Fabrication Process and Pressure Dependence of Critical Current Density in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ Superconducting HIP Wires

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Abstract. We demonstrate the fabrication of (Ba,K)Fe$_2$As$_2$ superconducting wires through a powder-in-tube (PIT) method and hot isostatic press (HIP) technique. Several fabrication processes with different sintering conditions of pressure and time were examined. The measurements of magnetization, $I$-$V$ characteristics, filling factor, and Vickers hardness clarified that the improvement of fabrication process and sintering condition is effective for the enhancement of critical temperature $T_c$, which leads to the enhancement of critical current density $J_c$ in HIP round wire. Wires which are fabricated in different processes show significantly different $T_c$ and $J_c$. The core density, which can be enhanced by sintering at higher pressures, is the main key factor for enhancement of $J_c$ in HIP wires, similar to pressed tapes. Changing the sintering time also slightly affect $J_c$.

1. Introduction
Iron-based superconductors (IBSs) are attractive candidates for future high-field applications of superconductors because they exhibit high critical temperature, $T_c$, large upper critical field, $H_{c2}$, and relatively low anisotropy compared with cuprate superconductors [1,2]. Among IBSs, $AE_{1-x}K_xFe_2As_2$ ($AE =$ Ba, Sr) compounds have been extensively studied as promising candidate for application, because high critical current density, $J_c$, is realized in superconducting wires and tapes using $AE_{1-x}K_xFe_2As_2$ [3-14]. In these studies, it is demonstrated that uniaxial pressing for tapes [3-7] and hot isostatic pressing (HIP) for round wires [8-14] are effective methods to enhance $J_c$. Pressing superconducting wires and tapes using these methods is an effective method for enhancing the density and eliminating weak links between superconducting grains in the core of wires and tapes. It is expected that the values of $J_c$ in these wires and tapes strongly depend on their fabrication and sintering processes. In the case of tapes, effects of fabrication process and pressing were revealed. For example, Gao et al. demonstrated that, final thickness of the pressed tapes and hardness of the core of the tapes strongly affect the $J_c$ in the tapes [3]. They showed positive correlation between the $J_c$ and...
hardness of the tape core [3]. The hardness, which is related to the core density, is controlled by the pressure of cold press. On the other hand, in the case of HIP round wire, how different fabrication processes and pressures affect the \( J_c \) have not been studied in detail. Reported HIP wires have been processed at high pressure of 100-200 MPa for 4-20 h [8,14]. However, sintering pressure and time dependence of \( J_c \) in the HIP wire have not been systematically studied yet. There is only one report where pressure dependence of \( J_c \) in the HIP wire was studied [12]. So the sintering condition for the HIP wire still has rooms for improvements. Furthermore, we have recently reported that drawing or rolling process for the wire has strong influence on \( J_c \) in the HIP wire [15]. Round wires drawn using dies with circular holes before groove-rolling (“drawn”) have higher \( J_c \) than those fabricated using only a groove roller with square grooves (“rolled”). The effect of different fabrication process of the wire on \( J_c \) should also be studied.

In this work, we first investigate how different mechanical deformation processes affect \( J_c \) characteristics of HIP wires by comparing properties of wires that are fabricated by different processes. Second, we systematically investigate how the sintering conditions such as pressure and time affect the \( J_c \) performance of HIP wires, by comparing wires prepared at different pressures and sintered for various times.

