Enhancement of Critical Current Densities in (Ba,K)Fe$_2$As$_2$ by 320 MeV Au Irradiation in Single Crystals and by High-Pressure Sintering in Powder-in-Tube Wires

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We demonstrate a large enhancement of critical current density ($J_c$) up to $1.0 \times 10^6$ A/cm$^2$ at 5 K under self-field in (Ba,K)Fe$_2$As$_2$ single crystals by irradiating 320 MeV Au ions. With the very promising potential of this material in mind, we have fabricated a (Ba,K)Fe$_2$As$_2$ superconducting wire through a powder-in-tube method combined with the hot isostatic pressing technique, whose effectiveness has been proven in industrial Bi2223 tapes. The $J_c$ in the wire at 4.2 K has reached $37 \, \text{kA/cm}^2$ under self-field and $3.0 \, \text{kA/cm}^2$ at 90 kOe. Magneto-optical imaging of the wire confirmed the large intergranular $J_c$ in the wire core. © 2013 The Japan Society of Applied Physics

The discovery of superconductivity in the iron-based superconductor (IBS)$^1$ has elicited great excitement and generated enormous amount of research activities. Many IBSs exhibit high critical temperatures ($T_c$'s)$^2-5$ and large upper critical fields ($H_{c2}$'s)$^6-10$ and relatively small anisotropies compared with cuprate superconductors.$^8,11,12$ They also show very high critical current density ($J_c$) up to $10^6$ A/cm$^2$ in high magnetic fields. This is almost the same level as $J_c$ of NbTi or Nb$_3$Sn which is practically applied for high-field magnets, so they are very promising for high-field applications.$^{13}$ e.g., high-performance superconducting wires and tapes. IBSs have attracted great interest in the fields of condensed matter physics and superconducting applications, based on such outstanding characteristics.

One of the fundamental questions in IBSs is how much $J_c$ can be enhanced. The $J_c$ is one of the extrinsic parameters that can be enhanced by introducing artificial defects. Irradiations of energetic particles are the well-known methods for this purpose. In particular, protons$^{14}$ and heavy ions$^{15}$ have been proven to be very effective in enhancing $J_c$ in single crystals of cuprate superconductors. These methods may not be practical, but it is well known that enhancements of $J_c$ by similar defect structures have been implemented in a more economical way in an industrial coated conductor of YBa$_2$Cu$_3$O$_y$. $^{16,17}$ Similar effects of particle irradiations have also been demonstrated in single crystals of IBSs with heavy ions$^{17-19}$ and protons.$^{20,21}$ Among various IBSs, 122-type materials, such as BaFe$_2$As$_2$, have been widely studied for the availability of their high-quality single crystals. Among them, (Ba,K)Fe$_2$As$_2$ with $T_c$ up to 38 K is reported to have the largest $J_c$ ($2 \times 10^6$ A/cm$^2$ at 5 K and 2 kOe) and it is enhanced up to $5 \times 10^6$ A/cm$^2$ at 5 K and 2 kOe after irradiating 1.4 GeV Pb ions with a dose-equivalent matching field of $B_{eq} = 210$ kOe.$^{18}$ By optimizing the condition of irradiation, further enhancement of $J_c$ in IBSs is expected, and the achieved value and behavior of $J_c$ can be a target for practical superconducting wires of IBSs.

In order to bring out the potential of remarkably high $J_c$ of IBSs for practical applications, several studies about superconducting wires have been performed. The best candidates for applications in IBSs are the 122-type. They have small anisotropies of 2–3, moderate $T_c$’s, large $H_{c2}$’s, and large $J_c$’s.$^{22}$ The $H_{c2}$ for (Ba,K)Fe$_2$As$_2$ exceeds those in both MgB$_2$ and Nb$_3$Sn at low temperatures, so (Ba,K)Fe$_2$As$_2$ attracts great technological interest.$^{10,22}$ In order to use 122-type materials for superconducting wires, growth processes of polycrystalline samples and wire fabrication techniques should be improved. Some studies for IBS wires suggest that weak links between superconducting grains are the main reason for the low transport $J_c$. The performance of the wires has been much improved by several methods, the combination of several times of cold press and hot press,$^{23}$ addition of Ag, Pb, and Sn,$^{24-29}$ or by texturing tapes,$^{30,28,29}$ and so on.

