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Estimation of critical current density and grain connectivity in superconducting MgB_2 bulk using Campbell's method

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Abstract

Many recent reports on the critical current density (J_c) in superconducting MgB_2 bulks indicated that improving the grain connectivity is important, since the obtained J_c values were generally much lower than those in other metallic superconductors and it was ascribed to the poor connectivity between grains in polycrystalline MgB_2. In this study, we focused on the estimation of the global critical current density, super-current path, grain connectivity and their relationships with the faults volume fraction in the MgB_2 bulks prepared by a modified PIT (powder in tube) method. Campbell's method was applied for the purpose of obtaining the penetrating AC flux profile and the characteristic of AC magnetic field vs. penetration depth from the sample's surface. A computer simulation on the penetrating AC flux profile in MgB_2 bulks with randomly distributed voids, oxidized grains and other faults was also carried out. J_c obtained by

Campbell's method turned out to be smaller than that obtained from the SQUID measurement, implying that the global super-current was reduced by the existence of various faults and the lack of the electrical connectivity. It was verified that the relationship between the global critical current characteristics and the faults contained in MgB_2 samples can be quantitatively clarified by comparing the simulated critical current densities and other factors with the experimental results.

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Keywords: Polycrystalline MgB_2 bulk; Campbell's method; AC flux profile; Grain connectivity

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1. Introduction

Since the discovery of superconductivity in MgB_2 [1] in 2001, great progress has been made in clarifying the superconducting properties, optimizing the synthesis methods, understanding the flux pinning mechanisms and so on. Because of its outstanding properties of high critical temperature and high upper critical field comparing with other metallic superconductors, as well as low anisotropy and less weak links at grain boundaries, a wide range of applications that using MgB_2 are greatly expected.

Many reports maintain that the grain boundaries are one of the dominant pinning centers in polycrystalline MgB_2. Several results succeeded to show an apparent correlation between the grain size and the critical current density in their MgB_2 samples. However, it is pointed out [2] that the critical current densities in polycrystalline MgB_2 are far below the values observed in Nb_3Sn under the same conditions, although it is considered that the flux pinning mechanisms originating in grain boundaries in these two materials resemble each other. Rowell [3] suggested that the reduction of the effective current-carrying cross-sectional area by the existence of voids, oxide phases present in the grain boundaries and other impurity factors in polycrystalline MgB_2, which directly lead to a reduction of super-current path, is one of the causes that largely suppress the critical current characteristic. Yamamoto [4] explained the relationship between the normal-state conductivity and the electrical connectivity in superconducting MgB_2 by using a site-percolation model, and concluded that the low packing factor and poor connectivity are the fatal reasons restricting the critical current density in MgB_2.

On the other hand, it is known that the AC inductive methods, for example, Campbell's method [4], are useful for the estimation of the critical current characteristics in polycrystalline superconducting materials. One of the examples is the study on the distinguishing between J_c's of the inter- and intra-grain in polycrystalline YBaCuO bulk by Campbell's method [5]. In this study, we applied Campbell's method as an effective mean for the discussion and estimation on the critical current density, super-current path and their relationships with the faults volume fraction in the polycrystalline MgB_2 bulks. We also carried out a computer simulation on the penetrating AC flux profile in MgB_2 bulks with randomly distributed voids, oxidized grains and other faults. By comparing the simulated critical current densities and other factors with the experimental results, we concluded that Campbell's method is an effective and reasonable method for understanding the electromagnetic behaviors in polycrystalline MgB_2.

2. Experimental

The samples were prepared by a modified PIT (Powder-In-Tube) method. Powders of magnesium (99.99%, 125 – 300 μm), amorphous boron (99.99%, 300 mesh) and commercial MgB_2 powder were used for the starting materials. The molar ratio of Mg : B : MgB_2 was 1.2:2:0.5. The mixture of B and MgB_2 was packed in a SUS pipe, accompanied with Mg powder filled at two ends of the mixture in the pipe. Then, the pipe was pressed and sealed at two ends by TIG welding. The pipes containing powders were heat treated at a fixed temperature of 900 ^{o}C, while the times of the heat-treatment were varied from 12 h to 300 h, in order to bring variations in the average grain size and the extent of oxidation in MgB_2. The densities of most of the samples, estimated by Archimedes method, were larger than the value of 90% of the theoretical density. In fact, SEM images of those samples with high densities show few voids observed, denoting that the packing factor was quite high. However, XRD results show some small peaks corresponding to MgO, implying that curtain oxidation occurred during the process of sample preparation.

