Fabrication of (Ba,Na)Fe$_2$As$_2$ round wires and tapes using HIP process

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Abstract. We fabricated round wires and tapes of (Ba,Na)Fe$_2$As$_2$ using hot isostatic press (HIP) process, and evaluated their critical current density ($J_c$). Polycrystalline powders were synthesized by two methods by mixing raw materials in a planetary ball mill or by using pre-synthesized precursors (BaAs, NaAs, Fe$_2$As). Transport $J_c$ of best wire reached 95 kA/cm$^2$ under self-field and 40 kA/cm$^2$ at 100 kOe. We also fabricated HIP tapes using the same powder and discuss their detailed characterizations. In particular, X-ray diffraction is extensively applied to the evaluation of the degree of texturing of the tape, and discuss its relationship with $J_c$.

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 IBSs were discovered. IBSs are superconductors with 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)Fe$_2$As$_2$ 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-types ((Ba,K)Fe$_2$As$_2$ and (Sr,K)Fe$_2$As$_2$) have been extensively studied. Recently, we have reported fabrication and characterizations of (Sr,Na)Fe$_2$As$_2$ tapes, and demonstrated that $J_c$ in this system is 47 kA/cm$^2$ even at 100 kOe at 4.2 K [14]. Very recently, $J_c$ in (Ba,Na)Fe$_2$As$_2$ HIP round wire processed using hot isostatic press (HIP) process reaches 150 and 20 kA/cm$^2$ at self-
field and 100 kOe, respectively [15-17]. These values are more than fifty percent of the largest $J_c$ in (Ba,K)Fe$_2$As$_2$ round wires, demonstrating the excellent performance of (Ba,Na)Fe$_2$As$_2$ wires.

In this study, we prepared high-quality polycrystalline powders of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ by two methods, and fabricated HIP round wires and tapes. The maximum transport $J_c$ at 4.2 K and 100 kOe of (Ba,Na)Fe$_2$As$_2$ wire reached 40 kA/cm$^2$, which is the largest among all IBS round wires. The maximum magnetic $J_c$ at 4.2 K and 40 kOe of (Ba,Na)Fe$_2$As$_2$ tape reached 66 kA/cm$^2$.

2. Experiments
Superconducting wires of (Ba,Na)Fe$_2$As$_2$ were fabricated by the ex situ powder-in-tube (PIT) method. In the present study, polycrystalline powders of (Ba,Na)Fe$_2$As$_2$ were prepared by two ways of solid-state reaction. In the first process, Ba pieces, Na ingots, Fe powder, and As pieces were used as starting materials. In order to compensate the loss of elements, the starting mixture contained 15% excess Na and 5% excess As. After filing Ba pieces and crushing As pieces, starting materials were mixed in an argon atmosphere more than 10 h using a ball-milling machine and densely packed into a niobium tube. The niobium tube was then put into a stainless steel tube and sealed in argon-filled glove box for heat treatment at 800, 850, and 900 °C for 30 h. It was then ground into powder using an agate mortar in argon-filled glove box. We define the powder, which was heated at 850 °C, as the #1 powder. In the second process, starting materials (BaAs, NaAs, Fe$_2$As) synthesized by a method reported in our work [17] were ground into powder with an agate mortar and pestle. The prepared precursors were weighed to a composition of Ba$_3$As : Na$_3$As : Fe$_2$As = 0.6 : (0.4 + α) : 1, and thoroughly mixed. Here, α (= 0.1) is additional NaAs content to compensate the loss of Na and As during the synthesis. Polycrystalline samples were synthesized at 820 °C for 24 h in an argon-filled stainless steel tube. The obtained powder is designated as the #2 powder.

The obtained two kinds powders were ground and filled into silver tubes (o.d.: 4.5 mm, i.d.: 3 mm). Ag tubes filled with #1 or #2 powders were cold drawn using dies with circular holes, or swaged using a rotary swaging machine, respectively. Both wires were formed into a round shape with a diameter of ~1.2 mm. After cutting them into short pieces, one of the pieces was put into 1/8 inch copper tube and redrawn into a square shape with a groove roller down to a diagonal dimension of 1.2 mm. Furthermore, a part of the wire was deformed into a tape form with 0.3 - 0.5 mm thickness. After the drawing process, both ends of the wire or tape were sealed using an arc welder. The sealed wires and tapes were sintered using the HIP technique. Some wires were heated for 0.5 h at 650-740 °C in an argon atmosphere under the pressures of 9 MPa. Other wires and tapes were heated for 4 h at 700 °C in an argon atmosphere under pressures of 175 MPa for wires and 200 MPa for tapes.

