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ARTICLE

ImprovedCritical Current Density and Magnetic Properties of Pr_6O_{11}-doped MgB_2 Bulks Synthesized by Mg-diffusion Method

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Abstract: The effects of the addition of Pr_6O_{11} nanopowder on the critical current density (J_c), irreversible field (H_{irr}) and upper critical field (H_{c2}) of the MgB_2 bulks prepared by a Mg-diffusion method were investigated. The results show that the Pr_6O_{11} nanopowder doping can significantly enhance the J_c, H_{irr} and H_{c2} of the MgB_2 bulks, but has little effect on the superconducting transition temperature (T_c). At 20 K in a self-field, the J_c value of the 1 wt% Pr_6O_{11}-doped MgB_2 sample is 3.61×10^5 A/cm^2, which is nearly 5 times larger than that of the undoped sample. In addition, at 10 K, the H_{c2} and H_{irr} of the bulk increase by 1.9 and 2.6 T, respectively. The effect of Pr_6O_{11}-doping on superconductivity and flux pinning mechanism was also discussed.

Key words: MgB_2; Pr_6O_{11} doping; critical current density; magnetic property

The discovery of superconductivity at 39 K in the MgB_2 compound has aroused a great interest in the field of applied superconductivity[1]. Extensive efforts have been made to improve the J_c, H_{irr} and H_{c2} of the materials, so that they can beappli ed to strong magnetic fields at 20 K. Experimental results have revealed that although grain boundaries are transparent to the current flow in MgB_2 [2,3], J_c drops rapidly as the strength of the magnetic field increases due to the lack of flux pinning centers and the poor connectivity between the material grains. Chemical doping has been considered as an effective and readily scalable technique for introducing pinning centers and improving J_c. Nanosized oxide inclusions including Al_2O_3, SiO_2, Y_2O_3, Dy_2O_3, Pr_6O_{11}, and TiO_2, have been reported to enhance the flux pinning in MgB_2 [4-8]. It has been found that nanosized coherent oxides can act as effective pin ning centers in MgB_2 materials[9,10], but large grains or high quantity of oxide evidently impair the grain connectivity.

MgB_2 bulks synthesized from a powder mixture of magnesium and boron under ambient pressure are usually porous with only 50% of theoretical density. Therefore, it is evident that the poor self-sintering nature of MgB_2 grains results in a large amount of holes in the material, leading to low density. There are several methods to produce highly dense MgB_2 samples without using any high pressure techniques, such as diffusion of magnesium into boron fibers[11], infiltration of liquid magnesium into powder boron[12] and the powder-in-closed-tube (PICT)-diffusion method[13,14]. The PICT-diffusion method has several advantages, including easy procedures and elimination of the MgO contamination. Ueda et al[11] has reported that a high critical current density (~0.86 MA/cm^2 at 20 K in self-field) can be achieved by the diffusion method, but it is found that the J_c value decreases as the strength of the resident magnetic field increases, and the J_c drops faster than that in the materials prepared by the solid-state reaction route. Therefore, we attempted to improve the J_c and magnetic properties of the MgB_2 bulks through a combination of nanopowder doping and the Mg-diffusion method.

1 Experiment

A series of Pr_6O_{11}-doped MgB_2 samples with a mass ratio of

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MgB_2:Pr_6O_{11}=1–x:x (x=0, 0.005, 0.01, 0.03) were prepared using the Mg-diffusion method under ambient pressure. The powders consisting of B (amorphous, purity 99.9%), Mg (purity 99%) and Pr_6O_{11} (purity 99.9%, particle size 100 nm), were mixed and pressed into cylinders with a diameter of 6 mm under a pressure of 4 MPa. The samples were heated at 850 °C for 24 h in a flowing Ar atmosphere and then cooled to room temperature. All of the samples had an average density of 1.77 g/cm^3, which was 68% of theoretical density. The magnetic properties of the samples were measured in a temperature range of 5~45 K, using a magnetic properties measurement system (MPMS, Quantum Design) and the magnetization measurements were performed using a 4 kA/m applied field in zero-field-cooling (ZFC). The magnetic J_c was calculated from the width ΔM of the magnetization loop (M-H) using the extended Bean model: J_c=20ΔM/[a(1–a/3b)], where a and b are the dimensions of the sample perpendicular to the direction of the applied magnetic field and a<b. The H_{irr} values of the samples were determined from the closure of hysteresis loops and the criterion is J_c=100 A·cm^{-2}.