The critical temperature T_c's of samples were measured by a SQUID magnetometer. It is found that all samples show the T_c values closing to 38.5 K, with the deviations smaller than 1 K. J_c was measured and estimated by DC magnetization (SQUID magnetometer) and Campbell's method, respectively. The temperature of sample was 4.2 K – 35 K, while the magnetic field was up to 6 T for SQUID measurement and 0.4 T for Campbell' method measurement. The frequency of the AC magnetic field in Campbell's method was varied from 37 Hz to 797 Hz.

3. Results and Discussion

The magnetic field dependence of J_c estimated from measured DC magnetization at T = 20 K is shown in Fig.1. As a pure MgB_2 bulk without any carbide doped, the sample with 12 hours heat-treatment shows a tolerable J_c value, while the samples experienced a longer heat-treatment time show an apparent decrease in J_c value. The fact could be considered as the result of the grain's coarsening in MgB_2, since the grain boundaries are one of the dominant pinning centers, and the measured grain size was increased according as the heat-treatment time of the samples lengthened. Another reason which reduces J_c value is probably the oxidizing progress of the surface of MgB_2 grains, since it is reasonable to consider that the longer heat-treatment time lasts, the more oxidizing proceeds.

As one of the measurements dealing with the electromagnetic properties in superconductor, Campbell's method measures the penetrating AC flux corresponding to the applied DC and variable AC magnetic fields. By analyzing the measured AC flux profile and AC magnetic field b_{ac} vs. penetration depth λ' curve, one can derive not only the critical current density but also the relationship between the force on and the displacement of the flux lines. In this study, we focused our attention on the AC flux profile itself and the estimation of global J_c in MgB_2 samples.

Taking the factor of electric field criterion, which defines the critical current density in superconductors, into account, J_c value from Campbell's method is usually overestimated, since the induced electric field under the applied AC magnetic field with several tens Hz or more should be larger than those in DC magnetization method or resistive method. If the n value in the current-voltage curve ($V \propto I^n$) is sufficient large, then, the error caused by the overestimation should be negligible. Fig.2 shows a comparison of J_c values at T = 20 K obtained from DC magnetization and Campbell's method, respectively. Contrary to our expectations, J_c from Campbell's method shows a smaller value. Considering the fact that in Campbell's method J_c was derived from the initial part of the b_{ac} vs. λ' curve, meaning that the value is corresponding to the global current in MgB_2 bulk, while J_c from DC magnetization was estimated based on the average magnetization per unit volume, meaning that the value is just an average one. The result strongly implies that the super current in MgB_2 bulk flows in a complicated path, bringing a reduction in current density value.

In order to give a more quantitative discussion, a computer simulation of penetrating AC flux profile was carried out. Supposing that MgB_2 bulk contains some certain amount of randomly distributed voids and grains which have an oxide layer on their surface, and AC magnetic field is sufficient small so that the influence on critical current density is negligible, then, the penetrating AC flux profile can be simulated and illustrated as Fig.3. A rectangle shape of cross section area was assumed and only half of the area was shown for the symmetricalness, i.e., the bottom line of Fig.3 is the center line of the MgB_2 bulk, and the AC flux penetrates from the rest of the three sides. In the simulation, the voids and the grains with oxide layer on the surface were approximated to a cubic normal phase with same size.

Based on the penetrating AC flux profile as Fig.3, the b_{ac} vs. λ' curve can be easily obtained according to the equation used in Campbell's method as

$$\lambda' = \frac{1}{4}(w+d)\left[1-\left(1-\frac{4}{(w+d)^2}\frac{\mathrm{d}\Phi}{\mathrm{d}b_{\mathrm{ac}}}\right)^{\frac{1}{2}}\right],$$

where Φ, w and d are the penetrating AC flux, sample's width and thickness, respectively. Fig.4 shows the calculated b_{ac} vs. λ' curves in the samples with different faults volume fractions, f. Since the critical current density is calculated from the gradient of the initial part of b_{ac} vs. λ' curve in Campbell's method, It is obvious that the increase of faults amount leads to a decrease of J_c value. As an example, normalized J_c derived from the calculated b_{ac} vs. λ' curve are shown in Fig.5.

