Comparison of J_c Characteristics among (Ba,Na)Fe_2As_2 Wires and Tapes Fabricated by Different Methods

T. Tamegai, R. Sakagami, T. Ito, K Takinoiri, A. Kikuchi, and M. Yamamoto

***Abstract*—In the present study, critical current density (J_c) characteristics and uniformity of (Ba,Na)Fe_2As_2 wires and tapes fabricated by different methods and sheath materials are compared. Impurity-free high-quality polycrystalline powders were prepared by two different methods; direct reaction method and precursor method. By optimizing the firing temperature and holding time of precursors, we have succeeded in obtaining high-quality precursor powders. In the case of wire drawing, a rotary swager with an automatic wire feeder is used to prevent sausaging. To further deform it into a tape form, cold press was used. In addition to the J_c characteristics of (Ba,Na)Fe_2As_2 wire and tapes, X-ray characterizations for the evaluation of texturing and uniformity are presented. Finally, a fine and long (Ba,Na)Fe_2As_2 wire is fabricated using Monel as an outer sheath material, and its characteristics are presented.**

***Index Terms*—Iron-based superconductors, (Ba,Na)Fe_2As_2, wires and tapes, critical current density**

I. INTRODUCTION

IRON-based superconductors (IBSs) have fascinating properties, such as high upper critical field (H_{c2}), modest critical temperature (T_c), and small electromagnetic anisotropy (γ), all of which are beneficial to utilize them under high magnetic fields. The fact that the critical grain boundary angle is much larger than that of cuprate superconductors [1] opens up a possibility that superconducting wire with reasonable critical current density (J_c) can be fabricated by an economic way of powder-in-tube (PIT) method using polycrystalline materials. Actually, several attempts have been performed to fabricate superconducting wires using PIT method in the very early stage of study [2], [3]. Since then, among various kinds of IBSs, 122-type materials represented by (AE,A)Fe_2As_2 (AE :Ba, Sr, A: K, Na) have been studied extensively to explore the possibility to be used as superconducting wires [4]. The practical level of critical current density J_c = 100 kA/cm^2 at 4.2 K under 100 kOe has been achieved by (Ba,K)Fe_2As_2 tapes prepared by uniaxial pressing [5]. Compared with tapes, round wires should have higher flexibility for the construction of various forms of high-field magnets. In addition, focusing on the excellent magnetic field dependence of J_c in (Ba,Na)Fe_2As_2 compared with that in (Ba,K)Fe_2As_2 despite its slightly lower T_c, we started fabrication of (Ba,Na)Fe_2As_2 round wires [6]. It leads to steady improvement of J_c in (Ba,A)Fe_2As_2 round wires, and now J_c of 71 and 49 kA/cm^2 at 4.2 K under 100 kOe have been achieved for (Ba,Na)Fe_2As_2 [7] and (Ba,K)Fe_2As_2 [8] round wires, respectively. On the other hand, improvements of J_c characteristics of Na-doped 122-type materials only started by the study on (Sr,Na)Fe_2As_2 tape in 2014 [9]. It was followed by the study of (Ba,Na)Fe_2As_2 tape, achieving J_c = 40 kA/cm^2 at 4.2 K under 40 kOe [10]. We have also fabricated $Sr_{0.45}Na_{0.55}Fe_2As_2$ tape using AgSn-sheath, and achieved J_c = 65 kA/cm^2 at 4.2 K under 100 kOe [11]. However, the largest J_c of (Ba,Na)Fe_2As_2 tape is still limited to J_c = 44 kA/cm^2 at 4.2 K under 100 kOe [12]. So, more careful refinement of powder preparation and tape fabrication is required. Another issue for the fabrication of superconducting wire is the non-uniformity of wires and tapes along the lengths of them, which is called sausaging effect. In the present study, we attempted to suppress the sausaging effect by using harder sheath material of Sn-doped Ag ($Ag_{1-x}Sn_x$). So far, most of wires and tapes have been fabricated from a powder prepared by directly reacting elemental materials. However, anticipating larger-scale production of IBS materials, usage of arsenides seem to be beneficial due to weaker sensitivity to oxygen and moisture of arsenides, and their easy handling. So, we also attempt to fabricate (Ba,Na)Fe_2As_2 round wire using arsenide precursors. Other important issues for the superconducting wire is the thermal stability and minimizing various kinds of losses, such as AC loss and coupling loss. All these requirements can be fulfilled by fabricating fine superconducting wires, and bundle them after twisting them. So, prepared fine (Ba,Na)Fe_2As_2 wire using harder an outer sheath material of Monel.