2 Results and Discussion

Fig.1 shows the XRD patterns of the samples MgB_2:Pr_6O_{11} =1–x:x (x=0, 0.01, 0.03). MgB_2 is the main phase in all samples. The undoped sample is composed of MgB_2 phase and a little amount of MgO. In the Pr_6O_{11}-doped samples, PrB_6 besides MgO can be identified as the impurities. With the increase of the Pr_6O_{11} doping level, the intensity of the PrB_6 peak increases. The a- and c-axis parameters calculated from the XRD patterns are also shown. Compared with the undoped samples, the lattice parameters of a and c in the Pr_6O_{11}-doped samples increase. The Pr ion radius is larger than the Mg ion radius, so the values of the lattice parameters become large. It is suggested that Pr substitution for Mg occurs in MgB_2 lattice.

Fig.2 shows the results of the temperature dependence of zero field cooled (ZFC) magnetization M(T) measured at 4 kA/m for the Pr_6O_{11}-doped samples. All samples exhibit a

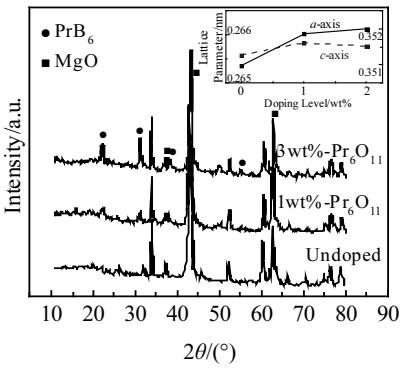

Fig.1 XRD patterns of the Pr_6O_{11}-doped MgB_2 samples after heating at 850 °C for 24 h (Inset: a, c-axis lattice parameters of MgB_2)

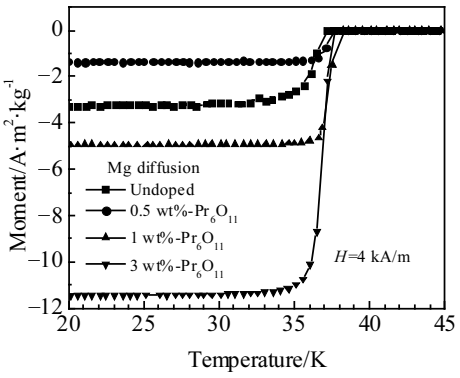

Fig.2 Temperature dependence of the magnetization measured at 4 kA/m

sharp transition. As can be seen, the highest transition temperature of the 1 wt% Pr_6O_{11}-doped sample is 38.3 K, while the lowest transition temperature of the undoped sample is 36.3 K and others are 37.6 K. Pan et al[6] reported that T_c is barely inhibited in the sample doped with nano-Pr_6O_{11} particles, and the pair-breaking effect of the magnetic moment of the rare-earth element is not significant in MgB_2. Therefore, in this work, the T_c increase in the Pr_6O_{11} doped samples may be attributed to the diversity of the quality in various samples, rather than the effect of the Pr_6O_{11} doping. On the other hand, the diamagnetic signal varies significantly with the doping levels. When the low doping levels x≤0.5wt%, the diamagnetic signal is small which may be attributed to low density, although the phase of the samples is relatively pure. With increasing doping level, the Pr_6O_{11} doping helps to increase the nucleation rate of MgB_2, thereby increasing the density and quantity of MgB_2. When the doping level x≥1wt%, the samples have a higher density, and although the MgB_2 phase has more impurities, the diamagnetic signals are enhanced at the same time.