On the other hand, the DC magnetization, which is usually obtained from SQUID magnetometer or other inductive measurements, can also be simulated by the penetrating AC flux profile. In this case, the penetration magnetic field B_p was defined as the AC magnetic field at which the AC flux reaches the center of the superconductor. The averaged value of the local and global J_c was calculated by applying the equation of the critical state model. Fig.6 shows a comparison with the experimental data [4] obtained from the DC magnetization of MgB_2 bulks with different packing factors. In Fig.6, the packing factor defined in reference [4] is identified with 1 – f in our simulation. The good agreement suggests that the assumptions of the faults existence and the tangled current path in MgB_2 bulk in our simulation are reasonable.

4. Conclusion

As an effective method for the estimation on the critical current density, super-current path, grain connectivity and their relationships with the faults volume fraction in the polycrystalline MgB_2 bulks, Campbell's method was applied for the purpose of obtaining the penetrating AC flux profile and the AC magnetic field vs. penetration depth from the sample's surface. A computer simulation on the penetrating

AC flux profile in MgB_2 bulks with randomly distributed voids, oxidized grains and other faults was also carried out. The critical current density obtained by Campbell's method turned out to be smaller than that obtained from the SQUID measurement, implying that the global super-current was reduced by the existence of various faults and the decrease of the electrical connectivity. By comparing the simulated critical current densities and other factors with the experimental results, it was concluded that Campbell's method is an effective and reasonable method for understanding the electromagnetic behaviors in polycrystalline MgB_2.

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Figure captions

Fig. 1 Magnetic field dependence of the critical current densities obtained from the SQUID measurement at T = 20 K. The MgB_2 bulks experienced different heat-treatment time of 12 hours to 300 hours.

Fig. 2 Comparison of the critical current densities at T = 20 K form DC magnetization method (square symbols) and Campbell's method (circle symbols). The sample experienced a 12 hours heat-treatment.

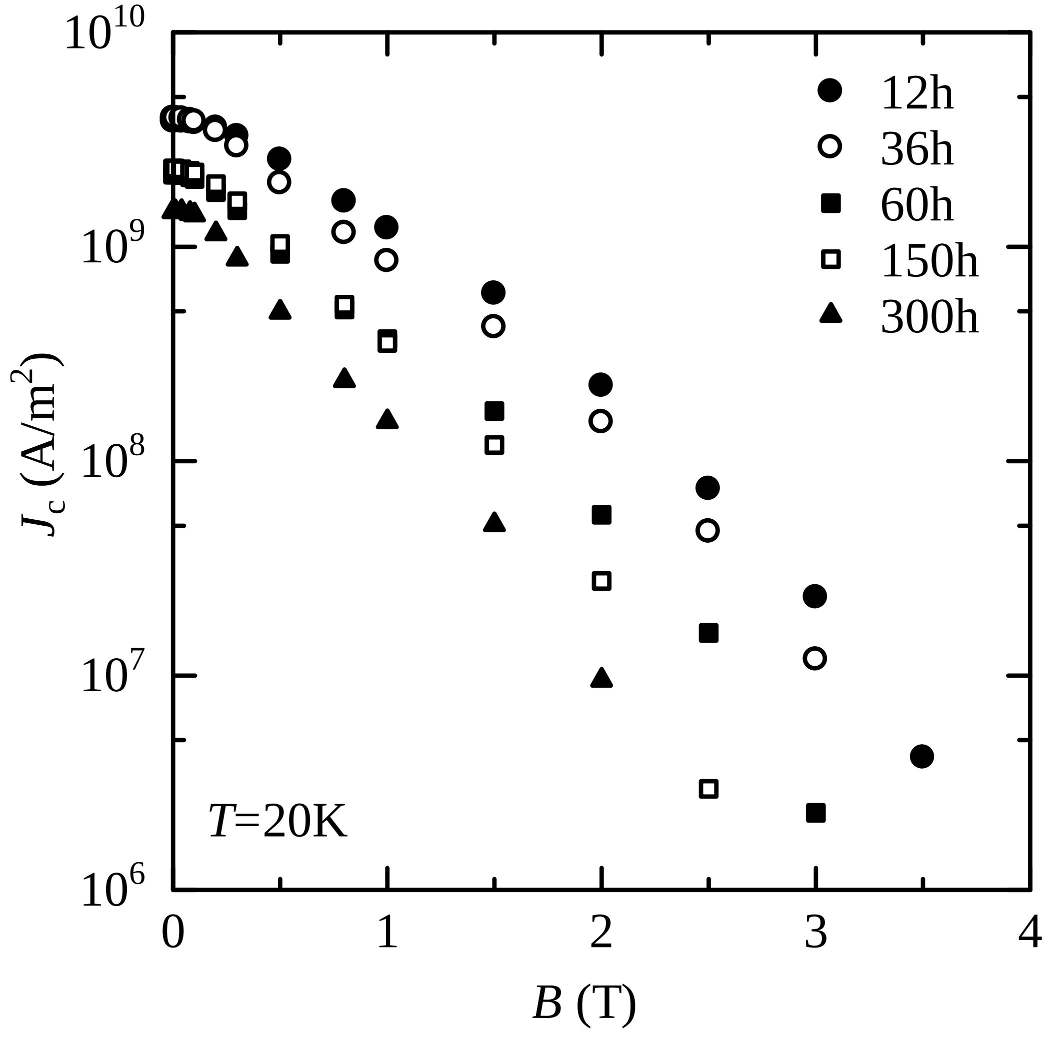
Fig. 3 Simulation of penetrating AC flux profile in polycrystalline MgB_2 bulk. It is supposed that MgB_2 bulk contains some certain amount of randomly distributed voids and grains which have an oxide layer on their surface. A rectangle shape of cross section area was assumed and only half of the area was shown, i.e., the bottom line of the image is the center line of the MgB_2 bulk, and the AC flux penetrates from the rest of the three sides. The voids and the grains with oxide layer on the surface were approximated to cubic normal phases with same size, which are denoted by blue square areas.

