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Addition effects of nanoscale NiO on microstructure and superconducting properties of MgB_2

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Abstract

We have investigated the addition effect of NiO magnetic nanoparticles on crystal structure, microstructure as well as superconducting properties of MgB_2. NiO-added MgB_2 samples were prepared by the solid-state reaction method. The superconducting transition temperature (T_c) of 37.91 K was obtained for pure MgB_2, and T_c was found to decrease systematically on increasing the addition level of NiO. X-ray diffraction (XRD) analysis revealed that no substitution of Ni for Mg in the lattice of MgB_2 was occurred. The microstructural analysis shows that the pure MgB_2 sample consists of plate shape MgB_2 grains, and the grains get refined to smaller size with the addition of NiO nanoparticles. At 5 K, high values of critical current density (J_c) were obtained for small amount NiO-added MgB_2 samples as compared to pure sample. The enhancement in J_c could be attributed to the refinement of MgB_2 grains which leads to high density of grain boundaries in NiO-added MgB_2 samples.

Keywords : MgB_2, nanoscale NiO, superconducting properties

1. INTRODUCTION

The high critical current density (J_c) at high fields and high temperatures is the prime requirement for MgB_2 to replace the low temperature superconductors, such as NbTi and Nb_3Sn for cryogen-free practical applications [1-3]. Some of these applications include superconducting magnets for magnetic resonance imaging (MRI), solenoids, long power cables, and sensors for measuring liquid H_2 level [4, 5]. In these applications, pinning of magnetic flux lines (vortices) is an essential requirement to maintain high critical current at high fields for operation with low noise and lower power loss. Carbon doping is known to be very effective for enhancing the high-field critical current properties of MgB_2 [6]. However, the J_c enhancement achieved at high fields are usually associated with J_c degradation in low fields due to stronger current carrier scattering and current-blocking effects on grain boundaries [7]. At the same time, we have to compromise with the reduction of superconducting transition temperature (T_c) in carbon doping. Therefore, we selected nanoscale NiO magnetic particles (mean particle size < 50 nm) as a dopant for MgB_2 to introduce magnetic impurities. Magnetic impurities usually have a stronger interaction with magnetic flux line than nonmagnetic impurities and may exert a stronger force to trap the flux lines if they can be properly introduced into the superconducting matrix [8]. In this work, we study the influence of NiO magnetic nanoparticles on crystal structure, microstructure and superconducting properties of MgB_2.

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2. EXPERIMENTAL

NiO-added MgB_2 bulk samples were synthesized by the solid-state reaction method. The NiO with 0, 1, 3, and 5 wt. % of total MgB_2 was mixed with an appropriate amount of boron and magnesium powders under the environment of ultra-high pure Ar gas in glove box. The mixed powders were pelletized under 10 Ton pressure using mechanical hydraulic press. The pellets were put into Fe tubes and sintered *in situ* at temperature 800 ℃ for 30 min under the continuous flow of pure Ar gas.

The crystal structures of NiO-added MgB_2 bulk superconductors were investigated by X-ray diffraction (Rigaku, D/Max 2500) using Cu Kα as an X-ray source. In order to observe the microstructures, the samples were examined by scanning electron microscopy (SEM). The magnetization measurements were carried out on all samples by using a quantum design vibrating sample magnetometer option (Quantum Design PPMS). The superconducting transition temperature (T_c) was obtained from zero-field-cooled (ZFC) measurement by first cooling the sample in zero field and then measuring the magnetic moment as the sample was warmed up in the field of 10 Oe. To estimate the critical current density (J_c) of samples, the magnetization hysteresis ($M-H$) loops were measured on all samples with magnetic field varying from -9 to 9 T applied parallel to the longest dimension of the sample.