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).

3. Results and discussion

3.1 Polycrystalline powders synthesized by two methods
Figure 1(a) shows $M$-$T$ measurements of both #1 and #2 Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ polycrystalline powders. Although the drop of the magnetization of the #1 powder is slightly sharper than that of the #2 powder, reasonably sharp transitions indicate that the qualities of both powders are higher than that of previous study [15-17]. Figure 1(b) shows XRD patterns of #1 and #2 powders. Peaks of (Ba,Na)Fe$_2$As$_2$ phase are strong and there are no peaks of impurities such as FeAs or Fe$_2$As.
polycrystalline powders #1 and #2. (b) X-ray diffraction patterns of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ polycrystalline powders #1 and #2.

3.2 HIP round wires
We fabricated HIP round wires using the #2 powder at 9 MPa for 0.5 h at differing temperatures (600 ~ 740 °C) to optimize the heating temperature during the HIP process. Magnetic $J_c$ as a function of magnetic field for wires sintered at different temperatures is shown in Fig. 2(a). Figure 2 (b) shows magnetic $J_c$ under self-field and at 40 kOe of HIP wires as functions of the sintering temperature. Although $J_c$ becomes higher as heating temperature becomes high up to 700 °C, it declines above 700 °C. So, we can conclude that the optimal sintering temperature during the HIP process for Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ Ag/Cu double sheath wire is 700 °C.

Figure 2. (a) Magnetic field dependence of magnetic $J_c$ at 4.2 K of HIP round wires sintered at 9 MPa for 0.5 h between 600 °C and 740 °C. (b) Magnetic $J_c$ under self-field and at 40 kOe as functions of sintering temperature during the HIP process.

Figure 3 shows the 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 processed at 700 °C for 4 h at 175 MPa. The transport $J_c$ of the wire #1 is 95 kA/cm$^2$ and 40 kA/cm$^2$ under self-field and at 100 kOe, respectively. It is noteworthy that the value of $J_c$ at 100 kOe of 40 kA/cm$^2$ is larger than that of the best (Ba,K)Fe$_2$As$_2$ HIP round wire [18]. On the other hand, $J_c$ at
100 kOe of the wire #2 is lower than that of #1, while $J_c$ under self-field is slightly larger. More detailed characterizations of these wires will be reported in a separate publication [19].

![Graph](image1)

**Figure 3.** Magnetic field dependence of transport $J_c$ at 4.2 K for the Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ HIP wires #1 and #2.

### 3.3 HIP tapes

Figure 4 shows the magnetic field dependence of magnetic $J_c$ of #1 and #2 HIP tapes with different thicknesses. The magnetic $J_c$ of both #1 and #2 tapes becomes large when the thickness of the tape becomes thinner. Among five kinds of tapes, the magnetic $J_c$ of the #1 tape with 0.3 mm thickness is the largest ($510$ kA/cm$^2$) under self-field, while $J_c$ of the #2 tape of 0.3 mm thickness is the largest ($66$ kA/cm$^2$) at 40 kOe. It should be noted that the magnetic $J_c$ under self-field of our HIP tapes is much larger than that of cold pressed (Ba,Na)Fe$_2$As$_2$ tape ($50$ kA/cm$^2$ under self-field) [20]. Even at a field of 40 kOe, the magnetic $J_c$ of our HIP tape is larger than the cold pressed tape ($43$ kA/cm$^2$).

![Graph](image2)

**Figure 4.** Magnetic field dependence of the magnetic $J_c$ of #1 and #2 HIP tapes with different thicknesses.
Figure 5 (a) and (b) show X-ray diffraction patterns of #1 and #2 tapes, respectively, with different thicknesses. It is obvious that all the tapes contain very few impurities. As a measure of texturing of the tape, we define $r$ as the ratio of the intensity of (103) and (002) peaks, $r = I_{(002)}/I_{(103)}$. $r$ is 0.27 and 0.34 for the #1 tape with thicknesses of 0.3 mm and 0.4 mm, respectively. For the #2 tape, $r$ is 0.68, 0.56, and 0.44 with the thicknesses of 0.3, 0.4, and 0.5 mm, respectively.

Figure 5. X-ray diffraction patterns of (a) #1 and (b) #2 HIP tapes with different thicknesses.