2. Experiment

Superconducting wires of \((\text{Ba,K})\text{Fe}_2\text{As}_2\) were fabricated by ex-situ powder-in-tube (PIT) method. Polycrystalline powders of \(\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2\) were prepared by the solid-state reaction. Used polycrystalline powder is the same as that used in the previous work [14], and details of powder preparation are described in ref. [14]. Ground powder was filled in a silver tube with an outer diameter of 4.5 mm and an inner diameter 3 mm. Here, three different fabrication processes were chosen. First, Ag tubes filled with powder were cold drawn into a round shape with a diameter \(\sim 1.2 \text{ mm}\) using dies with circular holes (“drawn”). Second, they were fabricated using only a groove roller with square grooves (“rolled”). Third, they were swaged using a rotary swaging machine (“swaged”). Obtained three kinds of wires were cut into short pieces. They are put into a 1/8 inch copper tube and redrawn into a square shape with a groove roller down to a diagonal dimension of 1.2 mm. After the drawing process, both ends of the wire were sealed by using an arc furnace. The sealed wires were sintered using the HIP technique. All wires were heated up to 700°C in argon atmosphere, and sintered under different pressures of 0.1-170 MPa and for 0.5-12 h. HIP process under the highest pressure 170 MPa were performed at National Institutes for Quantum and Radiological Science and Technology (QST), and others were performed at the University of Tokyo. In order to evaluate transport \( J_c \), a DC electric current up to 150 A was delivered through the wire at a ramping rate of 50-100 A/min. Measurements were performed in liquid helium to minimize the effect of Joule heating at the current leads. The critical current measurements in high magnetic fields were carried out by using the 15T-SM at High Field Laboratory for Superconducting Materials, IMR, Tohoku University. Current–voltage \((I-V)\) characteristics up to 140 kOe were measured by the four-probe method with solder for contacts. Bulk magnetization was measured to evaluate \( T_c \) and magnetic \( J_c \) by a superconducting quantum interference device (SQUID) magnetometer (MPMS-5XL, Quantum Design). Magnetic \( J_c \) is evaluated using the extended Bean model. The filling factor of the core was estimated from the volume and weight of cut and polished core of the wire. Vickers hardness, Hv, was measured on the polished surface of the wire core.

3. Results and discussion

Transport \( J_c \) and magnetic \( J_c \) of HIP wires sintered at various conditions are estimated. As shown in figure 1(a), “drawn” wire which was sintered at 170 MPa for 4 h shows the highest value of \( J_c \). At the self-field, transport \( J_c \) exceeds \( 10^5 \text{ A/cm}^2 \) which is the target value of \( J_c \) for application. At high field of 100 kOe, transport \( J_c \) reaches at \( 3.0 \times 10^4 \text{ A/cm}^2 \). Magnetic \( J_c \) also shows similar value and field dependence. This \( J_c \) value is larger than the previous world record of IBS round wire [14] and close to the value of the largest \( J_c \), which was achieved recently [15]. The difference between this wire and the
previous one [14] is just the fabrication process. This wire was “drawn” and previous one is “rolled”. Such a fabrication method dependence of \( J_c \) is also observed in other wires. In figure 1(a) magnetic field dependence of magnetic \( J_c \) in “drawn”, “swaged”, and “rolled” wire are described. These three wires were sintered in the same condition at 9 MPa for 0.5 h. Magnetic \( J_c \) in “drawn” or “swaged” wire is larger than that in “rolled” wire. One of the possible reasons why fabrication process affects the \( J_c \) is the different degrees of degradation of superconducting core because of different wire fabrications. We have shown that superconducting properties of the wire are degraded during the wire fabrication process, and recovered during the sintering process [11]. It is possible that the degree of the degradation depends on the fabrication method. As shown in figure 1(b), \( T_c \) of “drawn”, “swaged”, and “rolled” HIP wire are different to each other. In “rolled” wire with the smallest \( J_c \), \( T_c \) is also the lowest. The main factor for the suppression \( T_c \) is degradation of polycrystalline powders in the core of the wire during fabrication process [11]. Degraded superconducting property is partly recovered during sintering. The degree of recovery in the three kinds of wires must be the same because sintering temperature, pressure, and time is the same. So the degree of degradation should be different and they affect the \( J_c \) and \( T_c \) in the wire. It should also be noted that the highest \( T_c \) in these wires are realized in the “drawn” wire sintered at 170 MPa for 4 h. For recovery of superconducting properties, higher sintering pressure and longer sintering time are more effective. The other possible reason is the differences of grain orientations and texturing, which is reported in pressed tape [4,5]. Details of this possible reason are discussed in our former publication [15].

