the samples. The electrical properties were studied via dc electrical resistance, transition temperature, $T_c$, and critical current density, $J_c$, at 77 K. The XRD patterns of the Bi-2212/MgO compounds revealed the Bi-2212 as the dominant phase. EDAX analysis confirmed the presence of MgO in each of the nanosize MgO added sample. Furthermore, the SEM micrographs of the samples with more than 5% additions showed a more porous texture. The nanosize MgO addition did not affect $T_c$ of the Bi-2212 superconductor. However, the $J_c$ in the Bi-2212/MgO compounds decreased drastically for samples with MgO addition of more than 5%. The mechanical properties of the samples was studied by conducting the compression test at room temperature and from the results, the addition of 5% MgO particles produced the highest strength when compared with the other samples.

Keywords: High-temperature superconductor, electrical properties, mechanical properties, high current density, cryogenic temperatures.

1. INTRODUCTION

Fabrication of high-temperature superconductor ceramics with optimum electrical properties and stable mechanical behavior is essential for application in power engineering system such as fault current limiter and energy storage. In such applications, the Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi-2212) superconductor ceramics is an outstanding candidate for consideration due to its phase stability, high transition temperature ($T_c$), and excellent transport critical current density ($J_c$). For practical applications of high-temperature superconductors in power engineering, they must be fabricated into composite elements or sheathed tapes with the required microstructure to provide electrical and mechanical stability. In the application as a resistive superconducting current limiter for instance, the Bi-2212 bulk superconductor is an excellent candidate for consideration due to its homogeneous voltage characteristics over the whole bulk (Elschner et al., 2001). A process known as composite reaction texturing (CRT) is among the method employed for forming large highly textured bulk Bi-2212 superconductors with fully connected high quality grain boundaries (Watson et al., 1995). In the CRT method, inert MgO whiskers or fibers were employed to align the Bi-2212 superconducting grains and control their morphology, and thus produced an extremely high $J_c$ anisotropy (Watson et al., 1995; Soylu et al., 1993; Chen et al., 1995). Composites with different characteristic whisker alignment resulted in different types of textured microstructure and influenced the critical current density, $J_c$. On the other hand, lower degree of texturing is associated with the weak-link behavior and weak flux pinning center (Wei et al., 1998). Although the weak-link problem has been partially solved in Bi-2212 superconductor by partial-melt processing, the limitation from flux pinning, which determines the intrinsic critical current density, remains the most serious challenge for the practical application of Bi-2212 at elevated temperature. Therefore, an optimum technique for increasing the flux pinning in Bi-2212 has attracted a lot of interests from various groups of researchers (Wei et al., 1998; Ni et al., 1999; Agranovski et al., 2006). The additions of small MgO particles to Bi-2212 bulk and its effect on flux pinning were studied in order to introduce effective pinning centers into Bi-2212 bulk superconductor. To increase flux pinning in the Bi-2212 superconductor, the MgO particles must be trapped within the Bi-2212 superconducting grains as second-phase defects. It is believed that the extra pinning centers are due to the interface between MgO particles and the superconductor matrix. It has also been reported that the
improvement in $J_c$ could be achieved by improving the intergranular connectivity in high-$T_c$ superconducting bulk materials (Ren et al., 1994). The introduction of nanosize particles that resided in between the Bi-2212 superconducting grains would provide the perfect linkage for intergranular connectivity. Thus, enhancement in $J_c$ could also been attributed to improvement in texturing of the microstructure.

Nevertheless the Bi-2212 superconductor, like other high-temperature superconductors has poor mechanical properties such as low stiffness, strength and toughness. Low strength and low irreversible strain are among the factors that hindered the application of superconductor in power industry. In view of improving the mechanical property of Bi-2212 superconductor, there are numerous studies conducted by various groups. Nomura et al. (1994) have studied the influence of Ag-Au and Ag-Cu alloys on the critical current density, $J_c$ of Bi-2212 superconductors. Li et al. (1997) have discovered the enhancement of flux pinning in Bi-2212 single crystals by planar defects introduced via Ti-substitution. In all the studies, the behavior of bulk Bi-2212 superconductor with added elements is completely different from that of ordinary Bi-2212 superconductor. Parameters such as $T_c$ and $J_c$ are completely altered with the addition or substitution of foreign elements into the compound. Chen et al. (2003) have shown that MgO fibers could be used to reinforce composite materials and are be able to improve the mechanical properties of the compounds.

In view of fabricating highly textured Bi-2212 superconductor with high $J_c$ and excellent mechanical stability, this paper presents the effect of nanosize MgO additions on the electrical and mechanical properties of Bi-2212 phase superconductor. Optimum superconductivity properties with significant enhancement in $J_c$ are observed in samples with 5% MgO addition where lower grains orientation of Bi-2212 phase is observed. The addition of MgO minimize the high angle grain boundaries in the path of transport current, which should be parallel to the ab plane (Naylor et al., 1999). The nanosize MgO particles are found to reinforce the mechanical properties of the Bi-2212/MgO compounds.

