Flux pinning properties of (Nd, Eu, Gd)$Ba_2Cu_3O_y$ (NEG-123) superconductor with 211 phase particles

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Abstract. Flux pinning properties were investigated with focusing on the origin of the high peak critical current density and the high irreversibility field for $Nd_{0.33}Eu_{0.38}Gd_{0.28}Ba_2Cu_3O_y$ (NEG-123) bulk superconductors with addition of 211 secondary phase particles of the volume fractions up to 10 mol%. It was found that a negative correlation exists between the peak critical current density $J_{\rm cp}$ and the irreversibility field $B_{\rm i}$ under a variation in the fraction of added 211 phase. This suggests that the mechanisms which determine $J_{\rm cp}$ and $B_{\rm i}$ are different. The 211 particles do not contribute to the peak effect neither directly through the pinning mechanisms of the condensation energy interaction nor indirectly with the aid of the order-disorder transition of flux lines. Another possible defects are nano-lamella structures. Although these defects do not directly contribute to the peak effect by the pinning mechanism of condensation energy interaction, those may contribute to it with the aid of the order-disorder transition of flux lines. On the other hand, since $B_{\rm i}$ is deteriorated with addition of 211 particles, it is not determined by the flux pinning of 211 particles. The decrease in $B_{\rm i}$ is considered to be caused by the proximity effect of nano-lamella structures.

1. Introduction

Among high-temperature superconductors, $REBa_2Cu_3O_y$(RE denotes a rare earth element, RE-123) is one of the most interesting materials to study for its pinning properties, as it is considered to be a strong candidate for practical applications. Superior performance has been reported for higher peak critical current density J_{cp} and irreversibility field B_i in (Nd, Eu, Gd)-123 (NEG-123) bulk superconductors at 77.3 K as compared to Y-123 or Nd-123[1-4]. However, further improvement of these properties is still needed for facilitating applications. To achieve the higher J_{cp} and B_i, it is necessary to understand the flux pinning mechanism which brings about high J_{cp} and B_i.

For the explanation of the peak effect in RE-123 superconductors, two mechanisms have been proposed: the elementary field-induced pinning[5] and the order-disorder transition[6] of flux lines. In $NdBa_2Cu_3O_y$ (Nd-123) superconductor, it is known that there exists a low T_c phase in which Ba sites are substituted by Nd atoms. Although the substituted regions are superconducting at low fields and may not strongly contribute to the flux pinning, those become normal state at high fields and may contribute strongly to the flux pinning. This is the field-induced pinning mechanism and explains the peak effect[5]. The similar mechanism may also be expected for oxygen deficient regions with lower T_c in Y-123. That is, it is speculated that J_c starts to increase when the magnetic field reaches a lower edge of the distributed upper critical field of the low T_c phase. On the other hand, it has been proposed that the increase of J_c with magnetic field is ascribed to the transitional change from an ordered state of flux lines with a weak pinning force to a disordered state with a strong pinning force in the order-disorder transition model[6].

In previous measurements[7, 8], it was found that J_c decreased in the medium field region and the peak effect disappeared by addition of 211 phase particles to Y-123 bulk superconductor. Also, Mochida *et al.*[9] found that the peak effect at medium fields decreased by addition of Nd-422 particles in Nd-123. This means that the reduction in $J_{\rm cp}$ is ascribed to the interference between the repulsive pinning (positive pinning energy) of lower-$T_{\rm c}$ regions and the attractive pinning (negative pinning energy) of 211 phase particles (Nd-422 or Y-211)[8], suggesting that the pinning mechanism of lower-$T_{\rm c}$ regions is not the attractive one based on the field-induced pinning mechanism, a kind of the condensation energy interaction. The expected mechanism of lower-$T_{\rm c}$ regions is the repulsive kinetic energy interaction under the proximity effect[10].

