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CHARACTERIZATION OF THE HEAT TREATMENT MgB_2 RODS OBTAINED BY PIT TECHNIQUE WITH EXPLO9SIVE CONSOLIDATION METHOD

CHARAKTERYSTYKA WYGRZEWANYCH PRĘTÓW MgB_2 OTRZYMANYCH TECHNIKĄ PIT Z KONSOLIDACJĄ METODĄ WYBUCHOWA

$Cu/Fe/MgB_2$ rods have been prepared by the modified *PIT* technique with explosive consolidation method. In the next step, so obtained rods were additionally annealed. The heat treatments of the rods were performed at 750°C and 850°C for 1 hour in He atmosphere. The phase compositions, microstructure features were investigated by X-ray diffraction and scanning electron microscopy (SEM) techniques, while the values of lattice parameters were determined by Rietveld method. The contents of all involved phases were determined by Rietveld procedure. The superconducting properties for a core MgB_2 rods were also investigated by the AC susceptometry and SQUID magnetometry techniques. It was found that no impurity phases were formed in heat treatment processes of $Cu/Fe/MgB_2$ rods. In addition, it was found that MgB_2 powder was well consolidated. There were found some small voids in the heat treated rods. The high critical temperature T_c of 38.7 K and high critical current density $J_c \sim 10^5$ A/cm^2 were obtained in both rods.

Keywords: MgB_2, *PIT* technique with explosive consolidation method, XRD, SEM, Critical temperature, Critical current density

W pracy omówiono proces otrzymywania wygrzewanych prętów nadprzewodzącego MgB_2. W pierwszym etapie otrzymano pręty $Cu/Fe/MgB_2$ zmodyfikowaną techniką *PIT* z konsolidacją metod wybuchową, które w kolejnym etapie poddano obróbce cieplnej. Proces wygrzewania prętów odbywał się w temperaturze 750°C oraz 850°C w czasie godziny, w atmosferze ochronnej helu. Otrzymane pręty poddawano badaniom metalograficznym i rentgenowskim z wykorzystaniem skaningowego mikroskopu elektronowego (SEM) oraz dyfraktometru rentgenowskiego firmy Philips model 1130/00. W oparciu o uzyskane dyfraktogramy rentgenowskie określono ilościowy udział faz wchodzących w skład badanego materiału oraz stosując metodę Rietvelda określono wartości parametru sieci a_0 i c_0.

Badania magnetyczne *AC* podatności χ wykonano przy użyciu susceptometru *AC*, z kolei pomiary pętli histerezy namagnesowania przeprowadzono przy użyciu magnetometru SQUID. Przeprowadzona analiza fazowa prętów $Cu/Fe/MgB_2$ wygrzewanych w temperaturze 750°C i 850°C nie ujawniła występowania obcych faz. Badania mikrostruktury ujawniły w wygrzewanych prętach MgB_2 występowanie małej ilości pustek o małych rozmiarach. Pomiary magnetyczne ujawniły wysoką temperaturę krytyczną T_c = 38.7 K i wysoką krytyczną gęstość prądu $J_c \sim 10^5$ A/cm^2 (4.1 K, 0 T)

1. Introduction

The recently discovered superconductor MgB_2 with $T_c \approx 39$ K [1] has promoted many studies for practical applications. Several methods have been used to obtain bulk MgB_2 samples but from the application point of view the *Powder-In-Tube* (PIT) technique is the most promising method to obtain high quality MgB_2 wires and tapes [2]. There are two main processes of *PIT* MgB_2 fabrication: *ex-situ* and *in-situ* process. In the first step the initial powder (MgB_2 or Mg+2B) is packed into metallic tube. After the packing process tube with MgB_2 is drawn into wires or rolled into tapes. In some cases so obtained wires or tapes are annealed. In the last time development works are focused to obtain well consolidated with good properties MgB_2 samples. Several research groups reported MgB_2 fabrication by using the mentioned technique. Grasso et al. [3] successfully fabricated tapes with a *PIT* method without any heat treatment. Sumption et al. [4] obtained MgB_2 tapes with $J_c = 7.5 \times 10^4$ A/cm^2 after a rapid sintering process.

