Fabrication of (Ba,Na)Fe₂As₂ round wires using HIP process

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Abstract. We have fabricated round wires of Ba₀.₆Na₀.₄Fe₂As₂ using powder-in-tube technique and hot isostatic press (HIP) treatment. Transport \( J_c \) was measured at 4.2 K of two Ba₀.₆Na₀.₄Fe₂As₂ round wires with Ag/Cu double sheath which were processed at 700 °C for 4 h at 175 MPa. Transport \( J_c \) of one of the wires reached 76 kA/cm² and 24 kA/cm² at self-field and 100 kOe, respectively, while \( J_c \) of another wire is slightly larger at low fields. It means that \( J_c \) of Ba₀.₆Na₀.₄Fe₂As₂ wire is resilient against large magnetic field, which demonstrates very promising characteristics of this material. Detailed characterizations of wires using X-ray diffraction and compositional mapping by EDX are also presented. We also explore the better condition for synthesis of Ba₀.₆Na₀.₄Fe₂As₂ by changing the reaction temperature.

1. Introduction

The discovery of iron-based superconductors (IBSs) in 2008 has prompted a great interest in their application potentiality [1]. Soon after the discovery, several types of high temperature superconductors in iron pnictides were discovered. IBSs are high-temperature superconductors that have high critical temperature \( T_c \), high critical magnetic field \( H_{c2} \), small anisotropy \( \gamma \), and large critical current density \( J_c \) under high magnetic fields. Among them, superconducting wires and tapes have been fabricated mainly by using (AE,K)Fe₂As₂ materials (AE = Ba or Sr), so called ‘122-type’ [2-8], because of their high \( H_{c2} \) [9, 10] and low \( \gamma \) (~2) [10, 11], as well as their high \( T_c \) (~36-38 K) [12, 13]. They are expected to be put into practical applications such as wires for high-field magnets. In particular, superconducting tapes and wires using K-doped 122 type ((Ba,K)Fe₂As₂ and (Sr,K)Fe₂As₂) have been extensively studied. Recently, we have reported fabrication and characterization of (Sr,Na)Fe₂As₂ tapes, and demonstrated that \( J_c \) in this system is 47 kA/cm² even at 100 kOe at 4.2 K [14]. However, it became clear that segregation of Na in the wire core causes a problem in this system. Reduction of Na content is one way to achieve better performance. However, since antiferromagnetic phase is extended to \( x = 0.5 \) in \( \text{Sr}_1x\text{Na}_x\text{Fe}_2\text{As}_2 \) [15], pursuing underdoped sample is not a good option. So, we chose (Ba,Na)Fe As as an alternative, where...
2. Experiments

First, we synthesized polycrystalline samples of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$. Synthesis processes were conducted in an argon filled glove box. Starting materials (BaAs, NaAs, Fe$_2$As) were ground into powder with an agate mortar and pestle. Starting materials were synthesized by a method similar to that of (Sr,Na)Fe$_2$As$_2$ [17]. Polycrystalline samples were synthesized at 770 °C for 15 h in a stainless steel pipe, where pellets of the starting materials were placed. Then, the reacted materials were ground into powder, pelletized, and reacted at 770 °C for 15 h again. The prepared polycrystalline materials were ground into powder. The ground powder was tightly packed into a silver tube with outer and inner diameters of 4.5 and 3.0 mm, respectively. Some of the tubes were groove-rolled into a wire with 1.3 x 1.3 mm$^2$ cross section, and others were swaged using a rotary swager into the diameter of 1.3 mm. After these processes, both ends of the wire were sealed using an arc furnace. The sealed wires were sintered using the hot isostatic press (HIP) technique. Wires were heated for 4 h at 700 °C in an argon atmosphere under different pressures up to 175 MPa. HIP processes were performed at National Institutes for Quantum and Radiological Science and Technology. We also synthesized polycrystalline samples of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ at different reaction temperatures (800 °C, 820 °C, and 850 °C).

Magnetic measurements were conducted using a commercial SQUID magnetometer (MPMS-XL5, Quantum Design). Transport $J_c$ was measured at 4.2 K by the standard four-probe method in magnetic fields up to 140 kOe. Powder X-ray diffraction (XRD) measurements were conducted using Cu-Kα radiation (Smartlab, Rigaku). SEM images of the core were taken using SEM-EDX (S-4300, Hitachi High-Technologies equipped with EMAX x-act, HORIBA).

3. Results and discussion

3.1. Transport $J_c$ measurements at 4.2 K in magnetic fields up to 140 kOe

Figure 1 compares transport $J_c$ at 4.2 K of two Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ round wires with Ag/Cu double sheath which were processed at 700 °C for 4 h at 175 MPa. In the drawing process, wire #1 was swaged, and wire #2 was rolled. Transport $J_c$ of wire #1 is 76 kA/cm$^2$ and 24 kA/cm$^2$ at self-field and 100 kOe, respectively. On the other hand, $J_c$ at 100 kOe of wire #2 is lower than that of #1, while self-field $J_c$ is slightly larger. Even at a high magnetic field of 100 kOe, $J_c$ of wire #1 is reduced to only one third of the self-field value.