The irreversibility field is the upper limit of magnetic field for practical applications at which the critical current density is reduced to zero and can be explained by the flux creep theory[11] which is based on the thermal depinning mechanism of flux lines. Thus, the irreversibility field depends on the flux pinning strength. It was found in[9] that a high irreversibility field $B_{\rm i}$ can be achieved in Nd-123 sample with a large amount of Nd-422 inclusions. The $B_{\rm i}$ shifts to higher fields with increasing Nd-422 phase, indicating that Nd-422 are effective in enhancing $B_{\rm i}$. The enhancement in $B_{\rm i}$ by adding 211 phase can also be found in Y-123[12, 13, 14]. These results are consistent with the results[15] that $J_{\rm c}$ values in the high field region are increased with increasing $V_{\rm f}/d$, where $V_{\rm f}$ is the volume fraction and d is the average diameter of 211 particles. Hence, it can be concluded that the pinning mechanism of the irreversibility field and that of the peak effect are different in Y-123 or Nd-123. In fact $J_{\rm cp}$ is likely to decrease while $B_{\rm i}$ tends to increase by addition of 211 or 422 phase in these superconductors. That is, $B_{\rm i}$ is determined from the attractive pinning by 211 phase particles which is dominant at high magnetic fields, and $J_{\rm cp}$ is determined from the repulsive pinning interactions by oxygen deficient regions or substituted regions.

For practical applications, it is still required to improve $J_{\rm cp}$ and $B_{\rm i}$ of bulk superconductors. Recently, new $Nd_{0.33}Eu_{0.33}Gd_{0.33}Ba_2Cu_3O_y$ bulk superconductor called NEG-123 system has been developed and it has been demonstrated that the flux pinning properties of the new material are superior to those of Y-123 or Nd-123. Muralidhar *et al.*[16] observed the enhancement of $J_{\rm cp}$ and $B_{\rm i}$ at 77.3 K in $Nd_{0.33}Eu_{0.33}Gd_{0.33}Ba_2Cu_3O_y$ superconductors with addition of 10-40 mol% NEG-211 secondary phase. They found that J_{cp} increased and B_i also almost increased with increasing NEG-211 phase: a large J_{cp} of 70 kA/cm^2 at B=2.3 T and B_i over 7 T were achieved by addition of 40 mol% NEG-211 phase. Later, they measured[17] J_c-B properties at 77.3 K in $Nd_{0.33}Eu_{0.33}Gd_{0.33}Ba_2Cu_3O_y$ superconductors with addition of various 211 phases, i.e., NEG-211, EG-211, Eu-211 and Gd-211 of 0-50 mol%. In these measurements, J_{cp} almost increased with increasing volume fraction of various 211 phases, and B_i increased up to a maximum value at a certain mol% of 211 phase and then decreased. The maximum B_i depended on the kind of 211 phase and their amounts. Thus, the behaviors of J_{cp} and B_i with addition of 211 phase in the new superconductors are complicated.

In order to understand the change in J_c-B characteristics, they performed chemical analyses through microstructural observation and found a change in the chemical composition of a NEG-123 specimen with 211 phase addition, which might affect the J_{cp} and B_i. They speculated [16] that such peak effect was ascribed to the field-induced pinning by RE-rich clusters dispersed in NEG123 systems with 211 particles addition, and that such field-induced pinning was active in parallel with the pinning by 211 particles. On the other hand, a pronounced peak effect with J_{cp} of 70 kA/cm^2 (peak at B=4.5 T) and high B_i over 14 T were observed at 77.3 K in NEG-123 with different composition of Nd, Eu and Gd elements, i.e., $Nd_{0.33}Eu_{0.38}Gd_{0.28}Ba_2Cu_3O_y$ with 5 mol% NEG-211 inclusion[18]. They found new pinning centers, i.e., nanoscale lamellae structure of NEG/Ba-rich clusters and proposed that such new structures played the significant role for the increase in B_i. In previous measurements, however, a clear evidence was not given for the above speculation on the pinning centers and corresponding mechanisms responsible for the increases in J_{cp} and the decreases in B_i with 211 phase addition to NEG-123 materials. It was also found[1, 19, 20] that the pinning performance could be tailored in NEG-123 by controlling chemical ratio of Nd, Eu and Gd elements together with addition of 211 phase to achieve high J_{cp} and B_i. For further improvement of J_{cp} and B_i, such kind of explanation is very important.