In order to develop the superconducting wire of IBSs for practical applications, $J_c$ of the core material should be high and weak links between superconducting grains should be suppressed by optimizing the fabrication process. In this paper, we try to answer two fundamental questions in IBSs; (1) how much can $J_c$ be enhanced ultimately, and (2) what is the most important process in making high-performance superconducting wires of IBSs. Answers to these two questions can be a good target and guiding principle for making practical wires of IBSs. For this purpose we show the significant enhancement of $J_c$ both in single crystal and in superconducting wires of (Ba,K)Fe$_2$As$_2$ as a promising materials with high $J_c$. We demonstrate a strong enhancement of $J_c$ up to $1.0 \times 10^7$ A/cm$^2$ at 5 K under self-field in (Ba,K)Fe$_2$As$_2$ single crystals by 320 MeV Au irradiation. We also demonstrate the enhancement of transport $J_c$ up to $3.7 \times 10^7$ A/cm$^2$ at 4.2 K under self-field in (Ba,K)Fe$_2$As$_2$ powder-in-tube (PIT) wires by using only the hot isostatic pressing (HIP) technique without other special treatments. The HIP process has been proven to be effective in enhancing $J_c$ of industrial Bi2223 tapes.$^{30,31}$

Single crystals of (Ba,K)Fe$_2$As$_2$ were synthesized by the self-flux method. We used Ba pieces, K ingots, and FeAs powder as starting materials. FeAs was prepared by placing stoichiometric amounts of As pieces and Fe powder in an evacuated quartz tube and reacting them at 700 °C for 40 h after heating them at 500 °C for 10 h. A mixture with a ratio of Ba : K : FeAs = 0.6 : 0.44 : 4 was placed in an alumina crucible and sealed in a stainless steel container$^{32}$ in a nitrogen-filled glove box, and heated for 1 h at 1100 °C followed by cooling to 900 °C at a rate of 5 °C/h. 320 MeV Au ions were irradiated into (Ba,K)Fe$_2$As$_2$ along the c-axis using the tandem accelerator in JAEA to create columnar defects at dose-equivalent matching fields of $B_{eq}$ = 10 and 80 kG. (Ba,K)Fe$_2$As$_2$ superconducting wires were fabricated.
by the ex situ PIT method. Polycrystalline samples of (Ba,K)Fe$_2$As$_2$ were prepared by the solid-state reaction. A mixture with a ratio of Ba : K : FeAs = 0.6 : 0.44 : 2 was sealed in the same way as single-crystal synthesis. It was heated for 5 h at 1,100 °C after soaking for 5 h at 600 °C. The prepared (Ba,K)Fe$_2$As$_2$ precursor was ground into a fine powder in the nitrogen-filled glove box, and was filled into a silver tube with an outer diameter of 4.5 mm and an inner diameter of 3 mm, then cold drawn into a square wire with a diagonal dimension of about 1.2 mm. After cutting the drawn wire into ~3 cm pieces, each piece was put into a 1/8” copper tube and redrawn into the same size as the silver sheathed wire. Both ends of the wire were sealed using an arc furnace, and they were heated for 4 h at 600 °C in Ar atmosphere under a pressure of 120 MPa.

The phase identification of the sample was carried out by means of powder X-ray diffraction (MAC Science M18XHF) with Cu Kα radiation. Bulk magnetization was measured using a superconducting quantum interference device magnetometer (Quantum Design MPMS-5XL). Current–voltage ($I$–$V$) measurements up to 90 kOe were performed by the four-probe method. $I$–$V$ measurements were performed in a bath-type cryostat. The composition of (Ba,K)Fe$_2$As$_2$ is characterized by scanning electron microscopy and energy dispersive X-ray spectroscopy (SEM–EDX). For magneto-optical (MO) imaging, an iron-garnet indicator film is placed in direct contact with the sample, and the whole assembly was attached to the cold finger of a He-flow cryostat (Oxford Instruments Microstat-HR). MO images are acquired by using a cooled CCD camera with 12-bit resolution (Hamamatsu ORCA-ER).

The $J_c$ characteristics of the pristine and 320 MeV Au irradiated (Ba,K)Fe$_2$As$_2$ single crystals are summarized in Figs. 1(a) and 1(b). $J_c$'s are evaluated from the irreversible magnetization by using the extended Bean model. The $J_c$ value of the pristine crystal is about $2.0 \times 10^6$ A/cm$^2$ at 5 K under self-field, which is comparable to that reported in Ref. 18. It shows a significant increase up to $1.0 \times 10^7$ A/cm$^2$ by 320 MeV Au irradiation with a dose of $B_B = 80$ kOe. This $J_c$ value is twice larger than that for 1.4 GeV Pb-irradiated crystal ($B_B = 210$ kOe), and one of the largest values so far reported in IBS single crystals. A similar particle energy dependence of $J_c$ has been observed in Au-irradiated Ba(Fe,Co)$_2$As$_2$. One of the possibilities is that defects are splayed at lower ion energies by the sample stopping power, which was observed in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$. Splayed structure is more effective to suppress the motion of vortices than only parallel structure since magnetic relaxation via half-loop excitation becomes slower by forced entanglement of vortices, resulting in enhancement of $J_c$. The remarkably high $J_c$ value barely depends on the magnetic field, and it sustains the value of $7.0 \times 10^6$ A/cm$^2$ even at 40 kOe. The irradiated (Ba,K)Fe$_2$As$_2$ crystal also retains the high $J_c$ value of $1.0 \times 10^6$ A/cm$^2$ even at 30 K below 35 kOe. The extremely high $J_c$ and its retention at high magnetic fields and high temperatures indicate that (Ba,K)Fe$_2$As$_2$ is one of the promising candidates for high-field applications.