2. EXPERIMENTAL DETAILS

2.1 Synthesis of Materials

High-purity Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi-2212) powder, product of Alfa, USA with an average size of 5 μm was used in preparation of all the samples. The powder was mixed with nanosize MgO particles and grounded thoroughly. The nanosize MgO powder was product of Aldrich, USA with an average grain size of 100 nm and purity of 99.998%. Addition of MgO particles into the Bi-2212 powder was performed with molar fraction of 0%, 3%, 5%, and 8%, respectively. The powders were thoroughly mixed, followed by calcination for 48 hours at 830 °C. The calcination procedure was repeated twice to ensure homogeneity of the powder. The mixed powder was then reground and pressed into pellets with diameter of 13 mm using the hydraulic pellet press. The pellets were subjected to regimental heat treatments schedule as shown in Figure 1 with partial-melting temperature, $T_m$ of 865 °C. Partial melt processing of Bi-2212 significantly improve the microstructure of Bi-2212 phase superconductor.

![Fig. 1. Heat treatments schedule in partial melting process of Bi-2212/MgO compounds](image)

2.2 Characterization of Samples

The microstructure of the samples was investigated and analyzed by a Zeiss SUPRA 35 field emission scanning electron microscopy (FESEM) and energy dispersive x-ray (EDAX). The x-ray diffraction (XRD) patterns were recorded on a Siemens D5000 diffractometer using Cu Kα radiation. Electrical resistance measurements were carried out using the standard four-probe DC method with silver paint contacts in zero magnetic fields. The experimental set-up consisted of a Keithley multimeter (model 197), a closed cycle refrigerator from CTI Cryogenics (model 22), a temperature controller from Lake Shore (model 330), and a constant current source (Keithley 220). Zero transition temperature, $T_c$ zero was determined as the temperature at which the electrical resistance dropped to zero. The onset transition temperature, $T_c$ onset was taken as the temperature at which the tangent of the resistance against temperature curve at the normal state during cooling intersected the tangent of the part where the resistance dropped precipitously.

The transport critical current ($I_c$) were measured on bar-type samples with cross sectional area of approximately 0.2 cm x 0.7 cm. The critical current measurements were performed with criterion of 1 μV/cm at 77 K. The transport critical current density, $J_c$ was defined as the critical current divided by the cross-sectional area of oxide layer. The compression test of the samples was conducted using Instron Material Testing System model 5567, and the digital electronic model 647.25 was used for samples added with more than 5% nanosize MgO additions. The maximum load was recorded along with stress-strain data.
3. RESULTS AND DISCUSSION

3.1 Characterization

The Bi-2212 phase superconductor grains are known to have a platelet-like shape and thus their morphology are able to be controlled and fabricated into well-textured microstructure (Pavard, 1998; Buhl et al., 1996). The SEM micrographs in Figure 2 (a) to (d) show show the microstructure of Bi-2212 phase with addition of 0%, 3%, 5%, and 8% nanosize MgO, respectively. The size of the Bi-2212 platelets remains about the same in all the samples with an average size of about 1 μm with tendency to decrease in samples with higher additions.

![SEM micrographs](image)

Fig. 2. SEM micrograph for Bi-2212 superconductor with (a) 0%, (b) 3%, (c) 5%, and (d) 8% nanosize MgO addition, respectively

![EDAX analysis](image)

Fig. 3. EDAX analysis for Bi-2212 with 5% MgO addition

From the SEM micrographs and EDAX analysis in Figure 3, MgO particles are likely to reside near the grain boundaries of the Bi-2212 phase. When particles such as the MgO are added into a superconductor compound, they will exist either by substituting themselves at some suitable atomic sites or they will segregate as impurity phases and not become part of the structure (Jha et al., 1996). There are also cases in which both situations occurred.

Figure 4 shows the XRD patterns of the Bi-2212/MgO compounds that show a well defined peaks, all of which could be indexed on the basis of a Bi-2212 phase structure. The (002) peak, which is the characteristic of the Bi-2212 phase, can be observed clearly in all the samples. A few peaks which correspond to secondary phase such as Bi-2201 phase and other impurities are also observed. Those unidentified peaks are probably due to an inappropriate heat treatment during the sintering process which resulted in unidentified secondary phases. The MgO peak could not be observed due to low concentration of MgO additions. The MgO (111) peak should be located at 2θ = 36.8° (Shao et al., 2006). Nevertheless, the presence of nanosize MgO particles has been confirmed by the EDAX analysis. Thus, for samples with MgO additions, the SEM and XRD results showed that the nanosize MgO particles are segregated at the grain boundaries, and do not become part of the Bi-2212 phase structure.