3. RESULTS AND DISCUSSION

The X-ray diffraction (XRD) patterns for pure and different amounts of NiO-added MgB_2 samples are shown in Fig. 1. The MgB_2 diffraction peaks are clearly observed along with MgO peaks. MgO is really hard to avoid during synthesis process as Mg is a highly reactive material. No shift in MgB_2 peaks were observed for NiO-added samples (within the experimental error), this indicates that there are no changes in the *a*- or *c*-axis lattice parameters due to NiO addion. The unchanged lattice parameters suggest that there is no substitution of Ni for Mg was occurred. However, for 5 wt. % NiO-added sample, MgNi compound was seen as a secondary phase in the XRD graph, which indicates Mg reacted with Ni upon increasing the level of NiO addion. No traces of free Ni or NiO particles were detected in the XRD patterns, this might be due to the small amount of NiO was used for addion. In order to clarify the presence of NiO nanoparticles in MgB_2 matrix, whether they are at grain boundaries or inside the grains, we are performing further experiments including microstructure observations and composition analysis by transmission electron microscopy (TEM) and energy dispersive spectroscopy (EDS), respectively.

Fig. 2 shows the temperature dependences of the magnetization for pure and different amounts of NiO-added MgB_2 samples. In ZFC curves all samples exhibit a sharp superconducting transition, which indicates that the samples are of good quality. The superconducting transition temperature (T_c) of 37.91 K was obtained for pure MgB_2. When we add NiO into MgB_2, a systematic decrease in T_c was found with increasing the addition level of NiO. For 1 wt. % NiO-added sample, T_c decreased by 1.15 K, and it suppressed by 2.97 K for 5 wt. % NiO-added sample. Since Ni was not substituted for Mg in the lattice of

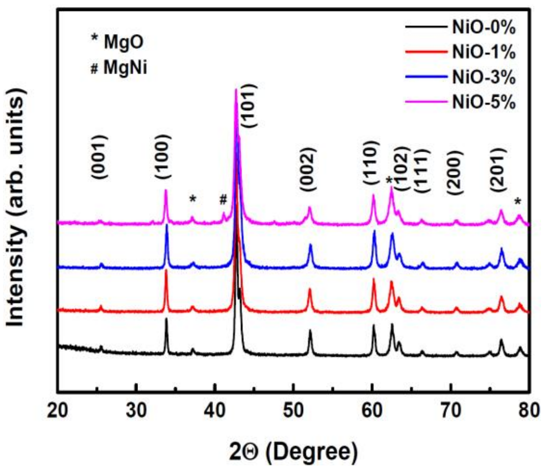

Fig. 1. X-ray diffraction patterns of pure and different amounts of NiO-added MgB_2 samples. No traces of free NiO particles were detected in doped samples.

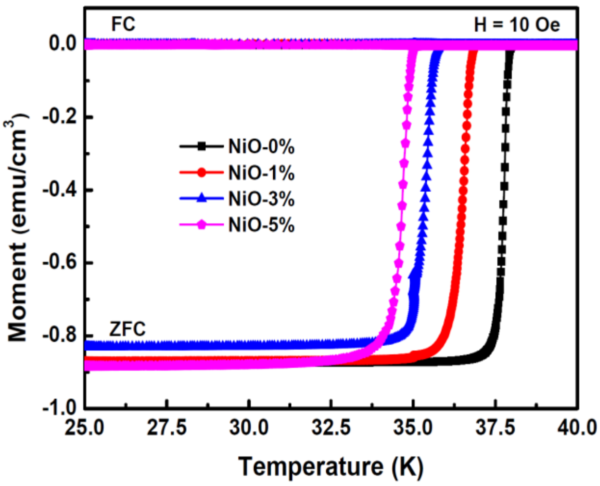

Fig. 2. Temperature dependence of magnetization measured in applied field of 10 Oe for pure and NiO-added MgB_2 samples. A systematic decrease in T_c was noticed with increasing the addition level of NiO.

MgB_2, therefore, the reduction in T_c because of pair breaking by magnetic NiO can be ruled out. On the other hand, the slow and systematic decrease of T_c with NiO addition indicates that the drop in T_c is most probably due to the change in stoichiometry of MgB_2 with varying the content of NiO and the reactions between Mg and Ni. The suppression of T_c due to stoichiometry change has also been reported for dopants addition and their reactions with MgB_2 [9].