Fig. 4 Calculated b_{ac} vs. λ' curves in the MgB_2 bulks with different faults volume fractions.

Fig. 5 Normalized J_c vs. f derived from the calculated b_{ac} vs. λ' curve shown in Fig.4.

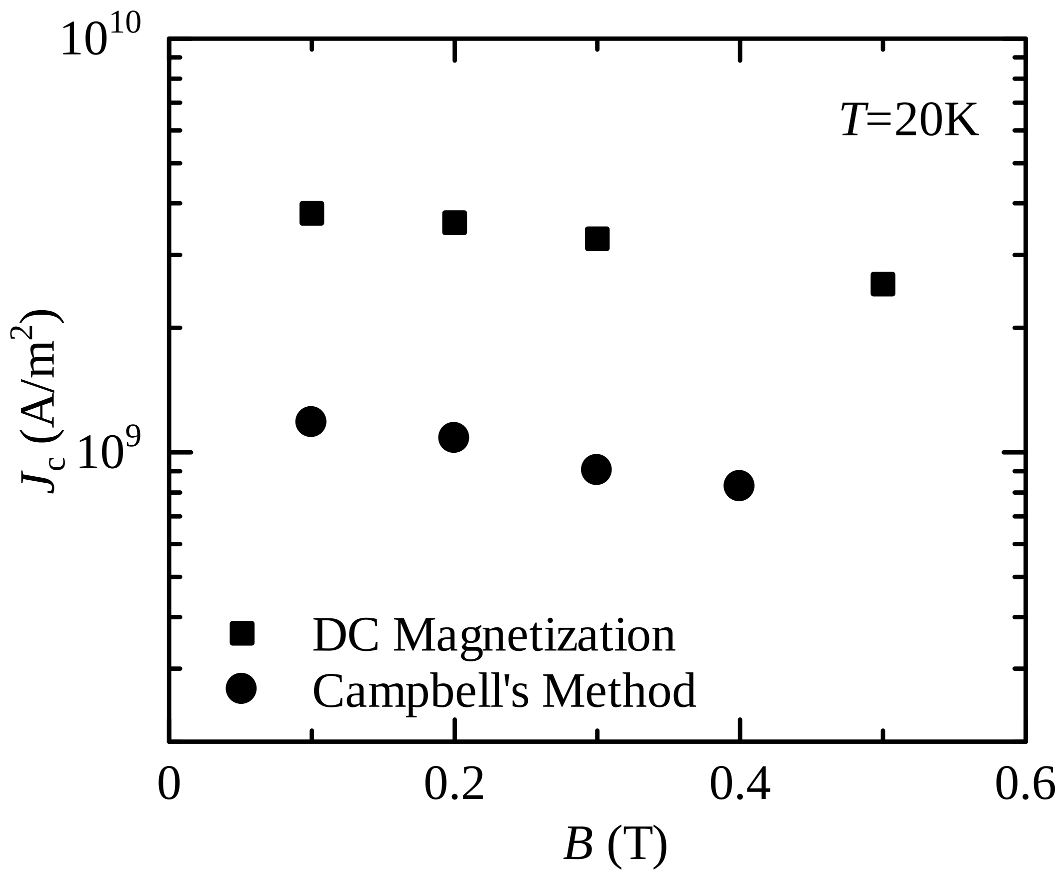
Fig. 6 Comparison of the calculated normalized J_c values and the experimental data [4] obtained from the DC magnetization of MgB_2 bulks with different packing factors. The packing factor defined in reference [4] is identified with $1 - f$.

