Effects of Re Addition into MgB_2 SuperconductorM. Takeda¹, M. Migita¹, Y. Nozaki¹, M. Uehara¹, T. Kuramoto¹, D. Hamane²,
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Re was added into the MgB_2 superconductor. The Mg, B and Re were sintered at 1173K for 30 minutes under H_2/Ar atmosphere in the electric furnace. The nominal $\text{Re}_x(\text{MgB}_2)_{1-x}$ samples were prepared in $0 \leq x \leq 0.05$. The field and x dependences of the critical current density J_c were analyzed by the extended critical state model. The enhancement of J_c was observed for the $x = 0.02$ sample. The effective pinning center should be ReB_2 nano-particles on the MgB_2 grain boundaries.

Key words: superconductor, MgB_2 , Re, magnetization, critical current density

1. INTRODUCTION

In earlier studies, the enhancement of critical current density J_c , at 20 K and 1 T was observed in X/MgB_2 ($\text{X}=\text{Ag-Mg}$, MgCu_2 , NbB_2 , TiB_2) sintered bulk samples, including a few mol % of X [1-5]. They were prepared by sintering the mixture of Mg, B and the oxides such as Ag_2O , CuO , Nb_2O_5 , and TiO_2 in the Ar/H_2 atmosphere at 1173 K for 30 minutes. Then the secondary phase of X was produced in them as a pinning center for the penetrated magnetic flux. The efficiency of small addition of metallic elements was also confirmed for sintered MgB_2 under high pressure of 3.5 GPa [5], and SiC/MgB_2 system [6]. Recently, it was found that the addition of 4d and 5d transition elements of Mo and W also enhanced the pinning property of MgB_2 [7,8].

As for the 5d transition metal of Re, the superconductivity was observed below T_c of 2.42 K [9]. Rhenium borides of ReB_2 and Re_3B were also superconductors with T_c of 4.5-6.3 K and 4.7 K, respectively [10]. Therefore Re or Re-borides are expected to be pinning centers at 20 K at which they are normal conductors. Especially, the ReB_2 has a possibility of mechanically hard pinning centers, since it has been known as an ultra-incompressible super-hard material [11-13].

In the present report, we show the superconducting transition temperature T_c and J_c results of the $\text{ReB}_2/\text{MgB}_2$ system, since the efficiency of ReB_2 has not been confirmed up to the present, especially at 20 K.

2. SAMPLE PREPARATION

The Re added MgB_2 bulk system was prepared by rapid sintering method. The powder mixtures of $x\text{Re}+1.6(1-x)\text{Mg}+2\text{B}$ were sintered at 1173 K for 30 minutes in the 10% H_2/Ar atmosphere, where x is the atomic concentration of Re in the

range of x between 0 and 0.05. The excess amount of 0.6Mg was expected to be evaporated during the above sintering process.

In Fig. 1, the $\text{CuK}\alpha$ powder X-ray diffraction (XRD) patterns are depicted for the sample of $x = 0, 0.02$ and 0.04 samples. The diffraction peaks of MgB_2 and ReB_2 were observed. They have commonly hexagonal crystal structure with the lattice parameters of $a = 0.308$ nm and $c = 0.353$ nm for the MgB_2 , and $a = 0.290$ nm and $c = 0.743$ nm for the ReB_2 . These results coincide with the values in the earlier literature [14,15], and there was no systematic change in the a and c by the Re amount. So we thought that the Re was scarcely substituted on the Mg- or B-sites, and the ReB_2 secondary phase was nearly isolated in the present system.

Fig.1 XRD patterns of $x = 0, 0.02$ and 0.05 samples.

The mean crystallographic correlation length or particle diameter d of ReB_2 phase was estimated from the half widths of XRD peaks by Sherrer's formula. We think that d is the averaged diameter of ReB_2 particles. The x -dependence of d is shown in Fig. 2. The d of ReB_2 was 14~16 nm which is comparable to the diameter of quantum vortex core of $2\xi \sim 10$ nm, where ξ is the coherent length of MgB_2 [16].

Fig.2 x -dependence of d of ReB_2 .

3. RESULTS AND DISCUSSION

The T_c 's were obtained from the temperature dependence of resistivity ρ by 4-terminals method. In Fig. 3, $\rho(T)$ curves of $x = 0, 0.02$ and 0.05 samples are shown. The x -dependences of T_c 's at onset, middle and end point of superconducting transition are shown in Fig. 4. They show no visible variation in $0 \leq x \leq 0.05$.