In the present study, we focus on the most promising ($Nd_{0.33}$, $Eu_{0.38}$, $Gd_{0.28}$)$Ba_2Cu_3O_y$ superconductor which shows a high performance at high fields and study the variations of $J_{\rm cp}$ and $B_{\rm i}$ with volume fractions of 211 phase to find out the mechanisms which determine the peak effect and $B_{\rm i}$. Then, a discussion is given on the possible defects and the corresponding pinning mechanism which can explain the variation in $J_{\rm cp}$ and $B_{\rm i}$ with the volume fractions of 211 phase.

2. Experimental

High-purity commercial powders of Nd_2O_3, Eu_2O_3, Gd_2O_3, $BaCO_3$ and CuO were mixed in the amounts corresponding to the nominal composition of ($Nd_{0.33}$, $Eu_{0.38}$, $Gd_{0.28}$)$Ba_2Cu_3O_y$. Next, commercial Nd-422, Eu-211 and Gd-211 powders were mixed in the ratio 1:1:1 for preparing NEG-211 secondary phase particles and then, Eu-211 and Gd-211 were mixed in the same ratio for EG-211 secondary phase particles. The preparation procedure was the same as described in Ref.[19,20]. Then, the samples added with the volume fractions of 3, 7, 10 mol% of NEG-211 and 3, 5, 7, 10 mol% of EG-211 were subjected to the oxygen-controlled melt-growth process in flowing 0.1% pO_2/Ar at 300 ml/min. The details of the heat treatment profile can be found in [1]. The size and dispersion of 211 phase in the 123 matrix depends on the initially added 211 size. In the present case, 75 nm sized 211 phase particles were used before melt processing and one can expect that the final size of 211 particles between the 100 to 500 nm, which was already confirmed by TEM analysis and also by high angle dark field scanning transmission electron microscopy with atmic resolution [21]. Muralidhar *et al.* [18, 20, 22] investigated also the microstructure for ($Nd_{0.33}$, $Eu_{0.38}$, $Gd_{0.28}$)$Ba_2Cu_3O_y$ with addition of 3, 5, 7, 10 and 40 mol% NEG-211 phase, which revealed that a new defects, i.e., the nano-lamella structure systematically appeared in samples with 3-10 mol% of NEG-211 phase and these nano-lamella structure was absent in the one with 40 mol% NEG-211 phase. From the STM analysis they found that the average spacing of the nano-lamella structure was around 3-4 nm (3 mol%) and again increased with increasing 211 phase, i.e., the change of nano-lamella structure was observed by addition of 211 phase [18, 20, 22].

The magnetization in a magnetic field along the *c*-axis was measured using the Quantum Design SQUID magnetometer. The onset superconducting transition temperature of all the samples was in the range 92.5-93.3 K. The critical current density J_c was estimated from the measured magnetization hysteresis using the Bean model. The irreversibility field was determined by the field at which J_c was reduced to 1×10^6A/m^2. The measurements were carried out in the magnetic field range 0.01 T-7 T and in the temperature range 77.3 K-80 K.