Fig.3 presents the variation of the J_c for the samples with magnetic field strength at 10 and 20 K. The J_c values of the MgB_2 bulks prepared by the Mg-diffusion method are obviously improved when the materials is doped with Pr_6O_{11} nanoparticles, especially in a stronger field. The J_c value of the undoped sample and the 1 wt% Pr_6O_{11}-doped sample is 6.0×10^4 A/cm^2 and 3.12×10^5 A/cm^2 at 10 K in 1 T, respectively. At 20 K in a self-field, the J_c value of 1 wt% Pr_6O_{11}-doped is 3.61×10^5 A/cm^2, whereas this value is only 7.23×10^4A/cm^2 for the undoped bulk, which is an increase of a factor of 5. Furthermore, Fig.3a also shows the J_c-μ_0H curves of the undoped and 1 wt% Pr_6O_{11}-doped MgB_2 bulks prepared by a solid-state reaction route. As can be seen, the J_c of the samples prepared by the Mg-diffusion method in the low filed exhibits much better performance than that of the samples prepared using the solid state method[15]. Therefore, the combination of Mg-diffusion and the Pr_6O_{11} nanoparticles doping method may

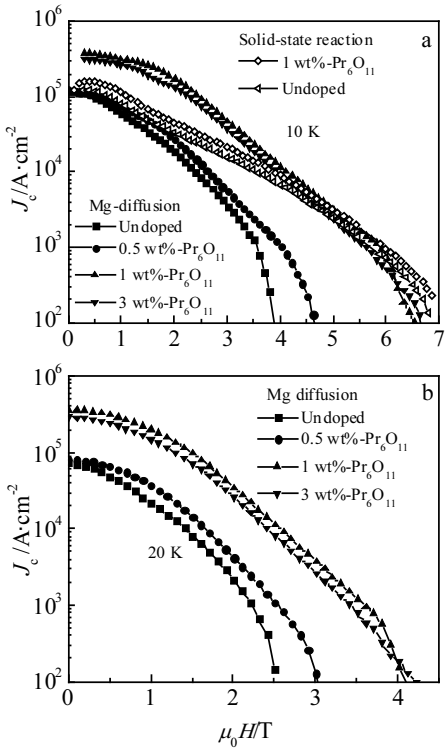

Fig.3 Magnetic field dependence of the critical density J_c properties at 10 K (a) and at 20 K (b)

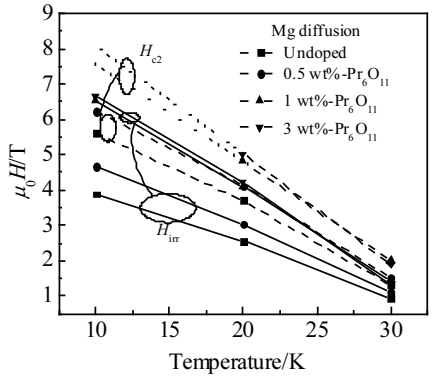

Fig.4 Temperature dependence of the irreversibility field H_{irr} and upper critical field H_{c2}

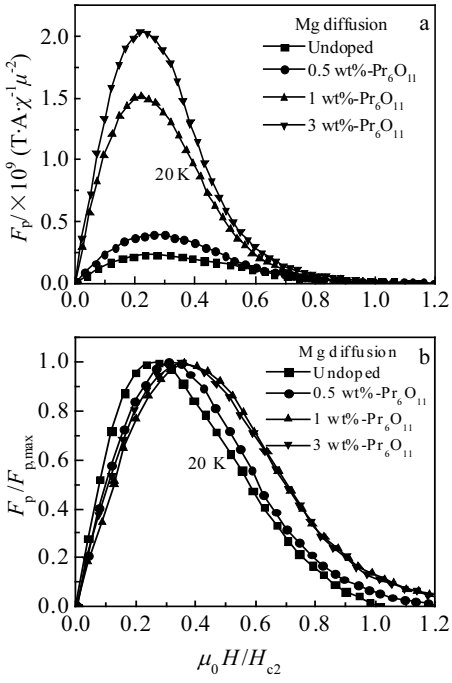

Fig.5 Flux pinning force as a function of applied magnetic field at 20 K: (a) flux pinning force F_p and (b) normalized flux pinning force $F_p/F_{p,max}$

be an underlying route for improving the superconducting performance of MgB_2.