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II. EXPERIMENTS

In the present study, superconducting wires and tapes of (Ba,Na)Fe_2As_2 were fabricated by *ex situ* powder-in-tube (PIT) method. In general, fabrication of IBS wires and tapes consists of three steps; (1) preparation of polycrystalline powder, (2) drawing of wires and tapes in metal sheaths, and (3) final heat treatments. Polycrystalline powders were prepared by two different methods of solid-state reaction. In the first method, raw elemental materials of Ba pieces, Na ingots, Fe powder, and As pieces were directly reacted after thoroughly mixing them in a planetary ball milling machine. We call this method as “direct reaction method”. In the second method, arsenides precursors such as Fe_2As, BaAs, and NaAs, were used to synthesize $(Ba,Na)Fe_2As_2$ powder. We call this method as “precursor method”. The mixed precursors and raw materials were heated at different temperatures between 800ºC and 890ºC for ~30 h. So far, the largest J_c has been achieved by using powder prepared by direct reaction method [7]. Details of firing conditions will be shown in each experimental section. The final reaction of precursors is done in Nb tube, which is sealed in a quartz tube.

The two kinds of reacted materials were pulverized and filled into silver tubes with outer and inner diameters of 4.5 mm and 3 mm, respectively. They were then either swaged using a rotary swaging machine or cold-drawn using dies with circular holes. The obtained wires with diameters 1.35– 1.5 mm were cut into short pieces, and one of them was inserted into 1/8 inch copper tube and redrawn down to diameters of 1.8 mm for tapes and 1.0 mm for wires. In the case of tapes, the final wire is rolled several times down to a final thickness of 500 μm. Before applying HIP treatment, both ends of the wire and tape were sealed using an arc welder. HIP treatments were done by heating for 4 h at 700 ◦C in an argon atmosphere under high pressures of 175 MPa. The transport J_c up to 140 kOe was measured using 15 T-SM at the High Field Laboratory for Superconducting Materials, IMR, Tohoku University by passing DC electric current up to 100 A through the wire in liquid He to minimize the Joule heating at the current leads. The bulk magnetization of short pieces of wires was measured for the characterization of T_c and magnetic J_c by a superconducting quantum interference device magnetometer (SQUID, MPMS-5XL, Quantum Design). Powder X-ray diffraction (XRD) measurements with Cu-Kα radiation were conducted using a powder X-ray diffractometer (Smartlab, Rigaku) on polycrystalline powders and the core of the wires to identify impurity phases and evaluate the degree of texturing. The wire core was carefully examined by a scanning electron microscope (S-4300, Hitachi High Technologies), and elemental mappings were conducted using energy-dispersive x-ray spectroscopy (EDX) with EMAX x-act (HORIBA).