Next, we focus on the effects of sintering pressure on physical properties in the HIP wire. To improve weak links between grains, higher density is advantageous. Sintering at higher pressure should be effective for that. As shown in figure 2(a), the filling factor shows clear positive correlation with the sintering pressure. At 170 MPa, the filling factor reaches 97%. Related to this correlation, Hv also shows positive correlation with the sintering pressure shown in figure 2(b). Increases in both the filling factor and Hv suggest the improvement of links in grains. Actually, \( J_c \) also shows a similar sintering pressure dependence as shown in figure 2(c). Hv dependence of \( J_c \) in HIP wires is summarized in figure 2(d). The positive correlation between \( J_c \) and Hv is observed. It is clearly confirmed for the first time systematically in IBS round wire that high core density plays a key role in achieving the practical level of \( J_c \), as discussed in IBS pressed tape [4]. It is worth noting that the effect of fabrication process on \( J_c \) cannot be ignored, although the effect of sintering pressure is more significant. As shown in figure 2(c), when the sintering pressure is about 9 MPa, \( J_c \) values of the wires are not the same. As discussed above, \( J_c \) is enhanced not only by improvements of weak link between

**Figure 1.** (a) Magnetic field dependence of transport \( J_c \) and magnetic \( J_c \) in the (Ba,K)Fe\(_2\)As\(_2\) HIP wires processed in different conditions. (b) Temperature dependence of normalized magnetization at 10 Oe for the (Ba,K)Fe\(_2\)As\(_2\) HIP wires.
grains using high pressure but by controlling core states such as superconducting properties and orientation of grains with changing fabrication process. For further enhancement of $J_c$, both changing sintering pressure and fabrication process should be effective.

We also investigated the effect of sintering time on the superconducting properties of the HIP wire. Sintering time dependences of $J_c$ in “swaged” HIP wire is shown in figure 3. Fabrication processes and sintering temperatures and pressures are the same in these wires. Compared with the sintering pressure dependence on $J_c$ as shown figure 2, the effect of sintering time on $J_c$ is small. However, slight enhancement is found. For example, $J_c$ of $7.8 \times 10^4$ A/cm$^2$ at sintering time of 0.5 h is enhanced to $10.1 \times 10^4$ A/cm$^2$ at sintering time of 8 h. Longer sintering time may be preferred for an increase of $J_c$ in HIP wire [15]. Enhancement of $J_c$ with increasing sintering time may be caused by higher sintering degree and improvement of superconducting properties and grain connections. This result suggests that further enhancement of $J_c$ is expected by making sintering time longer. It should be noted that the optimal condition of sintering time should depend on sintering temperature. For example, Weiss et al. fabricated HIP wire at lower temperature of 600°C and longer time of 20 h [8]. Systematic analyses of sintering time dependence of $J_c$ at different temperature has not been reported. Optimization of sintering time and temperature will realize certain degree of enhancement of $J_c$ in the HIP round wire.
4. Summary
We have fabricated (Ba,K)Fe$_2$As$_2$ superconducting wires through a PIT method and HIP technique. Several fabrication processes such as “drawing”, “swaging”, and “rolling”, and different sintering conditions of pressure and time were examined. The measurements of magnetization, $I$-$V$ characteristics clarified that fabrication process significantly affects the enhancement of $J_c$ in the HIP round wire. The estimation of filling factor of the core and $H_v$ in several wires revealed that the core density plays a key role for the improvement of weak links between grains in the core, and it can be controlled by changing sintering pressure. This relation is systematically evaluated and clarified for the first time in IBS round wires, similar to the pressed tapes. Changing the sintering time also slightly affect $J_c$.

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