![XRD patterns](image)

Fig. 4. XRD patterns of Bi-2212 phase with 0%, 3%, 5%, and 8% nanosize MgO addition, respectively

From the results of SEM micrographs and XRD patterns, a small addition of nanosize MgO particles of about 5% clearly improved the texture of the microstructure and thus enhanced the contact between the Bi-2212 phase grains. The superconducting grains in the samples are seen to align in much lower degree of orientation. The evidence could be clearly observed if comparison is made with the microstructure of the non-added sample. Nevertheless, for sample with 8% MgO addition, the SEM microstructure shows higher porosity and subsequently less contact existed between the adjacent Bi-2212 phase grains. Thus, lower $J_c$ could also be
attributed to the high porosity microstructure. In samples with high MgO addition, the orientation of the grains is much more perpendicular to the c-axis. As such, microstructures with lower grains orientation exhibited much higher transport critical current density, $J_c$. Therefore, beside the flux pinning factor, the intergranular connectivity that were provided by small addition of nanosize MgO particles, plays a very significant role in enhancing the $J_c$ of the samples.

**Table 1. Lattice parameters a, b, c for Bi-2212/MgO compounds**

<table>
<thead>
<tr>
<th>MgO addition</th>
<th>a (Å)</th>
<th>b (Å)</th>
<th>c (Å)</th>
<th>Volume (Å³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 %</td>
<td>5.401</td>
<td>5.396</td>
<td>30.78</td>
<td>896.9</td>
</tr>
<tr>
<td>3 %</td>
<td>5.413</td>
<td>5.400</td>
<td>30.91</td>
<td>903.3</td>
</tr>
<tr>
<td>5 %</td>
<td>5.413</td>
<td>5.340</td>
<td>30.80</td>
<td>900.3</td>
</tr>
<tr>
<td>8 %</td>
<td>5.413</td>
<td>5.400</td>
<td>30.97</td>
<td>905.2</td>
</tr>
</tbody>
</table>

Table 1 shows the lattice parameters $a$, $b$, and $c$ for the Bi-2212 phase superconductor with nanosize MgO addition of 0%, 3%, 5%, and 8%, respectively. As expected, the $c$ parameter for each sample increases with the increased in nanosize MgO addition due to the presence of the MgO particles in the compounds. But there is no systematic variation of the $c$ parameter, and this indicates that the addition of nanosize MgO does not stimulate a new degree of Bi-2212 phase superconductor grains. Thus, as mentioned earlier, the Bi-2212 structure remains unchanged and there is no solubility of MgO particles in the Bi-2212 phase.

**Table 2. The transport critical current density, $J_c$, and the transition temperature, $T_c$,zero and $T_c$,onset for Bi-2212/MgO compounds at 77 K**

<table>
<thead>
<tr>
<th>MgO addition</th>
<th>$J_c$ (A/cm²)</th>
<th>$T_c$,zero (K)</th>
<th>$T_c$,onset (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 %</td>
<td>1.44</td>
<td>84</td>
<td>96</td>
</tr>
<tr>
<td>3 %</td>
<td>1.10</td>
<td>84</td>
<td>95</td>
</tr>
<tr>
<td>5 %</td>
<td>2.07</td>
<td>84</td>
<td>99</td>
</tr>
<tr>
<td>8 %</td>
<td>0.93</td>
<td>80</td>
<td>99</td>
</tr>
</tbody>
</table>

Table 2 shows the results of critical current density, $J_c$, at 77 K, the transition temperature, $T_c$,zero, and $T_c$,onset for Bi-2212 superconductor with nanosize MgO addition of 0%, 3%, 5%, and 8%, respectively. The results show a distinct degrading in $J_c$ in the MgO added sample with 8% addition. This is due to the disruption of the connectivity among Bi-2212 superconducting grains by the MgO particles which resided near their boundaries. Furthermore, with higher MgO addition, the MgO particles do not act as pinning centers or contribute to flux pinning.

**3.2 Compression test**

The strength of the Bi-2212/MgO compounds was conducted using compression test at room temperature in which the plastic flow behavior and ductile fracture limits of the Bi-2212/MgO compounds was studied. Figure 5 shows the results of the compression test and a linear relationship between the load and the compression extension is observed until they reached the fracture limits. Thus, at room temperature the mechanical strength of the bulk Bi-2212 superconductor increases with the addition of MgO particles. Therefore, the higher mechanical strength of the superconductor is attributed to the presence of MgO particles. However, the strength is found to deteriorate with 8% MgO addition. As shown in the SEM micrograph, the porosity resulted in the formation of microcracks at the interface between Bi-2212 grains and MgO particles and thus the decreasing trend.

![Graph showing compression test results](image-url)
between the superconductor matrix and the added MgO particles before eventually fractured.

4. CONCLUSIONS

The improvement and enhancement of transport critical current density, $J_c$, in the Bi-2212 superconductor was observed in samples with small additions of nanosize MgO particles. The enhancement in $J_c$ may also be attributed to better texturing of the microstructure. With the employment of the partial-melt processing, small additions of 3% to 5% of nanosize MgO particles produced better texturing of the microstructure. The nanosize MgO particles that resided near the grain boundaries of the Bi-2212 phase lowered the grain orientation of the Bi-2212 phase, and subsequently improved the intergranular connectivity of the Bi-2212 phase superconductor. Furthermore, significantly higher stiffness, strength and toughness were recorded in the nanosize MgO added samples.

REFERENCES