Figure 1



B. Ni

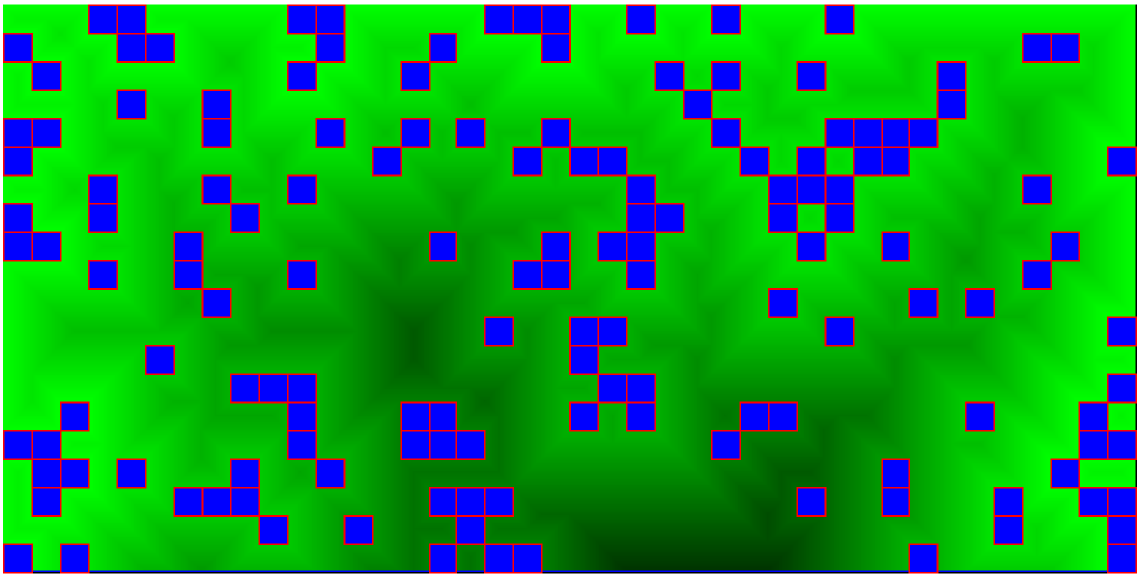
Figure 2



10^{10}
10^{9}
T=20K
DC Magnetization
Campbell's Method
J_c (A/m^2)
0
0.2
0.4
0.6
B (T)

