Low-Field Behavior of Ti-Added MgB_2/Cu Superconducting Wires

A. Gencer, A. Kılıç, S. Okur, N. Güçlü, L. Özyüzer, and I. Belenli

***Abstract*—We report on low-field magnetic properties of Ti-added (0–20 wt. % of Ti) Cu-clad MgB_2 superconducting wires. Wires were produced by mixing appropriate amount of Ti and reacted MgB_2 powder which was then placed inside Cu tubes with a diameter of 6 mm. The tubes were then cold worked by rolling or drawing to smaller diameters and then annealed at various temperatures to enhance the grain connectivity. XRD studies show that Ti addition results in new but minor phases. We have then measured ac susceptibilities in the temperature range between 20 K and 40 K in ac fields of 20–1600 A/m. The data show that an additional loss mechanism is established with Ti-addition. The calculated ac losses are increasing with increasing Ti-content in the main superconducting matrix.**

***Index Terms*—AC susceptibility, critical current density J_c, granularity, MgB_2/Cu wire.**

I. Introduction

AFTER THE discovery of superconductivity in an intermetallic binary compound, MgB_2 [1], there has been a considerable interest in the optimization of its superconducting properties [2]–[7] by doping and/or adding other elements into MgB_2. The compound has been found to be particularly attractive given the possibility that it can have higher J_c and T_c compared to the low temperature superconductors. When the proper mechanical requirements are met, it may be possible to use the material for low-field applications. The powder-in-tube (PIT) method [4]–[6] appears to be the most practical and advantageous technique, but requires a suitable, nonreactive sheath material, such as Fe [4], Cu [5], [6], Ag [7]. Copper is one of the most suitable sheath materials for the fabrication of MgB_2 composite wires due to its low cost and high ductility. In addition, the grain size of MgB_2 is an important factor for grain connectivity in these superconducting wires. A poor connection between grains and lack of pinning centers are the causes of low critical current densities in these wires. To improve the critical current density J_c in the wires, some elements such as Ti [8]–[11], Al [12] inside MgB_2 have been used. It is expected that these added elements enhance the electrical connection between grains. The mechanical properties also become improved with the addition of these metallic elements. As an adding material, Ti has a hexagonal crystal structure similar to MgB_2. It can fill the voids and connect the grain boundaries since Ti has less molecular volume than MgB_2. Ti is a good electrical conductor and anticipated to improve connectivity between grains at the boundaries [13]. Annealing temperature and duration are other parameters to be controlled for increasing the grain connectivity. When adding Ti into MgB_2 careful treatment must be done to ensure a good homogeneity of MgB_2 and Ti mixture.

In this paper, as an extension of our previously published work [14], we discuss the superconducting properties of copper-clad Ti-added MgB_2 wires for samples annealed at 800°C. Measurements of fundamental susceptibility are reported as a function of temperature, ac field amplitude and frequency. We note that the results are strongly dependent on the magnitude of penetration field H_p in relation to the ac field amplitude.

II. Experimental Procedure

Ti (−200 mesh) powder was mixed with pure MgB_2 (−325 mesh from Alfa Aesar) with various weight percentages from 0 to 20%. Then the mixture was filled into a Cu tube with a wall thickness of 1 mm and outer diameter of 6 mm in air. Only hand pressing was applied to increase the density of MgB_2 powder mixture in Cu tube. The filled tube was cold drawn in a number of steps with about 5% of cross-section reduction down to 3 mm outer diameter. Then two-axial rolling method was used to prepare a rectangular Cu-clad MgB_2 wire with one side being 0.7 mm and the other side being 0.8 mm. The outer diameter of the wire is 1.5 mm, while the superconducting filling factor corresponds to about 25%–30% for the whole conductor volume.