Fig.4 shows the temperature dependence of H_{c2} and H_{irr} obtained from the magnetic hysteresis measurements. The parameter H_{irr} is determined from the closure of hysteresis loops with a criterion of J_c=100 A·cm^{-2}. The H_{c2} is determined from the point at which the magnetization begins to deviate from the normal linear background. A systematic increase in the H_{c2} and H_{irr} is clearly observed for the Pr_6O_{11} doped samples. For the 1 wt% Pr_6O_{11}-doped sample, the H_{irr} and the H_{c2} are 6.5 T and 7.5 T at 10 K, respectively, which is increased by 2.6 T and 1.9 T compared to those of the undoped sample (3.9 T and 5.6 T), respectively. The H_{irr} and H_{c2} values increase with the increase of Pr_6O_{11} content. When the maximum quantity of Pr_6O_{11} is 3 wt%, the maximum values of H_{irr} and H_{c2} reach 6.7 and 8.1 T, respectively.

Fig.5 shows the pinning force as a function of applied magnetic field. In Fig.5a, a remarkable and systematic improvement in the pinning force can be seen in the Pr_6O_{11}-doped samples. The maximum pinning force F_p per volume unit for the 1 wt% Pr_6O_{11}-doped sample is 4 times larger than that in the undoped sample. Fig.5b shows the reduced field dependence of the normalized flux pinning force ($F_p/F_{p,max}$). The peaks in the Pr_6O_{11}-doped samples shift to higher field strengths and are much broader than those in the undoped sample, mainly denoted in the higher field, indicating that the pinning strength is improved in the higher field region. Therefore, the improvement of the MgB_2 properties can be explained by the doping of Pr_6O_{11}, which appears to increase the flux pinning force. Pan et al[6] has reported that Pr_6O_{11} nanoparticles in the MgB_2 samples are decomposed to form PrB_6 as revealed by XRD analysis. A partial substitution of Pr for Mg in MgB_2 crystal structure causes improved flux pinning behavior.

As well known, the behavior of J_c in MgB_2 bulks is influenced by both the flux pinning force and sample density. In this work, a series of highly compact MgB_2 samples with an average density of 1.77 g/cm^3 were prepared by the

Mg-diffusion method, while the usual density of MgB_2 bulks prepared by the standard solid-state reaction route at ambient pressure is 1.2 g/cm^3. A small amount of holes in the MgB_2 bulks prepared by the Mg-diffusion method caused an increase in the effective cross section for superconducting current, so the improved density is favorable for enhancing the J_c in MgB_2 bulks.

3 Conclusions

1) A series of Pr_6O_{11}-doped MgB_2 samples with various doping levels can be prepared using the Mg-diffusion method at ambient pressure, which have an averagede nsity of 1.77 g/cm^3.

2) The J_c, H_{irr} and H_{c2} in the Pr_6O_{11}-doped MgB_2 bulks increase significantly, but the T_c is not affected. The J_c reaches a maximum value in the 1 wt% Pr_6O_{11}-doped sample, and the 3 wt% Pr_6O_{11}-doped MgB_2 sample exhibits the maximum H_{irr} and H_{c2} values.

3) The combination of Pr_6O_{11} nanoparticles doping and the Mg-diffusion method can be an underlying pathway for improving the MgB_2 superconducting performance.

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镁扩散法制备 Pr_6O_{11} 掺杂的 MgB_2 块体的临界电流密度和磁场性能

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摘 要：采用镁扩散法制备了 Pr_6O_{11} 纳米颗粒掺杂的 MgB_2 超导块体，研究了 Pr_6O_{11} 掺杂对其临界电流密度(J_c)，不可逆磁场(H_{irr})和上临界磁场(H_{c2})的影响。实验结果表明，Pr_6O_{11} 纳米颗粒掺杂明显提高了块体的 J_c，H_{irr} 和 H_{c2}，但没有降低其超导转变温度 T_c。在 20 K 自场条件下，质量分数为 1% Pr_6O_{11} 掺杂 MgB_2 块体的 J_c 为未掺杂样品的近 5 倍，J_c=3.61×10^{5}A/cm^2。在 10 K 温度下，MgB_2 块体 H_{c2} 和 H_{irr} 较未掺杂样品分别提高了 1.9 和 2.6 T。同时讨论了 Pr_6O_{11} 纳米颗粒掺杂对 MgB_2 块体的超导性能和磁通钉扎机制的影响。

关键词：MgB_2；Pr_6O_{11} 掺杂；临界电流密度；磁场性能

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