With the very promising characteristics of (Ba,K)Fe$_2$As$_2$ single crystals described above, we fabricated and characterized PIT wires of (Ba,K)Fe$_2$As$_2$. First, polycrystalline precursor was characterized. Figure 2 shows the X-ray diffraction pattern of the precursor. The diffraction pattern has strong peaks of 122 phase indicating that the reaction to 122 phases is complete. Peaks from impurity phases such as FeAs are not detected. For this precursor, the formation of superconducting phase was confirmed by the signal of $J_c$ as FeAs are not detected. For this precursor, the formation of superconducting phase was confirmed by the signal of $J_c$ as FeAs are not detected. The onset $T_c$ of the precursor is about 38 K, which indicates that its composition is almost optimal.

Figure 3(a) shows the $E$–$J$ characteristics of the HIP-processed PIT wire of (Ba,K)Fe$_2$As$_2$ under different magnetic fields at 4.2 K. Here, we adopt $E = 1 \mu$V/cm as a criterion to define transport $J_c$ for the $E$–$J$ curves. The transport $J_c$ at 4.2 K has reached 37 kA/cm$^2$ under self-field and 3.0 kA/cm$^2$ at 90 kOe. $J_c$’s in 122 superconducting wires or tapes from recent publications are also plotted in Fig. 3(b). The self-field $J_c$ in the HIP-processed (Ba,K)Fe$_2$As$_2$ wire is several times larger than that of (Ba,K)Fe$_2$As$_2$ PIT wires, which are sintered at ambient pressure. Furthermore, it is almost 10 times larger at high magnetic fields. The obtained value of $J_c$ is roughly the same as the recently reported $J_c$'s in Sn-added (Ba,K)Fe$_2$As$_2$ textured tapes, which is 20–30% of $J_c$ in Sn-added (Sr,K)Fe$_2$As$_2$ textured tape, and is 20–30% of $J_c$ in the HIP-processed wire employing mechanical alloying with a low-temperature process. These results strongly indicate that our HIP
treatment for the PIT wire largely contributed to the improvement of \( J_c \).

The HIP-processed wire was further characterized by several physical measurements. The temperature dependence of the magnetic moment for the short segment of the wire was measured at \( H = 10 \text{ Oe} \) along the wire direction, as shown in Fig. 4(a). The onset of the main body of diamagnetism of the wire is slightly reduced to 30 K, but the shielding volume fraction is roughly 100%. We performed the SEM–EDX analysis for the wire. The analyzed potassium content \( x \), detected from the whole area of the cross section of wires of about \( 200 \times 200 \mu \text{m}^2 \), was 0.43 for \( \text{Ba}_1.63\text{K}_0.37\text{Fe}_2\text{As}_2 \). These values are almost the same as the nominal composition. However, from local area analysis around 10 \( \mu \text{m}^2 \), the grains that have rich or poor potassium contents, and small amounts of impurities such as FeAs and BaAs were also detected. The distribution of potassium content in 122 phases and impurities may be the main reason for the reduction of \( T_c \). To suppress the content of impurities and to obtain optimal 122 phases in wires, fabrication and synthesis processes should be improved.

In IBSs, vortices at grain boundaries are generally pinned more weakly than vortices in the grains, thus the grain boundaries become barriers for current flow. To investigate the quality of the grain boundaries, we performed MO measurements on the PIT wire. Figure 4(b) is an optical image of the transverse cross section in the HIP-processed wire. The area of the core part was approximately 0.0005 cm\(^2\), detected from the whole area of the cross section of wires of about \( 200 \times 200 \mu \text{m}^2 \), was 0.43 for \( \text{Ba}_1.63\text{K}_0.37\text{Fe}_2\text{As}_2 \). The thickness of this sample along the wire axis in the optical image is roughly 300 \( \mu \text{m} \). A higher magnification optical image of the core is shown in Fig. 4(c). Small grains of 122 phases with sizes less than 20 \( \mu \text{m} \) can be identified. No void is observed, which proves the high density of the wire core. Figures 4(d) and 4(e) depict MO images of the transverse cross section of the wire core in the remanent state after applying an 800 Oe field along the wire axis for 0.25 s which was subsequently...
Research Project by the Japan Society for the Promotion of Science (JSPS) for Young Scientists (B) (No. 24740238) and the Japan–China Bilateral Joint Project (No. 2374024). It is clear that weak links across grain boundaries are responsible for the trapping of large magnetic flux. We have fabricated a superconducting wire by the PIT method (2013) 025003.

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