Fig. 3 Temperature dependence of resistivity ρ of $x = 0, 0.02$ and 0.04 samples.

These results mean that the good pinning centers of ReB_2 could be expected in the MgB_2

system, because the superconducting property was not deteriorated by Re-doping.

Magnetization measurements were performed by the Quantum Design's MPMS SQUID magnetometer. The ZFC magnetization curves of $x = 0, 0.02, 0.04$ and 0.05 samples at 20 K are shown in Fig.5. The expressions for magnetization M by extended critical state model were used to estimate the critical current density J_c of each sample. The J_c was defined as $J_c(H) = B_{eq}^* (B_{eq}^* + 2B_0) / [2\mu_0 a \{ |B_{eq}(H)| + B_0 \}]$ [17-19], where μ_0 of $4\pi \times 10^{-7}$ [Tesla·m/A] is the vacuum permeability, and $2a$ is the thickness of slab sample.

Fig. 4 x -dependence of T_c 's at onset, middle and end point of $\text{Re}_x(\text{MgB}_2)_{1-x}$ system.

Fig. 5 ZFC magnetizations of $x = 0, 0.02$ and 0.04 samples of $\text{Re}_x(\text{MgB}_2)_{1-x}$ system at 20 K.

In the above expression of $J_c(H)$, $B_{eq}(H)$ of $\mu_0[H + M_{eq}(H)]$ is the flux density at the sample surface, where M_{eq} is the equilibrium magnetization. The B_{eq}^* represents $B_{eq}(H_p)$, where H_p is

the *full penetration field*, above which the internal flux density B_{int} becomes non-zero at the center of the slab. The B_0 can be called the *cross-over flux density* between Bean [20] and Anderson-Kim [21] states. Analyses of ZFC $M(H)$ -curves were performed using the equations given in earlier papers [17-19]. The parameters of B_{eq}^* and B_0 were evaluated from the least square fitting of experimental $M(H)$ -data to theoretical equations by Gauss-Jordan method.

Fig. 6 Field dependences of J_c 's of $\text{Re}_x(\text{MgB}_2)_{1-x}$ system at 20 K.

Fig. 7 $J_c(x)/J_c(0)$ curves of $\text{Re}_x(\text{MgB}_2)_{1-x}$ system at 20 K.

The field dependences of J_c 's and the x -dependence of $J_c(x)/J_c(0)$ were shown in Figs. 6 and 7 where $J_c(0)$ is the J_c of $x = 0$ sample. As shown in these figures, the enhancement of J_c was observed at $x=0.02$ where $J_c(x)/J_c(0)$ is about 1.3. Thus the efficiency of isolated ReB_2 phase is examined for the MgB_2 system. The authors emphasize that the size and density of ReB_2 pinning center may be optimum at $x = 0.02$.

Fig. 8 $F_p/F_{p\text{max}}$ vs. H/H_{max} curves at 20 K.

In Fig.8, normalized pinning force densities divided by their maximum values, $F_p/F_{p\text{max}}$, are shown for H/H_{max} , where $F_p = \mu_0 J_c H$ and H_{max} is the magnetic field at which the F_p has the maximum value of $F_{p\text{max}}$. The J_c above $H=1\text{T}$ was obtained from Bean's model [22]. Three kinds of lines in Fig.8 are the theoretical curves of $F_p/F_{p\text{max}}$ expressed by Shigeta et al. [23]. All of the experimental data between $x = 0$ and 0.05 are well fitted by the *surface pinning* curve. It shows that the main pinning centers are formed at the boundaries among the MgB_2 -grains and the ReB_2 nanoparticles in the grain boundaries strengthen the pinning force in MgB_2 . The SEM image of $x = 0.02$ sample in Fig. 9 shows that the white parts of ReB_2 phase exist mainly at the grain boundary between the MgB_2 phase, which indirectly supports the above fitting result.

Fig. 9 SEM image of $x = 0.02$ sample.

4. CONCLUSION

J_c enhancement in the nominal $\text{Re}_x(\text{MgB}_2)_{1-x}$ system was observed at $x=0.02$ where $J_c(x)/J_c(0)$ is about 1.3. The present system was prepared from the mixture of Mg, B and Re in the H_2/Ar atmosphere by rapid sintering at 1173 K for 30 minutes, followed by the cooling in a furnace. The secondary phase of ReB_2 is expected to

become good pinning centers on the MgB_2 grain boundaries for the penetrating magnetic flux.

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