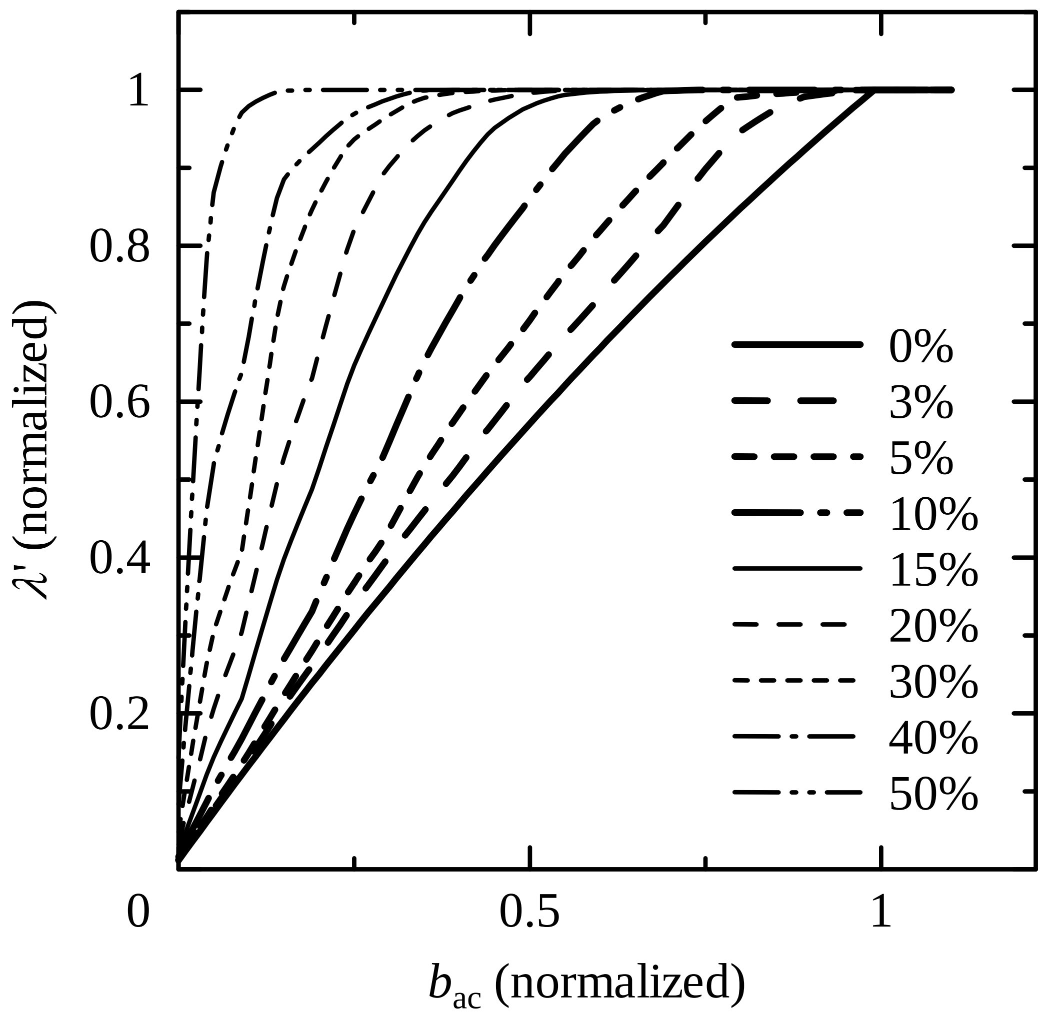
B. Ni

Figure 3



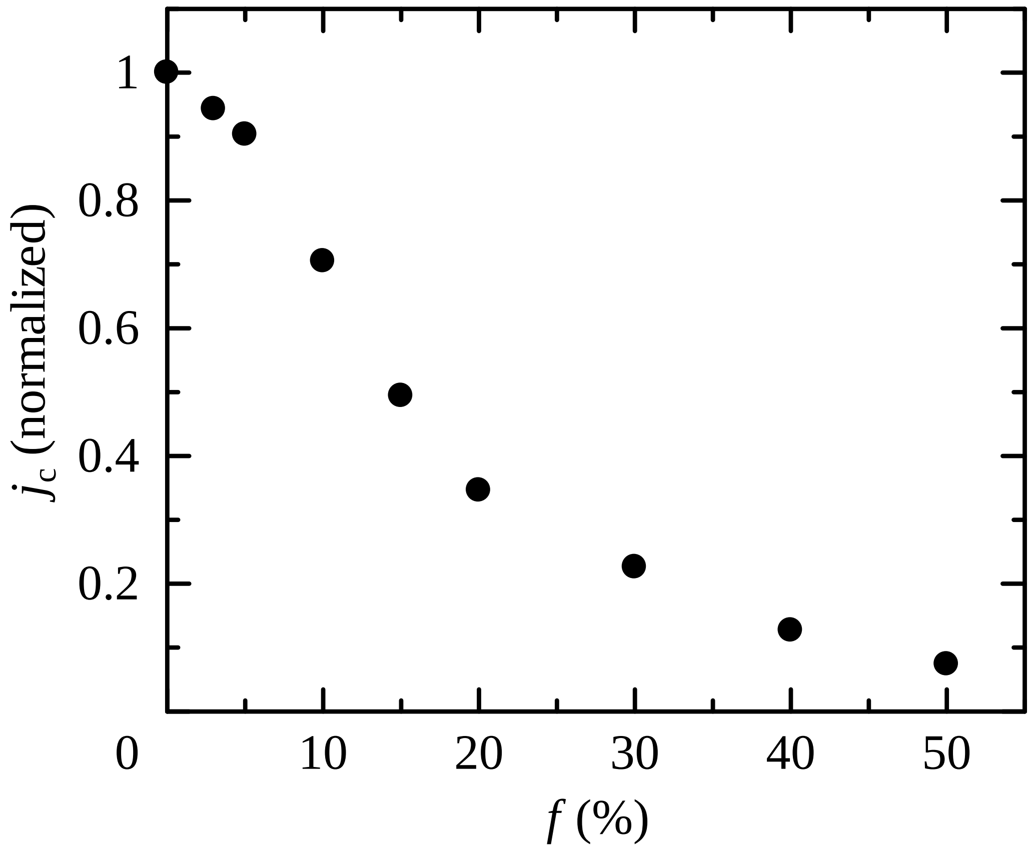
B. Ni

Figure 4


1
0.8
0.6
0.4
0.2
0
0.5
1
λ' (normalized)
b_ac (normalized)
0%
3%
5%
10%
15%