III. Results and Discussion

A. Improvements of $(Ba,Na)Fe_2As_2$ Tapes

Cu/Ag-sheathed wires drawn down to a diameter of 1.8 mm were made into tapes using a roller to the final thickness of 500 μm. The optical micrograph of the tape is shown as an inset for Fig. 1. The transport J_c as a function of magnetic field up to 140 kOe is shown in Fig. 1. Also shown is the results of ref. [10]. The J_c value at 4.2 K under 1 kOe for this tape reaches 140 kA/cm^2, and it sustains a high value of 48 kA/cm^2 even at 100 kOe. Obviously, J_c of our HIP processed tape has J_c more than 50% larger than that reported in ref. [10]. However, J_c value of 48 kA/cm^2 is lower than that for the largest J_c of round wires for the same material. Although a certain degree of under estimation of J_c is possible due to the choice of cross section, the value can be still lower than that for the best round wire. X-ray diffraction of this tape shows that the texturing parameter of r defined by $r = I((002)/I(103)$ is only 0.5, which is small for cold pressed tapes. We speculate that the reason for this low value of J_c can be due to combined effects of weaker texturing due to insufficient rolling and partly due to insufficient intergranular coupling of grains used for this tape.

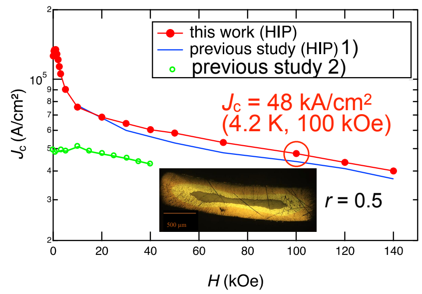

Fig. 1 Magnetic field dependence of transport J_c for HIP-processed $(Ba,Na)Fe_2As_2$ tape at 4.2 K. The same value for the cold-pressed tape reported in ref. [10] is shown with green circles.

B. $(Ba,Na)Fe_2As$ Wires with Ag/Sn Sheath

Effectiveness of HIP process is expected to be enhanced by using harder sheath material. So, here we report the effect of Sn addition into Ag sheath, which is in direct contact with the core material. Another important aspect of the usage of harder sheath material is the suppression of sausaging effect during the wire-drawing and/or swaging process. Figures 2(a)-(d) show the X-ray tomography image longitudinal cross section of series of $(Ba,Na)Fe_2As_2$ round wires with different amount of Sn in the Ag sheath. The degree of sausaging for these wires has been analyzed by using the auto-correlation function of the cross-sectional area along the length of the wire. It confirms our conjecture that the typical scale of sausaging is reduced by increasing the Sn content in Ag sheath. Fig. 3 shows the transport J_c as a function of magnetic field for these Ag/Sn-sheathed wires. Unexpectedly, values of J_c for all these wires are not as good as we expected. At 4.2 K and 100 kOe, J_c for the Ag-sheathed material is 18 kA/cm^2 and it decreased with increasing Sn content in the sheath. Since X-ray diffraction pattern of the powder used for these wires contains

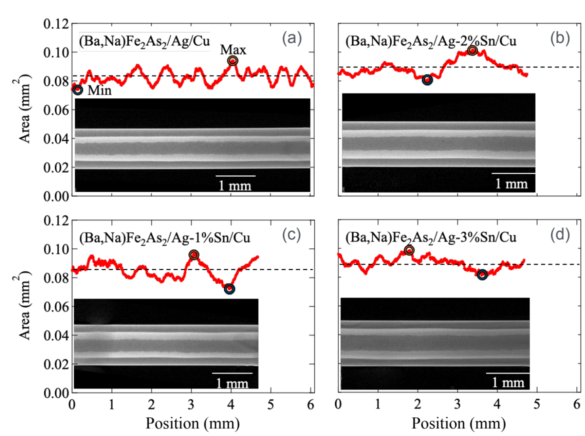

Fig. 2 Variation of cross sectional area of $Ag_{1-x}Sn_x$/Cu-sheathed $(Ba,Na)Fe_2As_2$ wire with (a) 0%, (b) 1%, (c) 2%, and (d) 3% Sn in Ag. Insets in each figure is X-ray tomography image of each wire.