3. Results and discussion

In Fig. 1(a) and 1(b), the magnetic field dependence of J_c with different volume fractions of NEG-211 and EG-211 phases at 80 K is shown, respectively. A pronounced increase in J_c at peak field region occurs by increasing the volume fraction of both NEG-211 and EG-211 phases. On the other hand, the J_c at high fields is deteriorated with addition of 211 phase suggesting a decrease in B_i. The dependences of J_{cp} and B_i on the volume fractions of 211 phase are shown in Fig. 2. Figure 3 shows the dependence of J_{cp} on the volume fractions of 211 phase at 77.3 K. The highest value of J_{cp} obtained for the specimen with 10 mol% NEG-211 reached almost 6×10^8A/m^2. At this temperature the irreversibility field exceeded 7 T and was too high to measure. It is found from Fig. 2 that J_{cp} increases with increasing volume fraction of EG-211 phase. A similar dependence is obtained also for addition of NEG-211 phase, although J_{cp} is minimum at 7 mol% of NEG-211 phase. The same behavior is observed at 77.3 K as shown in Fig. 3. On the other hand, B_i decreases with increasing volume fractions of EG-211 phase. The similar dependence is found also for addition of NEG-211 phase, although B_i is maximum and reaches 7 T even at 80 K at 7 mol% of NEG-211 phase. From the changes in J_{cp} and B_i with addition of 211 phases, it is found that there is no large difference in the effect of addition between EG-211 and NEG-211 phases. Such variations with addition of 211 phases are completely opposite to the cases of Y-123 and Nd-123.

However, it can be said from the above results that there exists a negative correlation between $J_{\rm cp}$ and $B_{\rm i}$. This strongly suggests that the pinning mechanisms which determine $J_{\rm cp}$ and $B_{\rm i}$ are different. The exceptional behavior of the specimen of 7 mol% of NEG-211 might be caused by smaller fraction of dispersed 211 phase than the nominal composition.

In order to explain responsible pinning centers and corresponding pinning mechanisms for the new behavior of $J_{\rm cp}$ and $B_{\rm i}$, a discussion is necessary, which is as follows:

Substituted regions/oxygen deficient regions which exist naturally are known to cause the peak effect in Nd-123 and Y-123. In the present measurements, however, the peak effect became pronounced by increasing the volume fractions of 211 phases, whereas substituted regions or oxygen deficient regions are not controlled. Hence, these defects are considered not to be responsible for the increase in $J_{\rm cp}$.

It is known that 211 particles act as strong pinning centers. In fact the critical current density of about 3×10^8 A/m^2 can be expected at 1 T for the specimen with 10 mol% 211 partciles from the relationship between $J_{\rm c}$ and surface area of 211 phase particles from the result in [26]. However, this will be reduced to about 2×10^8 A/m^2 at 2.5 T, and hence, the obserbed critical current density of about 6×10^8 A/m^2 at the peak field can not be explained by the mechanism of 211 phase particles. In addition, the pinning mechanisms of normal 211 particles is the condensation energy interaction with the pinning force which decreases monotonically with increasing magnetic field. Hence, the pinning by 211 particles does not directly play the role in the peak effect. Another possibility of the peak effect by 211 particles is the order-disorder transition of the flux lines, i.e., a transitional change of flux lines from the ordered state with a low $J_{\rm c}$ to the disordered state with a high $J_{\rm c}$. The enhancement of the pinning efficiency at the peak effect is considered to be caused by the softening of elasticity of flux lines. In the previous investigation on the peak effect and the irreversibility field for a single crystal $NdBa_2Cu_3O_y$ superconductor[23], the observed results were analysed using the flux creep-flow model[24], and the number of flux line in the flux bundle (g^2), the most probable value (A_m) and the distribution width (σ^2) of the pinning strength were determined so that the theoretical results fit well with the experimental results. It was found that g^2 decreased at the peak field, suggesting a softening of the flux line lattice, because g^2 was proportional to the shear modulus C_{66}. Such a softening of C_{66} is a direct proof of the order-disorder transition. For occurrence of the order-disorder transition, however, the pinning centers should be sufficiently small that an appreciable variation in the pinning efficiency could be realized by a slight deformation of flux lines. This requires a pinning potential quickly varying in space. On the other hand, the size of 211 particles is much larger than the flux lines spacing. Hence, the 211 particles do not seem to contribute to the peak effect also from a softening of flux lines for the disorder transition.