20%
30%
40%
50%

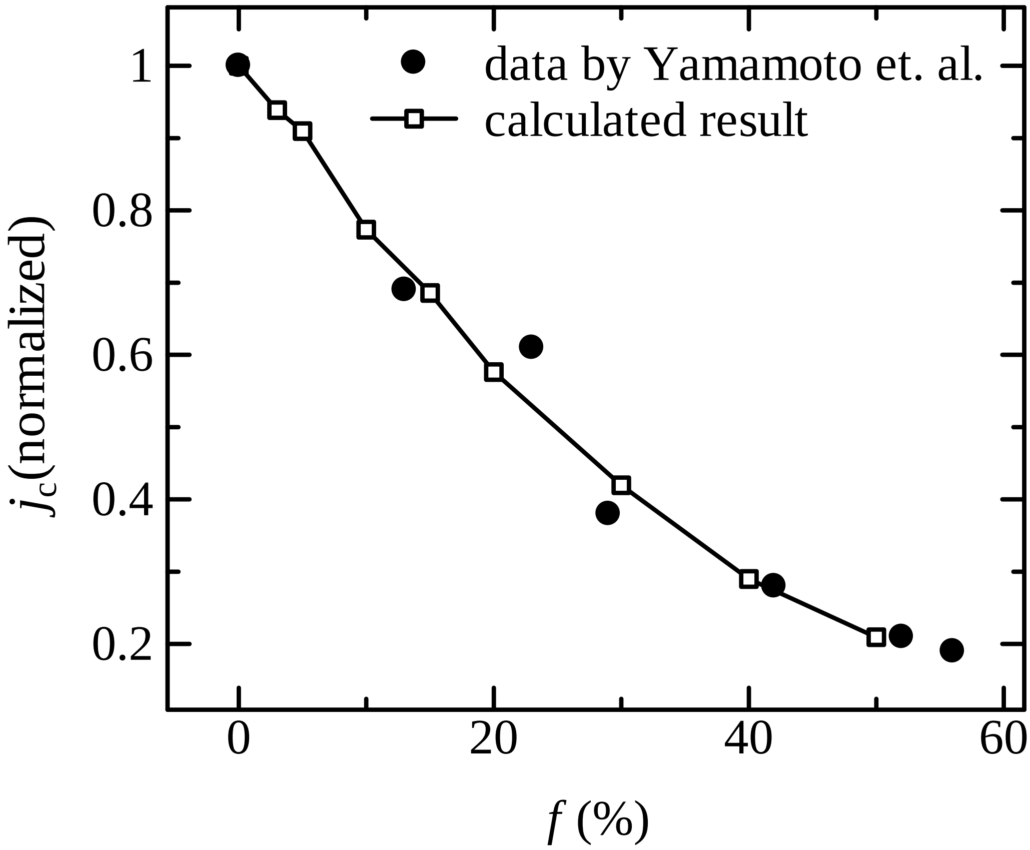
B. Ni

Figure 5



B. Ni

Figure 6



data by Yamamoto et. al.
calculated result
1
0.8
0.6
0.4
0.2
j_c(normalized)
0
20
40
60
f (%)

B. Ni