The high magnification SEM images of (a) pure and different amounts of NiO (b) 1 wt. %, (c) 3 wt. %, and (d) 5 wt. % added MgB_2 samples are presented in Fig. 3. The pure MgB_2 sample consists of plate shape crystalline grains with an average width of 120 nm and average length up to 425 nm. From SEM morphologies of NiO added samples, Figs. 3b to 3d, it appears that the grains are refined to smaller size with NiO addition. For 3 wt. % NiO-added sample, the average width of grains decreased from 120 nm of pure sample to 80 nm and the average length reduced from 425 nm to 400 nm. The grains refinement by NiO doping is also supported by the decrease in the full width at half maximum (FWHM) values (not shown here) for all diffraction peaks of MgB_2.

To estimate the critical current density, the magnetization (M) as a function of applied field (H) at different temperatures was measured for all samples. As NiO is magnetic in nature, therefore, to check its magnetic effect, we measured the $M-H$ loop at 40 K along with 5 and 20 K on a pellet sample of 3 wt. % NiO-added MgB_2, and the data shown in Fig. 4. From $M-H$ loop which was measured in the normal state (40 K), we can see that the signal from magnetic nanoparticles NiO-added MgB_2 sample is very negligible. This is most likely due to the small amount of NiO was added for doping purpose.

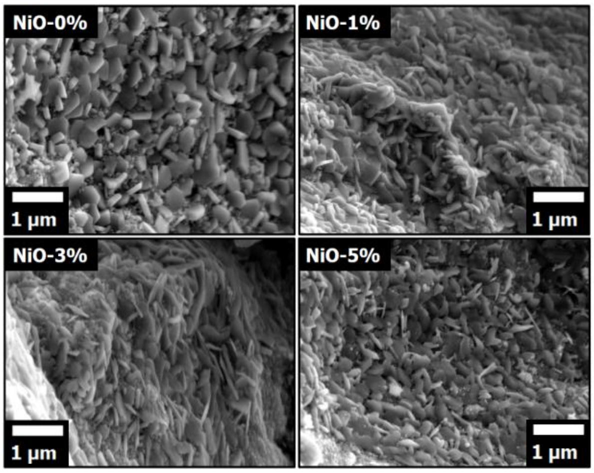

Fig. 3. The SEM morphologies of (a) pure and different amounts of nano-NiO (b) 1 wt. %, (c) 3 wt. %, and (d) 5 wt. % added MgB_2 samples. The grains get refined to smaller size with NiO addition.

Thus, we can deduce the critical current density for all samples directly from *M*−*H* loops.

The J_c was estimated from *M*–*H* loops by Bean's critical state model, $J_c = 20\Delta M/a(1\text{-}a/3b)$, where ΔM is the height of the *M*–*H* loop, *a* and *b* are the thickness and width of the sample, respectively. The magnetic field dependence of J_c at 5 and 20 K for pure and NiO-added MgB_2 samples are shown in Fig. 5a and 5b, respectively. At 5 K, the small amount 1 wt. % and 3 wt. % NiO-added samples show higher J_c both at low fields and up to 7 T as compared to pure sample. For further increasing the addition amount to 5 wt. %, the J_c is similar to pure sample upto 5 T, but after that it starts to fall. This indicates that only small addition of NiO is effective to pin the vortices and maintain high J_c. The high J_c could be attributed to the refinement of MgB_2 grains by NiO addition, which increases the area of grain