Another candidate for the pinning centers responsible for the peak effect is nano-lamellas. These defects are considered to be in the normal state, since the superconductivity in high-temperature superconductors is quite sensitive to the chemical composition. In fact, superconductivity has not been reported in single phase specimens with such a NEG-rich chemical composition. Thus, the pinning mechanism of nano-lamellas can not be the field-induced one but the usual condensation energy interaction, the strength of which decreases monotonically with increasing field. The observed peak effect can be explained only by the order-disorder transition of flux lines with the pinning by nano-lamella structures. This seems to be possible, since their size is sufficiently small that those can provide a sharp variation in the flux pinning strength by a slight rearrangement of flux lines. It should be noted that any kind of pinning centers or pinning mechanisms can contribute to the order-disorder transition. That is, the corresponding pinning centers are substituted regions or oxygen deficiencies in Nd-123 and Y-123 with repulsive pinning (kinetic energy interaction) and nano-lamella structures in the present system with attractive pinning (condensation energy interaction). But the important point is that the size of pinning centers should be sufficiently small.

The decrease in B_i with increasing volume fraction of 211 phase is incompatible with the flux pinning by 211 particles themselves. One possible explanation may be a deterioration of the Bose-glass state of flux lines. That is, the high irreversibility field attained by the stabilization of flux lines in the Bose-glass state by the c-axis correlated nano-lamellas [27] seems to be degraded by disturbance by 211 particles. However, the enhancement of the peak critical current density by the 211 particles addition suggests a rather stronger stabilization of the Bose-glass state by the addition, resulting in a contradiction with experiments, if the scenario of the stabilization of the Bose-glass state is believed. In addition, such an enhancement of the pinning strength can be simply explained by the correlated pinning independently of the Bose-glass state.

Since the pinning interaction of 211 particles is not so weak, there should exist some contribution to the irreversibility field from 211 particles. Hence, the degradation in the irreversibility can only be attributed to deterioration of some superconducting property. Such a deterioration seems to be caused by the proximity effect as in the substituted regions in Nd-123[25]. In the present specimens, since the number density of nano-lamella structures is very high, the proximity effect between thin superconducting region and layered nano-lamella structures seems to be significant, resulting in deterioration of the superconducting property such as H_c2 in the superconducting matrix. This explains the decrease in B_i through the decrease in the upper critical field, when the fraction of the added 211 phase is high.

Thus, it can be concluded that the dominant pinning centers which determine J_cp and B_i in NEG-123 superconductor are not the 211 particles but the nano-lamella structures. It is speculated that the peak effect is caused by the order-disorder transition of flux lines with the assistance of the pinning by nano-lamella structures and that the deterioration of B_i with the volume fraction of the 211 phase is caused by the proximity effect of thin nano-lamella structures.

4. Summary

In order to study the pinning mechanism of peak effect and irreversibility field in NEG-123 system, the variations in $J_{\rm cp}$ and $B_{\rm i}$ with volume fractions of EG-211 and NEG-211 phases were measured for ($Nd_{0.33}$, $Eu_{0.38}$, $Gd_{0.28}$)$Ba_2Cu_3O_y$ superconductor. It was found that $J_{\rm cp}$ increases but $B_{\rm i}$ decreases with increasing volume fraction both of EG-211 and NEG-211 phase and there was no large difference between these phases. This result shows a negative correlation between $J_{\rm cp}$ and $B_{\rm i}$, suggesting that the mechanisms which determine $J_{\rm cp}$ and $B_{\rm i}$ are different. Based on the discussion on these results the following conclusions are obtained:

1. Since the peak effect became pronounced by addition of 211 phase, the change can be attributed to the 211 particles themselves or to new defects nucleated by the addition. For the following reasons, it can be concluded that the 211 particles do not contribute for the peak effect: (a)the pinning force of 211 particles based on the pinning mechanisms of the condensation energy interaction decreases monotonically with increasing magnetic field and (b)since the size of 211 particles is much larger than the flux lines spacing, those can not contribute to the order-disorder transition of flux lines.
2. The pinning mechanism of the nano-lamella structures is the usual condensation energy interaction with the pinning strength which decreases monotonically with increasing field. Hence, these defects do not directly contribute to the peak effect. On the other hand, their size is sufficiently small that those can contribute to the peak effect with the aid of the order-disorder transition.
3. Since the irreversibility field is deteriorated by the addition of 211 phase, it can be concluded that the irreversibility field is not determined by the flux pinning of 211 particles. The decrease in $B_{\rm i}$ with the increasing fraction of 211 phase is considered

to be caused by the proximity effect between thin superconducting region and high density of nano-lamellas.

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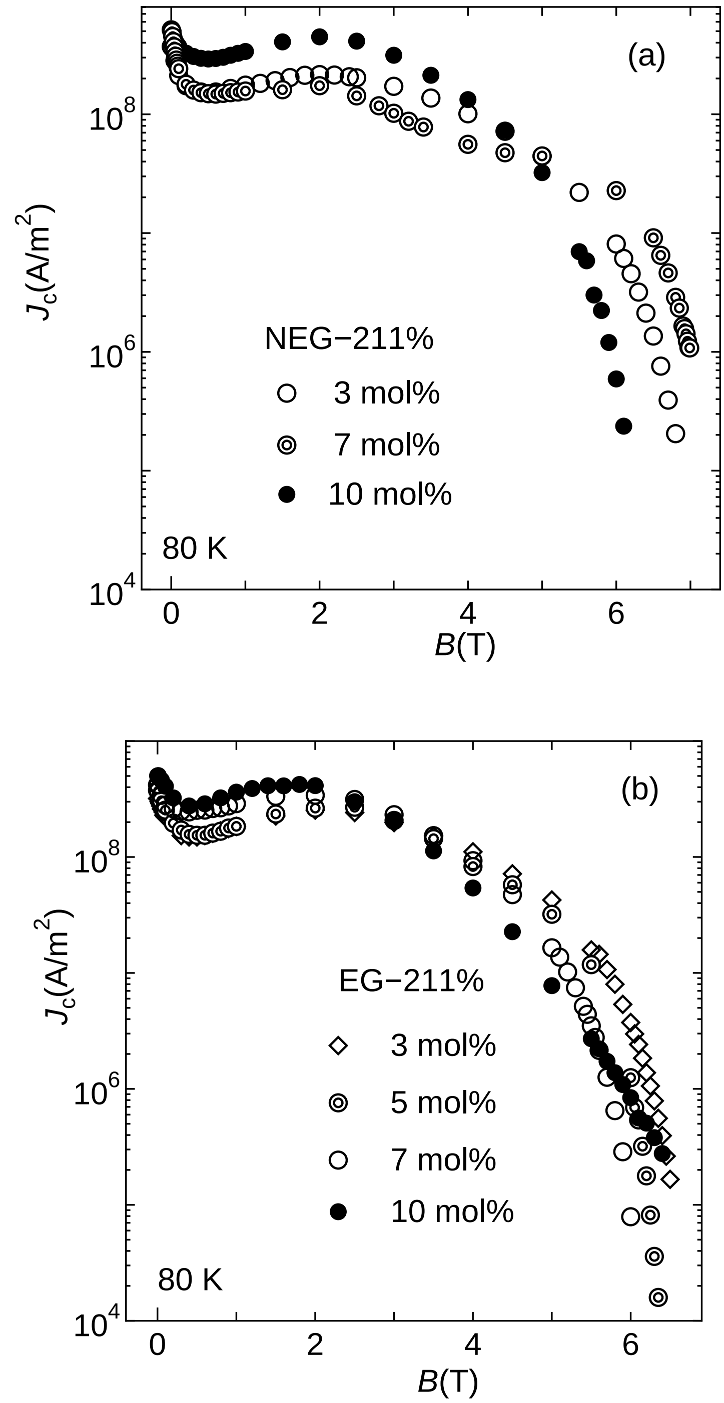

Fig.1 Field dependence critical current density J_c in NEG-123 system with different volume fraction of (a) NEG211 and (b) EG211 particles at 80 K.

