A comparative study of \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) and \( \text{SmFeAsO}_{1-x}\text{F}_x \) superconducting tapes by magneto-optical imaging

Chao Yao\(^1\), Chunlei Wang\(^1\), Xianping Zhang\(^1\), Dongliang Wang\(^1\), He Lin\(^1\), Qianjun Zhang\(^1\), Yanwei Ma\(^1\), Yuji Tsuchiya\(^2\), Yue Sun\(^2\) and Tsuyoshi Tamegai\(^2\)

\(^1\)Key Laboratory of Applied Superconductivity, Institute of Electrical Engineering, Chinese Academy of Sciences, Beijing 100190, China
\(^2\)Department of Applied Physics, The University of Tokyo, Hongo, Tokyo 113-8656, Japan

E-mail: ywma@mail.iee.ac.cn

Abstract

Using the magneto-optical imaging (MOI) technique, the intergranular critical current density \( J_c \) at various temperatures and the homogeneity of the local structure of the superconducting cores for the powder-in-tube (PIT) \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) and \( \text{SmFeAsO}_{1-x}\text{F}_x \) tapes are systematically investigated. These two tapes have large transport \( J_c \) over \( 10^4 \text{Acm}^{-2} \) in self-field at 20 K and 4.2 K respectively, but the \( J_c \) of the \( \text{SmFeAsO}_{1-x}\text{F}_x \) tape decreases rapidly with the increasing magnetic field. The MOI characterization indicates large bulk currents circulating through the whole sample for the both \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) and \( \text{SmFeAsO}_{1-x}\text{F}_x \) tapes, but also reveals the inhomogeneity inside the \( \text{SmFeAsO}_{1-x}\text{F}_x \) sample. The results obtained from the MO measurements can be confirmed by the magnetic hysteresis measurements \( M(H) \) and the SEM examination. The weak high-field performance of the \( \text{SmFeAsO}_{1-x}\text{F}_x \) tape may be ascribed to its short-time heat treatment.

Keywords: superconductivity, tapes, wires, magneto-optical imaging, critical current density

1. Introduction

The iron-based superconductors (IBS) discovered in 2008 [1] aroused extensive research works to elucidate the superconducting mechanism of this new class of superconductors. At the same time, enormous efforts were devoted to improve their superconducting properties for practical applications. With ultrahigh upper critical fields of about 300 T for 1111 type IBS (ReFeAsO, Re = rare earth elements) and over 100 T for 122 type IBS (AeFe\(_2\)As\(_2\), Ae = alkali or alkali earth elements) [2-5], superconducting wires fabricated with these two types of IBS have great potential for high-field applications. Soon after the discovery of the IBS materials, the first 1111 and 122 type IBS superconducting wires were made using the powder-in-tube (PIT) method [6, 7]. However, it was found that the transport currents of IBS wires were largely deteriorated by high-angle grain boundaries [8, 9] and defects such as cracks, pores, and impurities [10-12]. In recent years, by using the \textit{ex situ} method instead of the \textit{in situ} method used at the early stage, the critical current density \( J_c \) of both 1111 and 122 type IBS superconducting wires has been improved continuously [13]. To date, the \( J_c \) values of \textit{ex situ} 122 IBS wires have rapidly increased above \( 10^4 \text{Acm}^{-2} \) at 4.2 K, 0 T or \( 10^4 \text{Acm}^{-2} \) at 4.2 K, 10 T, by employing a hot isostatic pressing (HIP) [14], a combination technique of chemical addition and rolling texture [15], and cycles of cold deformation and heat treatment [16]. On the other hand, recently \( J_c \) values over \( 2 \times 10^4 \text{Acm}^{-2} \) at 4.2 K, 0 T have been achieved in the Sn added \textit{ex situ} 1111 IBS wires. [17, 18].