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In our paper we report the fabrication of the heat treated Cu/Fe/MgB_2 rods obtained by the modified *PIT* technique with explosive consolidation method. We used two different annealing temperatures to compare the effect of the heat treatment on the phase formations, microstructure and superconducting properties.

2. Experimental

As we previously described [5, 6] the modified *PIT* technique with explosive consolidation method was used to prepare the monocore rods. The commercially available MgB_2 powder (Alfa Aesar) was tightly packed into Fe (OD: 4.85 mm, ID: 4 mm, length: 160 mm) tubes. This packing process was carried out by hand in air. Next, the tubes with initial MgB_2 powder were closed by Fe plugs. The filled and closed tubes were placed in additional Cu tubes (OD: 8.3 mm, ID: 4.9 mm). The so-obtained double arrangements of tubes with initial powders (Cu/Fe/MgB_2) in length equal 185 mm, were closed with Cu plugs. In this way obtained arrangements of tubes were consolidated using an explosive method. The consolidation set-up and the explosive materials applied were described elsewhere [7].

After explosive consolidation method we obtained superconducting Cu/Fe/MgB_2 rods. These rods were cut into samples of 5 mm in length. After that, samples were annealed at two different temperatures. The heat treatments were performed in 750±1°C and 850±1°C for 1 hour in He atmosphere and followed by furnace cooling to room temperature. After those processes, we obtained two heat treated Cu/Fe/MgB_2 rods at 750°C and 850°C (hereafter referred to as S1 and S2 samples, respectively).

In our experiments we applied two different heat treatment conditions in order to increase the density of so obtained rods and to compare the phase composition, structure and superconducting properties such MgB_2 rods.

The phase composition and microstructure of S1 and S2 samples were examined by XRD and SEM methods. XRD characterizations were carried out with Philips (1130/00) diffractometer using CuK_α radiation (λ = 1.54178 Å) after mechanically peeling off the sheath material and grinding the core into powder. The phase composition of each MgB_2 rods was determined on the basis of the X-ray diffraction patterns taken in the angle range 2θ = 20-140° with step 0.05° and time measurement t_p = 4 s. Metallographic observations were carried out with the use of scanning electron microscope JOEL JSM-6480.

The analysis of XRD patterns was performed by the use of Rietveld method [8]. Rietveld method appeared to be useful in the microstructure characterization and also in the verification of the qualitative phase composition of multiphase materials [9]. The detailed information on the structure of concerned phases is necessary for the estimation of phase abundance in multiphase materials [10]. The procedure of quantitative phase analysis based on the values of scale factors determined by Rietveld refinement was introduced by Hill and Howard [11].

Magnetic measurements were performed on the core samples. Cu/Fe sheaths were removed by peeling. The *AC* susceptibility was measured in the temperature range 2-50 K by means of the *AC* susceptometry technique. Magnetization hysteresis loops (M-H hysteresis loops) for heat treated MgB_2 rods were carried out using a SQUID magnetometer in the temperature 4.1 K and in applied fields up to 5.0 Tesla. The magnetic critical current density values (J_c) were calculated from hysteresis loops obtained for magnetic field (H), using the Bean's critical state model [12, 13] given by $J_c = 30 \times \Delta M/d$, where ΔM is the hysteresis of magnetization per volume (emu/cm^3) at a given field and d is the mean size of the particles [14]. Assuming an average particle size of 2.5 $\times 10^{-2}$ cm for both samples, the critical current density J_c obtained at various fields are plotted in Fig. 5.

3. Results and discussion

Figure 1 shows the X-ray diffraction patterns of S1 (Fig. 1a) and S2 (Fig. 1b) samples. It may be noted that in both cases the main phase is MgB_2 with only a few impurity phases of SiO_2 and MgO. The XRD analysis indicates that there is almost no reaction between MgB_2 and the inner Fe tubes during the all preparation process. This result indicates that the Fe tube is good material for preparing heat treated rods after explosive consolidation method.