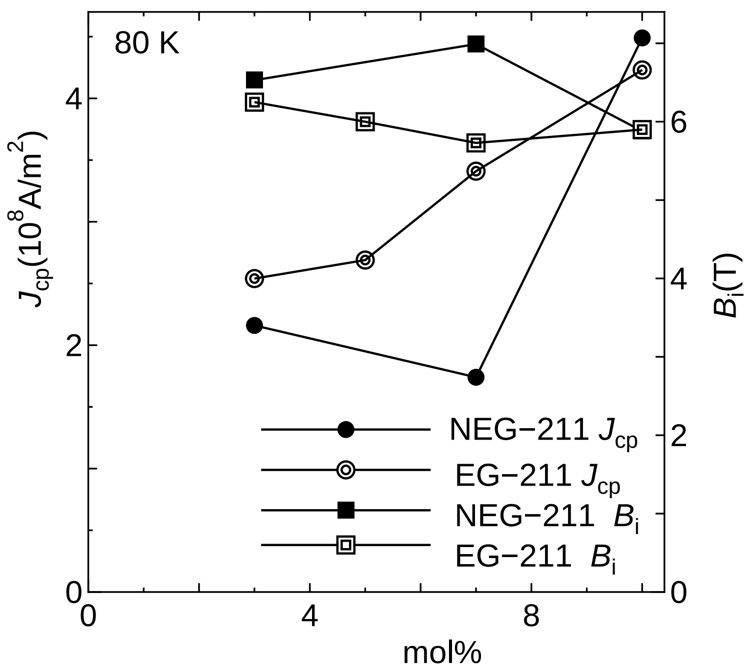

Fig.2 Variation of the peak critical current density J_{cp} and irreversibility field B_i with different volume fraction of 211 particles at 80 K.

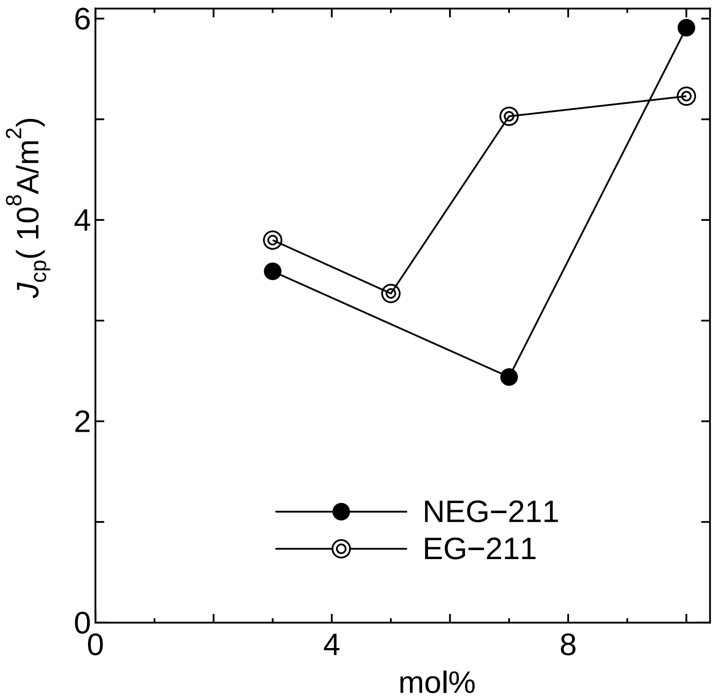

Fig.3 Variation of the peak critical current density $J_{\rm cp}$ and with different volume fraction of 211 particles at 77.3 K.