In order to further improve the transport properties of the IBS wires and tapes, it is necessary to investigate the intergranular \( J_c \) and the magnetic flux distribution of the material. Magneto-optical imaging (MOI) is an effective technique to visualize the local and real-time distribution of the magnetic flux and reveal the detailed information of the IBS polycrystalline material such as the granularity, homogeneity, intra- and intergranular \( J_c \) [14, 19-22]. In this paper, we report a comparative study of the 122 and 1111 IBS tape samples made by our group. One is the \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) tape with rolling-induced texture made by the low-temperature annealing process with a high transport \( J_c \) value of about \( 10^4 \text{Acm}^{-2} \) at 20 K in self-field, as reported in [23]. The other is the \( \text{SmFeAsO}_{1-x}\text{F}_x \) tape with the highest transport \( J_c \) (\( 2.2 \times 10^4 \text{Acm}^{-2} \) at 4.2 K, 0 T) in 1111 IBS wires, which was heat treated at high temperature up to 1100 °C for a very short time [18]. Both with transport \( J_c \) above \( 10^4 \text{Acm}^{-2} \) at 4.2 K, 0 T, and the \( J_c \) of these two tapes have very different magnetic field dependence. In contrast to the \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) sample, the \( J_c \) of the \( \text{SmFeAsO}_{1-x}\text{F}_x \) sample...
decreases very rapidly with increasing field. Therefore, it is interesting to compare their magnetic performance using the MOI technique. By investigating the intergranular \( J_c \) at various temperatures and the homogeneity of the local structure, it is suggested that the \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) sample has a better quality than the \( \text{SmFeAsO}_1\text{F}_x \) sample, which was caused by a different heat treatment process.

2. Experimental details

The iron-sheathed \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) tape and \( \text{SmFeAsO}_1\text{F}_x \) tape were both fabricated by the \textit{ex situ} PIT process. The detail of the starting materials and the cold deformation processes were reported in [23] and [18], respectively. In contrast to the pure \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) and \( \text{SmFeAsO}_1\text{F}_x \) tapes, for which no transport \( J_c \) was measured, the 5 wt.% Sn added \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) tapes and 30 wt.% Sn added \( \text{SmFeAsO}_1\text{F}_x \) tapes showed high transport \( J_c \) due to the improved grain connectivity. Therefore, in this work, these two kinds of tapes were selected for a comparative study. As mentioned in the introduction, for the heat treatment, the as-rolled \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) tapes were sintered at 800-950 °C for 5-30 min, while the \( \text{SmFeAsO}_1\text{F}_x \) tapes were submitted to a short high-temperature annealing at 1100 °C for 50 s.

The magneto-optical imaging (MOI) characterization was conducted in the Prof. Tamegai’s group at the University of Tokyo. After polishing the superconducting core of the tapes with lapping films, the \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) and \( \text{SmFeAsO}_1\text{F}_x \) samples were cut into small rectangular pieces with dimensions of about 0.82×0.80×0.10 mm\(^3\) and 0.96×0.89×0.06 mm\(^3\), respectively. A Bi-substituted iron-garnet indicator film was placed in direct contact with the sample, and the whole assembly was attached to the cold finger of a He-flow cryostat (Microstat-HR, Oxford Instruments). The MO images of the reflected light were obtained by using a cooled CCD camera with 12-bit resolution (ORCA-ER, Hamamatsu). A differential imaging technique was used to enhance the visibility of the local magnetic induction and remove the contrasts induced by the impurity phases and scratches of the garnet film [24, 25]. In the MO images in this paper, as can be seen in figure 2, there are still a few scratches, but they do not affect the analysis of the results. The transport \( I_c \) was measured at 4.2 K using short tape samples of 3 cm length with the standard four-probe method and evaluated with a criterion of 1\( \mu \)V/cm. The transport \( J_c \) was calculated by dividing the critical current through the cross-sectional area of the superconducting core. The magnetic hysteresis measurements were carried out on a Quantum Design’s physical property measurement system (PPMS). The microstructure of samples was analyzed with a Hitachi S4800 scanning electron microscope (SEM)

![Figure 1. The field dependent transport critical current density \( J_c \) of the \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) (Sr-122) tape and \( \text{SmFeAsO}_1\text{F}_x \) (Sm-1111) tape at 4.2 K.](image)