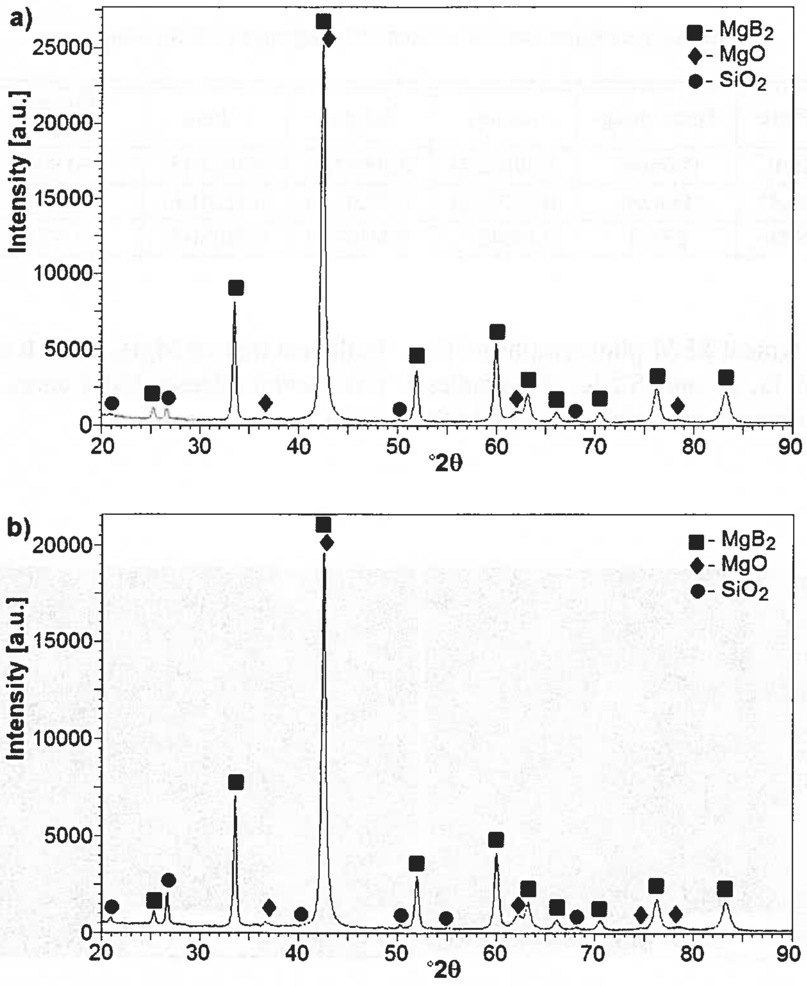

Fig. 1. X-ray diffraction patterns of (a) S1 and (b) S2 samplese

The values of lattice parameters determined by Rietveld method and the contents of all involved phases determined by Rietveld procedure are given in Tables 1 and 2. The accuracy of the lattice parameter determination (equal to ±0.015%) was estimated using SRM 1976 alumina plate as a standard. The heat treatment processes reveals that S1 samples annealed at 850°C has higher lattice parameter than S2 samples annealed at 750°C. The changes of lattice parameters were observed for MgB_2 and SiO_2 phases (Table 1 and 2). Moreover these parameter of MgO were stable. The reason for these changes was initial powder relaxation after explosive consolidation method. The increase contents of MgO phase in S2 sample was the result of the higher heat treatment process and presence of pure Mg in initial powder [7]. The numerical phases analysis reveal loss of MgB_2 phase contents (~3 wt.%).

TABLE 1

Lattice parameters and the contents of components for S1 sample

Phase	Space group	a_0[nm]	b_0[nm]	c_0[nm]	Contents [wt.%]
MgB_2	P6/mmm	0.30837(4)	0.30837(4)	0.35218(5)	94.6(6)
MgO	Fm-3m	0.42209(6)	0.42209(6)	0.42209(6)	5.1(5)
SiO_2	P3121	0.49090(7)	0.49090(7)	0.54066(8)	0.3(1)

TABLE 2

Lattice parameters and the contents of components for S2 sample

Phase	Space group	a_0[nm]	b_0[nm]	c_0[nm]	Contents [wt.%]
MgB_2	P6/mmm	0.30842(4)	0.30842(4)	0.35221(5)	90.9(9)
MgO	Fm-3m	0.42201(6)	0.42201(6)	0.42201(6)	7.5(3)
SiO_2	P3121	0.49142(7)	0.49142(7)	0.54051(8)	1.6(2)

Figure 2 shows the typical SEM photographs of the cross-sections of the S1 (a, b) and S2 (c, d) samples. The boundaries between barrier materials are visible for both heat treated MgB_2 rods. It can be seen that the both rods contain dense (bulk) cores with a small amount of voids.