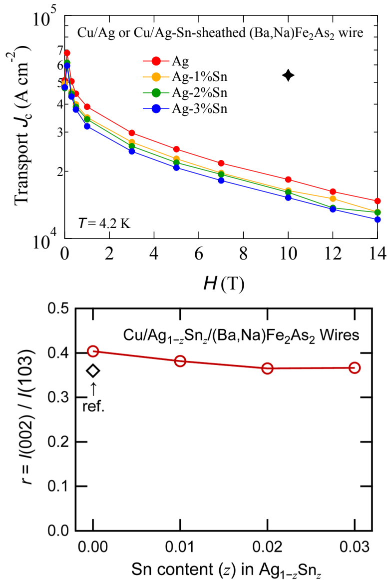

Fig. 3 (a) Magnetic field dependence of transport J_c and (b) texturing parameter as a function of Sn content in Ag/Sn-sheathed (Ba,Na)Fe_2As_2 wires.

negligible amount of impurities, the reason for the low J_c value can be associated with weak-link nature of gains. Despite low value of J_c, the texturing parameter r is large as shown in Fig. 3(b), which is even larger than the r value for the wire that achieved the largest J_c [10]. It turns out that the chemical composition of the superconducting core of all these wires are more that 0.4 as shown in Table I-I possibly due to the contribution from the excess NaAs. Simulations of the texturing parameter for (Ba,Na)Fe_2As_2 with different amounts of Na demonstrate that it can be enhanced by increasing the Na content. So, a lesson learned from these experiments is that we need to take care of the chemical composition when we compare the texturing parameter for wires fabricated in different processes.

TABLE I
CHEMICAL COMPOSITION OF AG/SN-SHEATHED (BA,NA)FE_2AS_2 TAPES.

x	Ba	Na	Fe	As
0	0.597	0.439	2.04	1.925
0.01	0.58	0.433	2.043	1.95
0.02	0.597	0.43	2.034	1.94
0.03	0.60	0.44	2.03	1.930

C. Improvements (Ba,Na)Fe_2As_2 Wires using Precursors

Here, we re-examined all the processes for the wire fabrication using precursors. In order to make sure the chemical composition of the prepared powders of precursors, we thoroughly examined the preparation process of all precursor arsenides such as Fe_2As, BaAs, and NaAs. The XRD patterns of arsenide powders prepared in the present experiments are shown in Figs. 4(a)-(c). As for Fe_2As, the powder obtained after the first firing (800ºC, 40 h) consists mainly of Fe_2As with ~5% of FeAs impurity. Since there are no volatile components, and the inner wall of the quartz tube used for the synthesis was clean, we judge that only one firing is enough to obtain high-quality of Fe_2As. On the other hand, in the case of BaAs fired at 700ºC for 20 h, since 1:1 ratio of BaAs does not exists, the obtained powder is a mixture of different barium arsenides with general formula of $BaAs_x$, and it is not easy to judge whether the obtained powder is good to be used for the synthesis of (Ba,Na)Fe_2As_2. Hence, we fired the same material twice after pulverizing carefully, and sealing it in another quartz tube. The XRD pattern after the second firing is shown at the bottom of Fig. 4(b). Obviously, the pattern changed significantly from that after the 1st firing. So, we fired it for the third time, and took another XRD pattern. The XRD pattern after the 3rd firing (not shown) is almost identical to that after the 2nd firing. Hence, we judge that firing twice is enough to obtain stable mixture of $BaAs_x$. Fig. 4(c) shows the XRD pattern of NaAs after the 1st and 2nd firing (800ºC, 24 h). Since these patterns are almost identical, we judge that firing only once is enough to obtain pure NaAs.