3. Results and discussion

Figure 1 shows the field dependent transport \( J_c \) of the \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) and \( \text{SmFeAsO}_1\text{F}_x \) tapes at 4.2 K. The magnetic fields were applied parallel to the tape surface. Though the transport \( J_c \) of the \( \text{SmFeAsO}_1\text{F}_x \) tape is as high as 2.2×10\(^4\) Acm\(^{-2}\) in self-field, it drops very rapidly with the increase of the applied field. When the applied field is larger than 1 T, the \( J_c \) value decreases more than one order of magnitude. One the other hand, for the \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) tape, the transport \( J_c \) decreases very slowly from 1 T to 5 T, suggesting a much better high-field performance. In addition, the value of the transport \( I_c \) in self-field for this tape is beyond the maximum (~200 A) of the current source used in the \( I_c \) measurement, so its transport \( J_c \) in self-field is supposed to be higher than 2.8×10\(^4\) Acm\(^{-2}\).

Figure 2 presents the MO images of the magnetic flux penetration after zero-field cooling (ZFC) the samples to 5 K. The external magnetic fields were applied perpendicular to the sample surface. The bright regions represent the part where the magnetic flux exists. For the \( \text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) sample, due to the strong shielding currents, the magnetic flux does not penetrate into the sample when the external field is less than 20 mT. When the field is further increased, it gradually moves towards the sample center from the edge, but even when the applied is increased to 50 mT, it still just partially penetrates the sample. This result indicates strong bulk circulating currents in the sample, which can be ascribed to the good grain connectivity and the reduced mismatch of the grains with textured microstructure. The magnetic flux penetration patterns of our tapes are similar to that of the bulk material synthesized by the HIP technique [14], and completely different from that of the weak linked 122 IBS wire sample [21]. For the \( \text{SmFeAsO}_1\text{F}_x \) sample as shown in figure 2(b), the penetration of the magnetic flux
into the sample towards the center appears faster than the 122 sample in figure 2(a) with increasing field. However, we can still find the shielding currents go through the whole sample when the field is lower than 20 mT. This result is better than the previously reported Sm-1111 IBS bulk sample [19], indicating the Sn addition in our sample can effectively improve the connectivity between grains. On the other hand, due to the poor crystallinity and large grain-misalignment, the global shielding currents of this sample cannot be kept as well as that of the Sr$_{1-x}$K$_x$Fe$_2$As$_2$ sample when increasing the applied fields.

In order to reveal the relationship between the global $J_c$ and the microstructure of the samples, we have investigated the remnant trapped fields after removing the applied fields at 5 K and 20 K for the zero-field-cooled (ZFC) samples. Figure 3(a) shows the magnetic induction line profile along the red line on the MO image. After cycling the field up to 80 mT for 0.2 s at 5 K, the magnetic flux still does not reach the center of the Sr$_{1-x}$K$_x$Fe$_2$As$_2$ sample, indicating very strong shielding currents in the sample. In this case, the intergranular $J_c$ can be estimated using the equation [26, 27],

$$J_c = \frac{H_{ex}c}{4d\cosh^{-1}[w/(w-2p)]}$$

where $H_{ex}$ is the external field, $d$ is the sample thickness, $w$ is the sample width, and $p$ is the flux penetration distance from the sample edge. In figure 3(a) the right blue dashed line indicates the edge of the sample, and the left blue dashed line indicates the front edge of the flux penetration. Therefore, with $H_{ex}$=800 Oe, $c$= 10, $d$= 100 μm, $w$= 820 μm and $p$= 274 μm, the $J_c$ at 5 K for the Sr$_{1-x}$K$_x$Fe$_2$As$_2$ sample is calculated to be about $1.1\times10^5$ Acm$^{-2}$. In figure 3(c), at 20 K
we obtain a very uniform roof top pattern of the fully trapped magnetic flux, and the line profile calculated from the MO image data also shows a roof shape, suggesting bulk currents flow over the whole sample. Since the cycling external field penetrates the whole sample at 20 K, we can estimate the intergranular $J_c$ using the equation,

$$J_c \sim \frac{\Delta B}{d} \quad (2)$$

where $\Delta B$ is the trapped field, and $d$ is the thickness of the sample. According to the magnetic induction line profile, the global $J_c$ at 20 K of this sample is estimated up to $4 \times 10^4$ Acm$^{-2}$. Similarly, as reported in [18], the global $J_c$ at 5 K for the SmFeAsO$_{1-x}$F$_x$ sample estimated from figure 3(b) is about $3.6 \times 10^4$ Acm$^{-2}$. Though the trapped magnetic flux in the SmFeAsO$_{1-x}$F$_x$ sample also shows a roof pattern at 5 K, it can be found that the sample is not as homogenous as the 122 sample at 20 K in figure 3(d). We can see some bright local regions in the remnant state MO image, and the curve of the line profile has sharp spikes, indicating that the quality of the grains is not uniform and there are still weak links between grains.