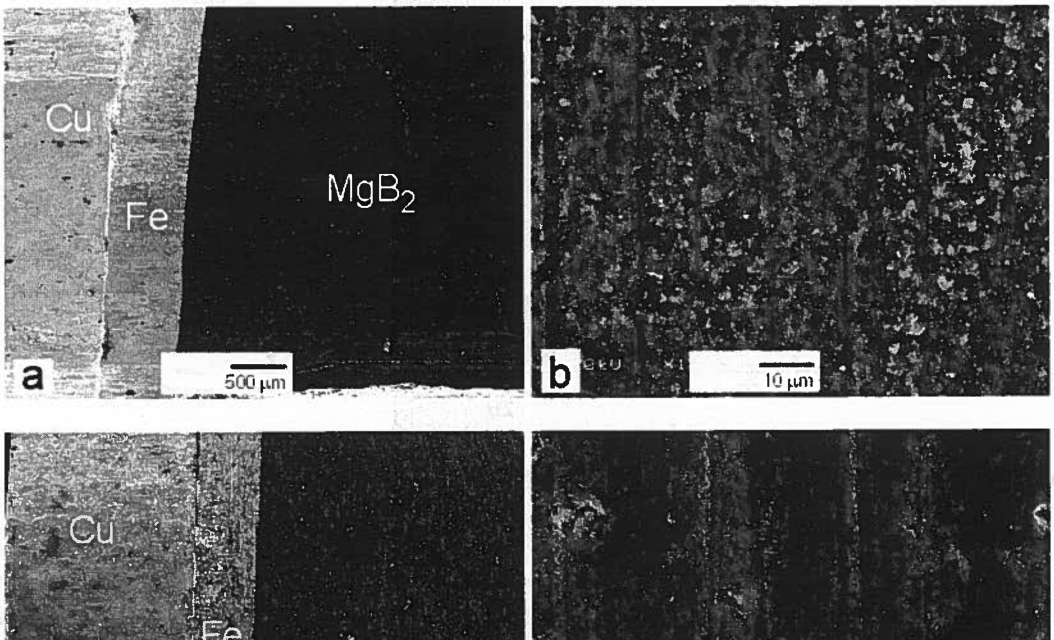

Fig. 2. SEM images of the MgB_2 parts in Cu/Fe tubes: (a) – annealed at 750°C, (c) – annealed at 850°C and SEM images of the MgB_2 cores: (b) – annealed at 750°C, (d) – annealed at 850°C

The temperature dependence of *AC* susceptibility for S1 and S2 samples is shown in Figs. 3a and 3b. The real part of *AC* susceptibility χ' (Fig. 3a) and the imaginary part of *AC* susceptibility χ" (Fig. 3b) reveal that sample S1 has one-step transition temperature, e.g. intragranular temperature (T_c). The real part of *AC* susceptibility χ' (Fig. 3a) and the imaginary part of AC susceptibility χ" (Fig. 3b) reveal that sample S2 has two-step transition temperature, e.g. intragranular temperature (T_c) and intergranular temperature (T_{int}). As seen within the imaginary part of susceptibility curves, there are the narrow maxima in both transition temperatures. The presented data show that the characteristic intragranular transition temperature for S1 and S2 samples are T_c = 38.7 K and intergranular temperature for S2 sample is T_{int} = 36.7 K.