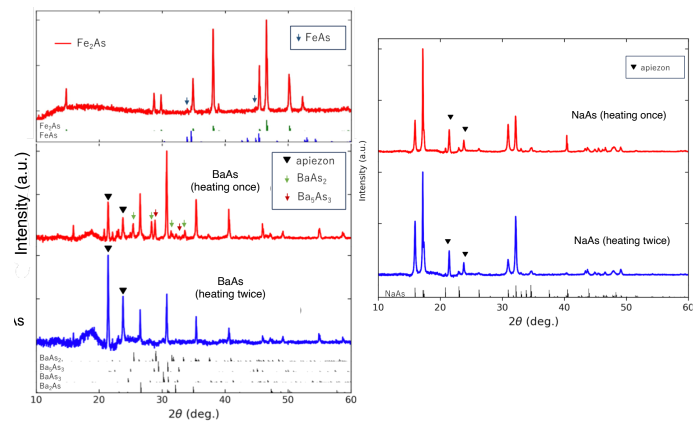

Fig. 4 XRD patterns of (a) Fe_2As, (b) BaAs (1st firing: top, and 2nd firing: bottom, and (c) NaAs.

By using well-characterized arsenide powders, we optimized the synthesis temperature by changing the holding temperature, and found out that the impurity contents are minimum when the reaction temperature is between 820ºC and 850ºC. After all these preparation, we started the syntheses of larger amount of (Ba,Na)Fe_2As_2 powder using arsenides precursors. In doing so, anticipating the loss of Na during the synthesis, we started from the composition of Fe_2As:BaAs:NaAs = 1:0.6:0.5, amd prepared three kinds of (Ba,Na)Fe_2As_2 powders using the same starting arsenides.

Figure 5(a) shows XRD patterns of three kinds of powders synthesized at 835ºC or 850ºC. Except for powder #2, there are very few impurity phases. Temperature dependence of normalized magnetization for three kinds of powders are shown in Fig. 5(b) in open symbols. Both powder b and c show sharp superconducting transition starting from 35 K. Temperature dependence of magnetization for three kinds of wires using each power are shown also in Fig. 5(b) in solid symbols. All three wires show onset of diamagnetic response at ~33-32 K, among which wire C shows the sharpest

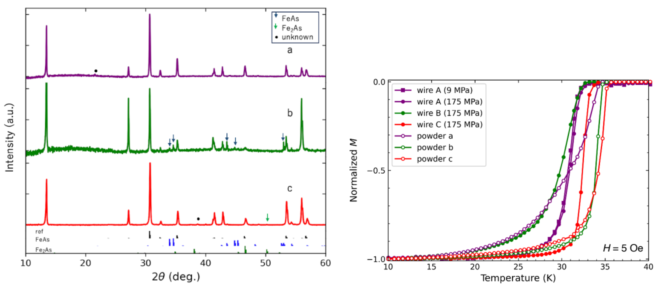

Fig. 5 (a) XRD patterns of three kinds of (Ba,Na)Fe_2As_2 powders. (b) Temperature dependence of normalized magnetization for three kinds of powders and four kinds of wires processed by HIP.

transition close to 33 K. Figure 6 summarizes magnetic field dependence of the transport J_c for all three wires. We also plot the data for the J_c - H curve that recorded the largest J_c at 100 kOe prepared by direct reaction method [7] and another showing the largest J_c among all wires prepared starting from precursors [13]. All wires show very similar J_c - H curves, and only the scales for J_c are different from wire to wire. In the present experiment, wire *C* prepared from powder *c* exhibited the largest transport J_c of 41 kA/cm^2 among all round wires prepared from precursors. The value of J_c for wire *A* is unexpectedly low, lower than that of wire *C* by a factor of 7. The reason for such a big discrepancy in J_c is not clear at the present stage.

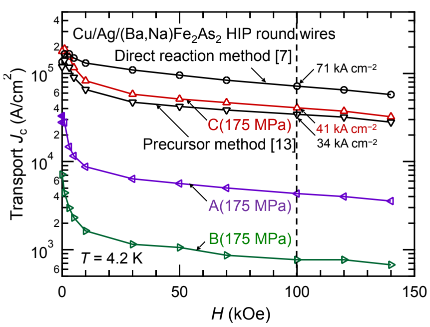

Fig. 6 Magnetic field dependence of J_c for three (Ba,Na)Fe_2As_2 wires A, B, and C, prepared by using precursors. The same data for wires from refs. [7] and [13] are also plotted for comparison.