To confirm the intergranular $J_c$ estimated from the MOI results, we measure the magnetic hysteresis at various temperatures of the two samples used in the MO measurements, as shown in figure 4 (a) and (c). The magnetic fields were applied perpendicular to the sample surface. The bulk magnetic $J_c$ can be derived from the magnetic hysteresis loops using the Bean model [28].

Figure 4. (a) The magnetic hysteresis loops and (b) the field dependent magnetic $J_c$ for the Sr$_{1-x}$K$_x$Fe$_2$As$_2$ sample at various temperatures. (c) The magnetic hysteresis loops and (d) the field dependent magnetic $J_c$ for the SmFeAsO$_{1-x}$F$_x$ sample at various temperatures.
Figure 6. Microstructure of the superconducting cores investigated by SEM. (a) SEM image of the Sr\textsubscript{1-x}K\textsubscript{x}Fe\textsubscript{2}As\textsubscript{2} sample showing a well-coupled planar structure with partial c-axis texture. (b) SEM image of the SmFeAsO\textsubscript{1-x}F\textsubscript{x} sample showing an inhomogenous structure with cracks and pores.

By SEM analysis, we examined the microstructure of the samples. In figure 6(a), the superconducting core of the Sr\textsubscript{1-x}K\textsubscript{x}Fe\textsubscript{2}As\textsubscript{2} tape shows a well-coupled planar structure with a flat smooth surface, indicating the c-axis texture along the tape surface. There are only a few micro cracks and pores in the grains. For the SmFeAsO\textsubscript{1-x}F\textsubscript{x} sample, as shown in figure 6(b), its grains have a much smaller size than the 122 sample. It is obvious that the quality of the grain is not homogenous, and the grain alignment is not as uniform as the textured Sr\textsubscript{1-x}K\textsubscript{x}Fe\textsubscript{2}As\textsubscript{2} grains, which can result in the weak link at the grain boundaries, and are in consistence with the results of the MOI characterization. The weak performance of the current carrying ability when increasing the fields and temperature of the SmFeAsO\textsubscript{1-x}F\textsubscript{x} tape may be due to the short annealing time and the quick quenching process, which will cause the low quality of grains and the inhomogeneity of microstructures. On the other hand, if increasing the time for the heat treatment, the superconducting core may react with the sheath material, and the loss of fluorine element will be more at the temperature as high as 1100 °C. If the heat treatment process for the 1111 IBS tapes can be improved, its transport properties will be largely enhanced in the future.

4. Conclusions

In this work, we use the MOI technique to characterize the superconducting properties of the powder-in-tube Sr\textsubscript{1-x}K\textsubscript{x}Fe\textsubscript{2}As\textsubscript{2} and SmFeAsO\textsubscript{1-x}F\textsubscript{x} tapes. The MO measurement show that the textured Sr\textsubscript{1-x}K\textsubscript{x}Fe\textsubscript{2}As\textsubscript{2} sample heat treated at low temperature has very uniform and well-coupled grains. On the other hand, similar to the Sr\textsubscript{1-x}K\textsubscript{x}Fe\textsubscript{2}As\textsubscript{2} sample, the SmFeAsO\textsubscript{1-x}F\textsubscript{x} tape made by the short high-temperature sintering shows bulk shielding currents in the whole sample region at low temperature around 5 K, but at 20 K the MO measurement reveal inhomogeneity inside the sample, which may cause the rapid decrease of its transport properties when increasing the field and temperature. The results of the MO measurements are in accordance with the M(H) measurements, suggesting that the weak performance of the SmFeAsO\textsubscript{1-x}F\textsubscript{x} tape may be the result of the short annealing time during the heat treatment.

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