![Figure 1](image_url)

Figure 1. Magnetic field dependence of transport $J_c$ at 4.2 K of two Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ HIP round wires processed at 700 °C and 175 MPa for 4 h.
### 3.2. Magnetic susceptibility measurements

Figure 2 shows $M$-$T$ measurements of $\text{Ba}_{0.6}\text{Na}_{0.4}\text{Fe}_2\text{As}_2$ polycrystalline powders sintered once and twice at 770 °C, and the HIP wire. Comparing polycrystals sintered once and twice, the transition of twice sintered became a little sharper. Although $T_c$ of the HIP wire became lower than that of polycrystals, the transition became much sharper.

![Normalized magnetization at $H = 5$ Oe as a function of temperature of $\text{Ba}_{0.6}\text{Na}_{0.4}\text{Fe}_2\text{As}_2$ polycrystalline powders sintered once and twice at 770 °C, and the HIP wire.](image)

Figure 3 shows normalized magnetization as a function of temperature of $\text{Ba}_{0.6}\text{Na}_{0.4}\text{Fe}_2\text{As}_2$ polycrystalline powders sintered at different temperatures. By increasing the sintering temperature from 770 °C, the transition became sharper as in samples sintered at 800 °C and 820 °C. However, when the sintering temperature reached 850 °C, $T_c$ dropped markedly and the transition became broader.

![Normalized magnetization at $H = 5$ Oe as a function of temperature of $\text{Ba}_{0.6}\text{Na}_{0.4}\text{Fe}_2\text{As}_2$ polycrystalline powders sintered twice at different temperatures (770, 800, 820, and 850 °C).](image)
3.3. X-ray diffraction analyses and SEM images

Figure 4 shows X-ray diffraction patterns of the HIP wire and polycrystalline powders (sintered at 770 °C) of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$. Compared to the polycrystalline powders, peaks of the HIP wire are sharper. Although peaks of impurities (Fe$_2$As and FeAs) can be seen in powder sample, the amount of impurities in the HIP wire is less. SEM images in Fig. 5 shows Fe$_2$As impurities exist in the core of the HIP wire, and some parts are Ba rich. In addition, Na-rich region can also be identified as in the case of (Sr,Na)Fe$_2$As$_2$ [14].

![X-ray diffraction patterns](image)

**Figure 4.** X-ray diffraction patterns of the HIP wire and polycrystalline powders of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ prepared at 770 °C. Reference data is calculated as explained in ref. [18].

![SEM image and EDX mappings](image)

**Figure 5.** (a) SEM image of the cross section of the HIP wire. (b) Compositional mappings of the HIP wire by EDX.

In Figure 6, polycrystals sintered at 770 °C had most impurities of FeAs and Fe$_2$As. Polycrystals sintered at 800 and 820 °C had less impurities, and polycrystals sintered at 850 °C had least impurities. As the sintering temperature increases, the amount of impurities became less.
Figure 6. X-ray diffraction patterns of polycrystalline powders of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ sintered at 770, 800, 820, and 850 °C. Reference data is calculated as explained in ref. [18].

Although there are still impurities in the HIP wire using polycrystalline sample sintered at 770 °C, $J_c$ was as large as that in (Ba,K)Fe$_2$As$_2$. By comparing data in Fig. 6 and Fig. 3, it is clear that the polycrystals sintered at 800 and 820 °C had less impurities and sharper transition. Although $T_c$ of the polycrystals sintered at 850 °C is lower, the content of impurities is minimum. Compositional mapping of the sample sintered at 850 °C by EDX shows that the average content of Na is 0.3 as compared with the initial value of 0.4. We speculate that this reduction of average Na content is the origin of the suppression of $T_c$ in sample sintered at 850 °C. We thus conclude that the optimal temperature for the preparation of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ is between 800 °C and 820 °C. $J_c$ of HIP wires using polycrystals sintered at the optimal temperature is expected to be high.

4. Summary
We fabricated round wires of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ using powder-in-tube technique and HIP treatment. The maximum transport $J_c$ at 4.2 K and 100 kOe of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ wire sintered at 770 °C reaches 24 kA/cm$^2$. It is remarkable that $J_c$ in this wire is reduced to only one third of the self-field value at 100 kOe. Although there are still impurities in the HIP wire using polycrystalline powder sample sintered at 770 °C and it is not optimal, $J_c$ is comparable to that in (Ba,K)Fe$_2$As$_2$ wire. From $M$-$T$ and XRD measurements, we propose that the optimal temperature for the preparation of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ is between 800 °C and 820 °C.

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