The samples were then annealed in a tube furnace in a high purity Ar gas flow under ambient pressure at 800°C for 1 hour and then cooled in air. The phase assemblage of the Cu-clad MgB_2 filaments was analyzed by x-ray diffraction technique using Philips Expert Plus with Cu-K α. In our measurements of fundamental harmonic susceptibilities, we observe only a small frequency dependence and therefore use the critical state model as a good approximation for our wires.

III. Results and Discussion

We first present measurements. In Fig. 1, we plot XRD intensities as a function of 2 θ for nonannealed MgB_2 wire, 5%Ti added MgB_2, 10% Ti-added MgB_2, 15% Ti-added MgB_2

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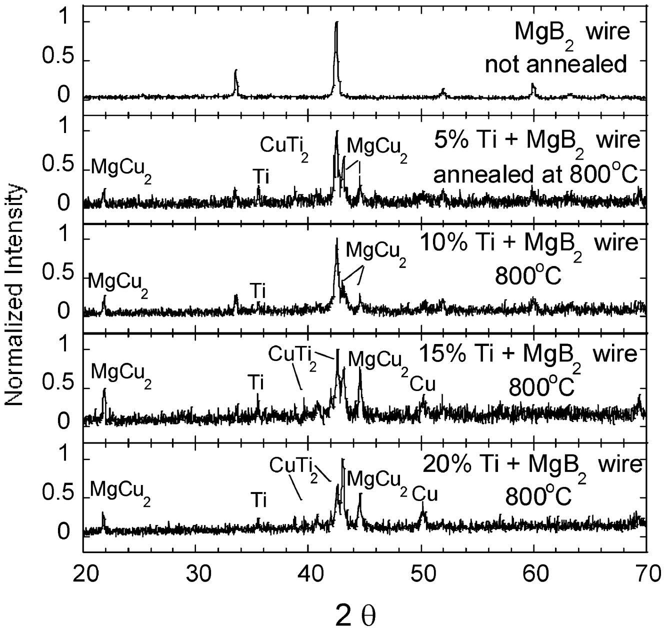

Fig. 1. X-ray diffraction patterns of Ti doped MgB_2/Cu wire after annealing at 800°C for 1 h in argon atmosphere.

and 20% Ti-added MgB_2. Ti-added samples were annealed at 800°C for 1 hour. Ti-addition has resulted in the production of new phases. These phases are $MgCu_2$, Ti, Cu and $CuTi_2$ which are also marked in graphics of Fig. 1. There are also some additional unidentified small peaks.

The peaks for Ti and $CuTi_2$ appear in the x-ray patterns. $MgCu_2$ and $CuTi_2$ peaks become visible due to interaction between Cu sheath and MgB_2+Ti core after an annealing process at 800°C for 1 hour. The superconducting MgB_2 peaks also disappear due to diffusion of Mg during annealing from the superconducting core into the interface between sheath and core to form inter-metallic $MgCu_2$ layer on the tube walls. The structural analysis dictates that the main peak for MgB_2 becomes smaller with increasing Ti-content. Although the purpose of Ti-addition was to fill the regions between grain boundaries, Ti also reacted with Cu and $CuTi_2$ peaks are observed. After completing x-ray characterization of our samples, we have then measured their ac susceptibilities between 20 K and 40 K.

In Fig. 2, we have plotted AC susceptibility $\chi_{AC}(T)$, for ac field amplitude of 20 A/m and 111 Hz frequency. The onset of diamagnetic transition of about 37.8 K is the same for all samples. However, there is a remarkable change resulting from Ti-addition in the shape of transitions. First, the magnitude of the out-of-phase component increases with increasing Ti-content. This observation implies that the ac losses become larger with Ti-addition. When a closes are large, the area in the M-H loop becomes larger. The large magnetization corresponds to the higher critical current densities. The magnitude of the in-phase component also increases with increasing Ti-content. There exists a single peak (upper peak) with tails for pure MgB_2, %5 and %10 Ti added samples. Two peaks are observed for % 15 and %20 Ti additions. Additional peaks have sometimes been observed, especially in the out-of-phase component