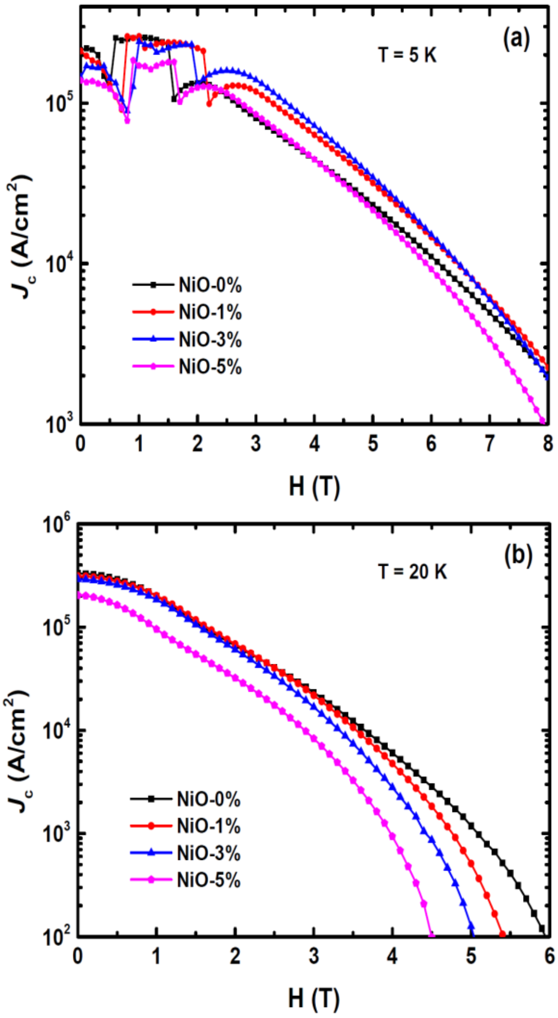

Fig. 5. The critical current density as a function of magnetic field for pure and NiO-added MgB_2 samples measured at (a) 5 K and (b) 20 K. High J_c values were obtained for small amount added NiO samples as compared to pure sample.

boundaries in MgB_2. At 20 K, the J_c of pure and NiO 1 wt. % and 3 wt. % added MgB_2 samples is almost overlapped with each other below 2 T field, after that the rate of decrease of J_c increased upon increasing the addition level of NiO. More degradation in J_c over the whole field was observed for 5 wt. % NiO-added sample. This indicates that at high temperature (20 K) and at high fields the pinning efficiency of NiO magnetic nanoparticles is reduced. The size and the distribution of NiO magnetic nanoparticles in MgB_2 matrix and their flux-pinning mechanism will be our future research work.

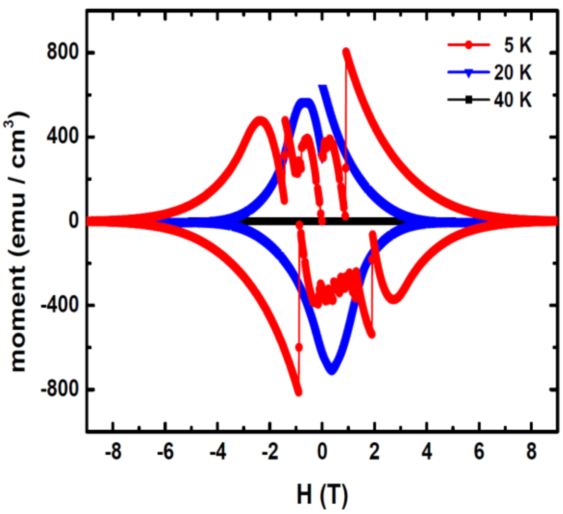

Fig. 4. The magnetization hysteresis loops measured at different temperatures for 3 wt. % NiO-added MgB_2 sample.

4. CONCLUSION

In summary, the influence of NiO magnetic nanoparticles on crystal structure, microstructures as well as superconducting properties of MgB_2 were investigated. The unchanged lattice parameters and no shift in the diffraction peaks of MgB_2 indicate that there no substitution of Ni for Mg was occurred. A systematic decrease in T_c was found upon increasing the level of NiO addition, which is most probably due to the change in stoichiometry of MgB_2. It is observed that only small amount of NiO magnetic nanoparticles (≤ 3 wt. %) are effective in enhancement of J_c, especially at low temperature of 5 K. The J_c enhancement could be attributed to the refinement of MgB_2 grains by NiO addition, which increases the area of grain boundaries in MgB_2, and the grain boundaries are known to be the dominant pinning source in MgB_2 superconductor.

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