For the #1 tape, however, the relation between $r$ and $J_c$ is not consistent and $r$ is smaller than that of the #2 tape. In spite of better performance of the #1 HIP wire, the #1 tape may have some problems which is not seen in the #2 tape. For the #2 tape, $J_c$ becomes systematically larger as $r$ becomes larger. On the other hand, for the #1 tape, $r$ is smaller than that for the #2 tape with the same thickness. In addition, $r$ becomes smaller in the thinner tape with larger magnetic $J_c$ in the #1 tapes. It clearly indicates that there is another factor governing the value of $J_c$ in the tape. Further studies are necessary including the shape and orientation of the core in the tape, which is known to be nonideal.

4. Summary
We fabricated round wires and tapes of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ using powder-in-tube technique and HIP treatment. We succeeded in preparing high-quality polycrystalline powders of Ba$_{0.6}$Na$_{0.4}$Fe$_2$As$_2$ by two methods. We optimized sintering temperature during the HIP process of the wire. The maximum transport $J_c$ at 4.2 K and 100 kOe of (Ba,Na)Fe$_2$As$_2$ wire reached 40 kA/cm$^2$, which is the largest
among all IBS round wires. The maximum magnetic $J_c$ at 4.2 K and 40 kOe of (Ba,Na)Fe$_2$As$_2$ tape reached 66 kA/cm$^2$.

References
[1] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[2] Ma Y W, Gao Z S, Qi Y P, Zhang X P, Wang L, Zhang Z Y and Wang D L 2009 Physica C 469 651
[3] Togano K, Matsumoto A and Kumakura H 2011 Appl. Phys. Express 4 043101
[4] Pyon S, Tsuchiya Y, Inoue H, Kajitani H, Koizumi N, Awaji S, Watanabe K and Tamegai T 2014 Supercond. Sci. Technol. 27 095002
[5] Pyon S, Yamasaki Y, Kajitani H, Koizumi N, Tsuchiya Y, Awaji S, Watanabe K and Tamegai T 2015 Supercond. Sci. Technol. 28 125014
[6] Weiss J D, Tarantini C, Jiang J, Kametani F, Polyanskii A A, Larbalestier D C and Hellstrom E 2012 Nat. Mater. 11 682
[7] Pyon S, Suwa T, Park A, Kajitani H, Koizumi N, Tsuchiya Y, Awaji S, Watanabe K and Tamegai T 2016 Supercond. Sci. Technol. 29 115002
[8] Tamegai T, Suwa T, Pyon S, Kajitani H, Takano K, Koizumi N, Awaji S and Watanabe K 2017 IOP Conf. Ser. Mater. Sci. Eng. 279 012028
[9] Putti M et al. 2010 Supercond. Sci. Technol. 23 034003
[10] Wang Z S, Luo H Q, Ren C and Wen H H 2008 Phys. Rev. B 78 140501R
[11] Yuan H Q, Singleton J, Balakirev F F, Baily S A, Chen G F, Luo J L and Wang N L 2009 Nature 457 565
[12] Rotter M M, Tegel M, Johrendt D, Schellenberg I, Hermes W and Pöttgen R 2008 Phys. Rev. B 78 020503
[13] Sasmal K, Lv B, Lorenz B, Guloy A M, Chen F, Xue Y Y and Chu C W 2008 Phys. Rev. Lett. 101 107007
[14] Suwa T, Pyon S, Tamegai T, and Awaji S 2018 Appl. Phys. Express 11 063101
[15] Tamegai T, Suwa T, Miyawaki D, Pyon S, Takano K, Kajitani H, Koizumi N, and Awaji S 2019 IEEE Trans. Appl. Supercond. 29 7300605
[16] Miyawaki D, Pyon S, Tamegai T, Awaji T, Takano K, Kajitani H, and Koizumi N 2019 J. Phys.: Conf. Ser. 1293 012043.
[17] Miyawaki D, Pyon S, Suwa T, Takano K, Kajitani H, Koizumi N, Awaji S and Tamegai T to be published in Physica C
[18] Pyon S, Suwa T, Tamegai T, Takano K, Kajitani H, Koizumi N, Awaji S, Zhou N, and Shi Z X 2018 Supercond. Sci. Technol. 31 055016.
[19] Pyon S, Miyawaki D, Tamegai T, Awaji S, Kito H, Ishida S, Yoshida Y submitted to Supercond. Sci. Technol.
[20] Imai S, Itou S, Ishida S, Tsuchiya T, Iyo A, Eisaki H, Matsuzaki K, Nishio T, and Yoshida Y 2019 Sci. Rep. 9 13064.