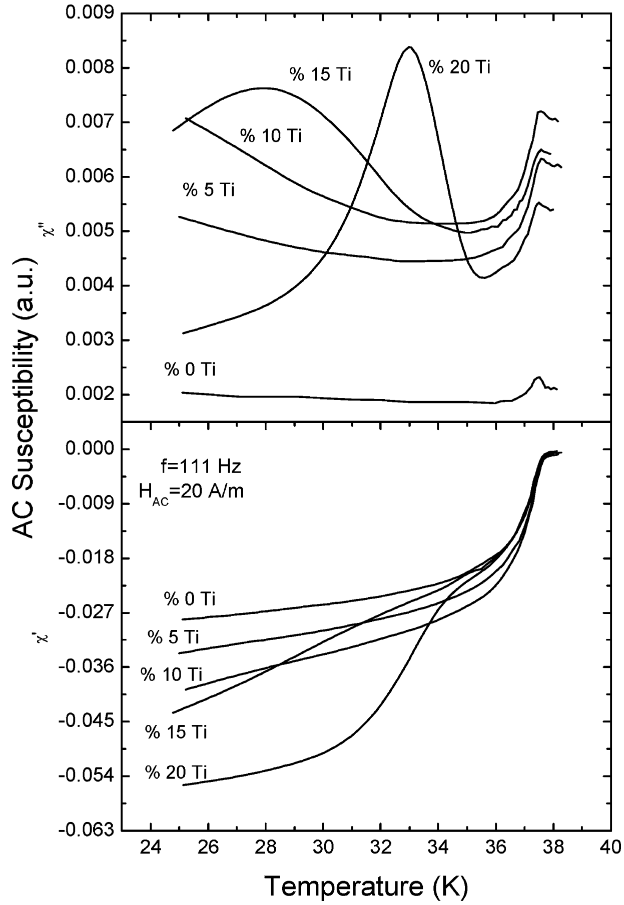

Fig. 2. AC susceptibility versus temperature at $f = 111$ Hz and $Hac = 20$ A/m for the samples studied, %0 Ti, %5 Ti, %10 Ti, %15 Ti, %20 Ti doped MgB_2/Cu wire.

of the fundamental susceptibility for high temperature superconductors [15], which could be attributed to several different loss mechanisms.

There could indeed be many reasons for the occurrence of a double peak. The most obvious one in our case appears to be inhomogeneous flux penetration into the samples. The inhomogeneous flux penetration can easily produce a two- or more-stage magnetic transition giving rise to the double-peak feature. Inhomogeneous flux penetration of this kind may result from the presence of macroscopic cracks, intergrowths and misoriented domains, nonuniform impurities and/or spatial variation of superconducting matrix [16]. Let us now suppose that our samples (%15 Ti-addition and 20% Ti-addition) have at least two dominating superconducting distinct regions behaving more or less independently, one with low current carrying capacity (matrix 1) and the other with high current carrying capacity (matrix 2). For larger ac fields, some explanations for the occurrence of the double peaks in AC susceptibility are possible. First, the occurrence of a small tail becoming a progressively larger peak with increasing Ti-content may be related to the weak inter-domain coupling as in a granular superconductor [15]. In this scenario, for sufficiently large ac amplitudes, the upper peak in out-of-phase component is associated with the irreversible motion in and out of the domains themselves, while the lower peak is related to irreversibility of