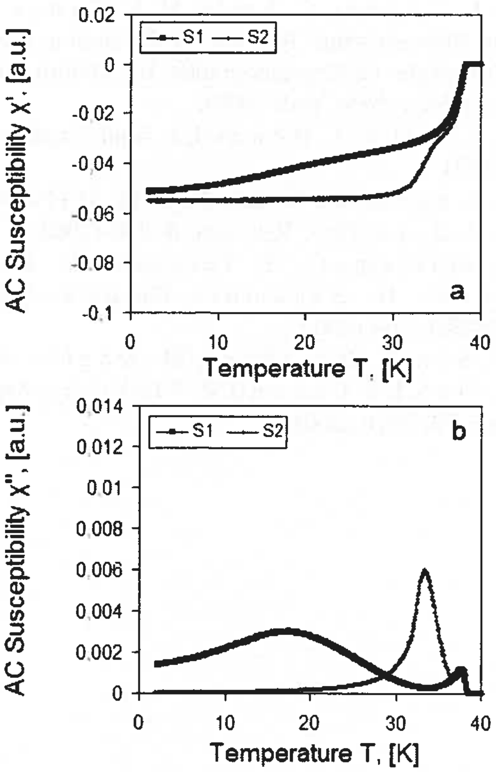

Fig. 3. The real (a) and the imaginary (b) parts of *AC* susceptibility χ' and χ'' for S1 and S2 samples

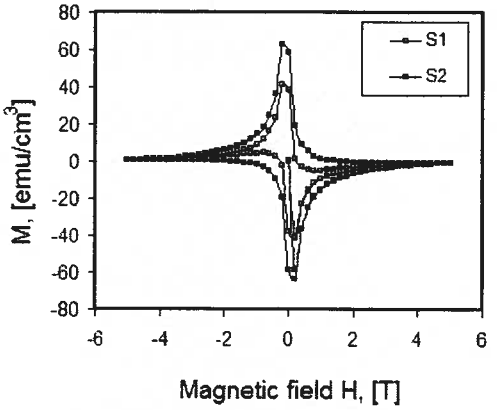

Fig. 4. Magnetic hysteresis loops measured at 4.1 K for samples S1 and S2

Figure 4 illustrates *M-H* hysteresis loops for S1 and S2 samples. The dependence of J_c on H, calculated by using the data of Fig. 4 is shown in Fig. 5. It can be seen that the obtained values of J_c at 4.1 K and 0 T field for sample S1 and sample S2 are 9.16×10^4 and 1.4×10^5 A/cm^2, respectively. This relation shows that the J_c monotonously decreases and for magnetic field equal of 5.0 T critical current density $J_c = 0$. These critical current density values are comparable with critical current density $J_c = 2.3 \times 10^5$ A/cm^2 (4.2 K, 1.5 T) obtained by Suo et al. [15] for sintered tapes.

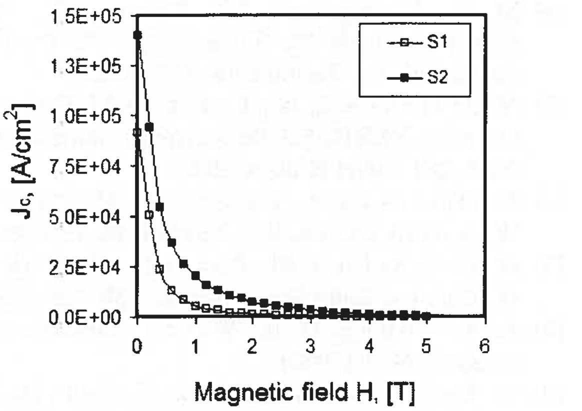

Fig. 5. Critical current density as a function of magnetic field for S1 and S2 samples

4. Conclusions

The two annealed Cu/Fe/MgB$_2$ rods were prepared successfully by the modified *PIT* technique with explosive consolidation method. The obtained rods were annealed at 750±1°C and 850±1°C for 1 hour in He atmosphere. XRD results show that the main phase is MgB$_2$ including strange MgO and SiO$_2$ phases. It was found that there are no reaction between Fe tubes and MgB$_2$ in both cases. The heat treatment process reveal that the Cu/Fe/MgB$_2$ rods annealed at 850°C has higher value lattice parameter of MgB$_2$ and SiO$_2$ than the Cu/Fe/MgB$_2$ rods annealed at 750°C. In the case of MgO the lattice parameter was stable. The numerical phases analysis reveals loss of the total content of MgB$_2$ phase. Both rods exhibit high intragranular transition temperature T_c = 38.7 K. The critical current density J_c for the Cu/Fe/MgB$_2$ rods heated at 850±1°C is equel 1.4×10^5 A/cm^2 (4.1 K, 0T) and it is higher than the critical current density J_c for the Cu/Fe/MgB$_2$ rods heated at 750±1°C. These results imply that the rods annealed at 850°C reveal more dense cores and better powder particles connection than rods annealed at 750°C.

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