D. (Ba,Na)Fe_2As_2 Fine Long Wire

Figure 6 shows the (Ba,Na)Fe_2As_2 powder sealed in Ag sheath and a Monel tube used as the outer sheath for the wire. A long wire with a diameter of 300 μm was drawn after inserting the Ag-sheathed powder inserted into the Monel tube. using a draw bench. The final length of the drawn wire reached a length of 90 m, out of which ~20 m section contains the superconducting core. Optical micrographs of transverse and longitudinal cross sections of Ag/Monel-sheathed (Ba,Na)Fe_2As_2 fine wire are shown in Figs. 7(a) and (b), respectively. The shape of the superconducting core is deformed from circular shape, indicating that certain degree of sausaging is happening along the length of the wire. The drawn wire was cut into a few cm pieces and heat treated to sinter superconducting grains at temperatures between 700ºC and 850ºC. Unfortunately, however, I – V measurements of all wires did not show finite critical current densities. Magnetic measurements also indicated negligible superconducting signal superimposed on top of very strong ferromagnetic signal of the Monel. We speculate the reason for the loss of superconductivity can be as follows; (1) Strong deformation during the wire drawing process severely damaged the structure of the material causing disorder induced pair-breaking. (2) Either Ni or Cu in the Monel diffuse into the wire core and caused pair-breaking. (3) Strong deformation of the core during the drawing process caused numerous micro-cracks that hinders the superconducting current flow from one end of the wire to another. Since very small diameter of the wire causes some challenges for further studies, we prepared Ag/Monel wire with a larger diameter. Such preliminary studies using larger diameter wires also demonstrated the weakening of superconductivity in Ag/Monel-sheathed (Ba,Na)Fe_2As_2 wire. Further studies to sort out the true origin of the loss of superconductivity is now in progress.

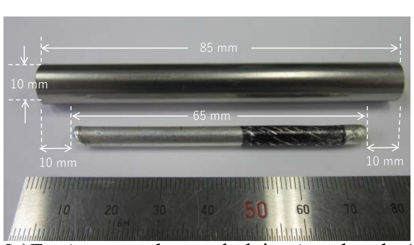

Fig. 7 (Ba,Na)Fe_2As_2 powder sealed in Ag sheath (front) and a Monel tube that serves as an outer sheath (back) .

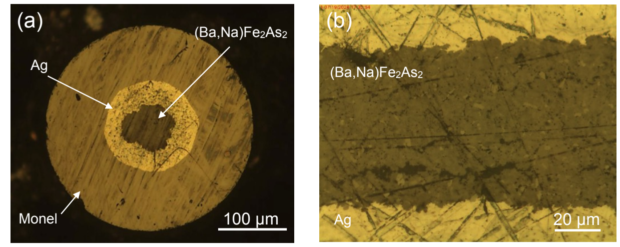

Fig. 8 Optical micrographs of (a) Transverse and (b) longitudinal cross sections of Monel/Ag-sheathed (Ba,Na)Fe_2As_2 wire with 300 μm diameters.

IV. Conclusion

J_c characteristics and uniformity of (Ba,Na)Fe_2As_2 wires and tapes fabricated by different methods and sheath materials are compared. By optimizing the firing and sintering temperatures of powders, we have achieved J_c(4.2 K, 100 kOe) = 48 and 41 kA/cm^2 for a tape fabricated using direct reaction and a wire fabricated by precursor method, respectively. We also succeeded in suppressing the sausaging effect by using AgSn sheath. We also succeeded in drawing fine (300 μm diameter) and long (20 m) wire of (Ba,Na)Fe_2As_2 using Monel as an outer sheath.

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