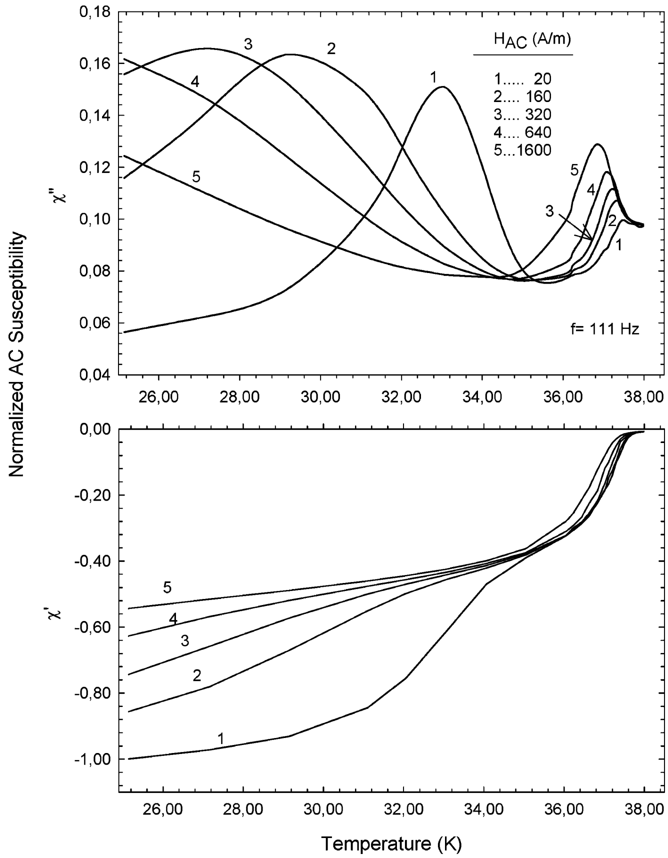

Fig. 3. Shows fundamental susceptibility of 20% Ti-added MgB_2 superconductor as a function of temperature for various ac field amplitudes at measuring frequency of 111 Hz.

flux line motion in regions between superconducting domains (assumed to be composed of well connected grains). One may conclude that Ti-addition has resulted in the occurrence of granularity in the superconductor. The ac fields that can be applied in our measuring system are limited to 1600 A/m (r.m.s). With the application of such fields, one can observe double-peak feature for higher Ti-added samples. A similar behavior has been observed in a single phase Bi-2212 sample [16]. The presence of double peak in the out-of-phase component appears to be caused by the granular character of Ti-added MgB_2 samples. Similar double peak behavior in the out-of-phase component for lower Ti-added samples (%5 and %10) would have been observed if higher ac field amplitudes were applied. However, note that we have used relatively large ac field amplitudes than that of routine ac characterization measurements.

Although ac losses seems to be increasing with Ti-addition, the bulk superconductivity is established at lower temperatures.

In Fig. 3, we plot the fundamental susceptibility of 20% Ti-added MgB_2 as a function of temperature for various ac field amplitudes. The height of the upper peak increases with increasing ac field amplitude. This is associated with irreversible flux penetration into the intra-grains of superconducting MgB_2. In addition, the curves shift to lower temperatures with increasing ac field amplitude. We only show the ac susceptibility of the sample with the highest Ti-content (20% Ti). The results for other samples are not shown since they have similar features. The measurements are normalized at 25 K in order

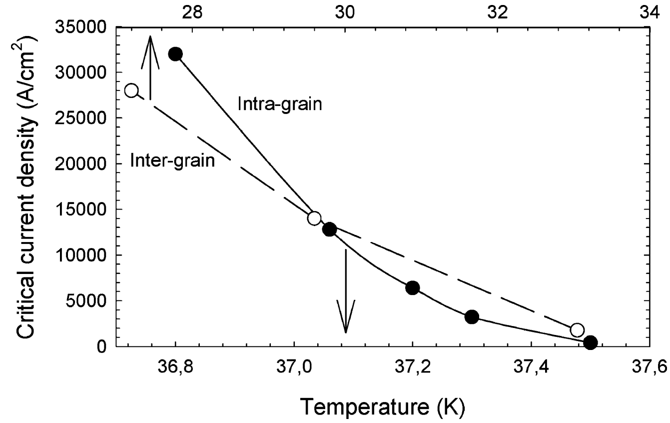

Fig. 4. Inter- and intra-grain critical current density for 20% Ti doped MgB_2/Cu wire calculated from AC susceptibilities at $f = 111$ Hz and various ac field amplitudes.

to make quantitative comparison. For the field of 20 A/m, the magnetic transition has a two-stage (lower graph). The upper sharp decrease is associated with the occurrence of superconductivity within the grains (intra-grain), while the gradual change in the in-phase component at lower temperatures is attributable to the screening currents between superconducting grains (inter-grain). This indicates accumulation of Ti at grain boundaries. Chemical x-ray mapping of a polished cross-section, which is in progress, may give more direct evidence. In the literature, the most recent study by Wilke *et al.* [17] shows that Ti forms precipitates of either TiB or TiB_2 that enhance the flux pinning and raise J_c. They also found that while J_c is increasing, H_{c2} becomes decreased. Therefore, our interpretation of the observed dependences are consistent with the findings in [17].

The bulk superconductivity is established at around 25 K for the field of 20 A/m. The bulk superconductivity is established at lower temperatures for higher ac field amplitudes. The peak positions in the out-of-phase component also shifts to lower temperatures with increasing ac field amplitudes. From the positions of the peaks, one can estimate the critical current densities at the peak temperatures. In this case, there exist two different critical current densities for intra-grain and inter-grain properties.

In Fig. 4, we plot magnetically estimated critical current densities for intra-grain and inter-grain regions at temperatures close to T_c. The upper axis is used for temperature scaling for inter-grain (transport) critical current density, while the lower axis is for intra-grain critical current density. The diameters of the wires are about 1 mm. The data of Fig. 4 is obtained as follows: When the out-of-phase component in ac susceptibility peaks, full flux penetration occurs. At the peak temperature, for a cylinder with radius r, the magnetically estimated Jc is given as: $J_c = H_{ac}/r$. For intra-grain super-currents, we take average grain size instead of the radius, r. Note that this is only a qualitative method. From the comparison, the inter-grain critical current densities are expected to be lower than the intra-grain critical currents simply because of temperature shift.

It is well known that the out-of phase component of the fundamental ac susceptibility is a direct measure of the ac losses as the Bean model estimates that $\chi'' = A_H/\mu_0 \pi H_{ac}^2$, where A_H is

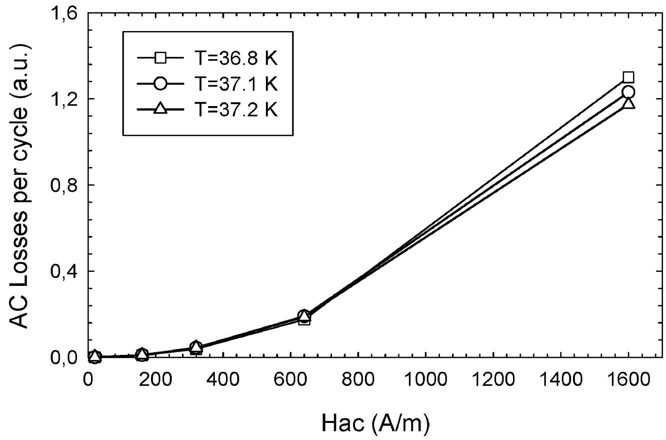

Fig. 5. The plot of experimental ac losses versus H_{ac} for three different temperatures is given.

the area embraced by the hysteresis curve. We have also plotted the experimentally measured ac losses versus ac ?eld amplitude in Fig. 5 for three different temperatures ($T = 37.2$, 37.1 and 36.8 K). It can be found from Fig. 5 that ac losses exhibit a parabolic curvature as a function of H_{ac}^2. The measurements are qualitatively consistent with Bean model.

IV. Conclusion

In summary, Ti-added MgB_2 composite wires have been characterized by use of XRD and ac susceptibility measurements. Ti-addition results in the increase of ac losses. The increase in ac losses appears to be an indication of the presence of the higher critical current densities. However, granularity and accumulation of Ti at the grain boundaries become apparent with increasing Ti-addition.

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