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Large transport J_c in Cu-sheathed $\text{Sr}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ superconducting tape conductors

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Copper sheath is the first choice for manufacturing high- T_c superconducting wires and tapes because of its high electrical and thermal conductivities, low-cost and good mechanical properties. However, Cu can easily react with superconducting cores, such as BSCCO, MgB_2 and pnictides, and therefore drastically decrease the transport J_c . Here, we report the fabrication of Cu-sheathed $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ tapes with superior J_c performance using a simple hot pressing method that is capable of eliminating the lengthy high-temperature sintering. We obtained high-quality $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ tapes with processing at 800 °C for 30 minutes and measured high T_c and sharp transition. By this rapid fabrication, Cu sheath does not give rise to apparent reaction layer, and only slightly diffuses into Sr-122 core. As a consequence, we achieved high transport J_c of $3.1 \times 10^4 \text{ A/cm}^2$ in 10 T and $2.7 \times 10^4 \text{ A/cm}^2$ in 14 T at 4.2 K. The in-field J_c performance is by far the highest reported for Cu-sheathed high- T_c conductors. More importantly, Cu-sheathed Sr-122 tapes also showed a high J_e value of $1.0 \times 10^4 \text{ A/cm}^2$ in 10 T at 4.2 K, which has reached the widely accepted practical level for applications. These results demonstrate that Cu is a very promising sheath for the practical application of pnictide conductors.

The discovery of the iron-based superconductors, with a relatively high critical temperature T_c , ultra-high upper critical fields H_{c2} and low anisotropy γ , has inspired worldwide research efforts^{1–5}. Studies on single crystals and thin films reveal that iron-pnictides have large in-field critical current density J_c due to strong intrinsic flux pinning^{6–8}. To explore the potential of using this new material for magnet applications, significant efforts had been focused on developing wires and tapes with in-field high J_c . Our research group prepared $\text{LaFeAsO}_{1-x}\text{F}_x$ and $\text{SmFeAsO}_{1-x}\text{F}_x$ superconducting wires using *in-situ* powder-in-tube (PIT) method⁵. A Ta tube or Fe tube with an inner Ti sheath was used to prevent the reaction between the tubes and the superconducting compounds. Zhang *et al.* systematically studied the effect of various sheath materials (Nb, Ta and Fe/Ti) on the microstructure and superconducting properties of $\text{SmFeAsO}_{1-x}\text{F}_x$ wires and reported consistent formation of a thick reaction layer between the core and metal sheath, which hinders the performance of transport J_c ⁹. Therefore, a key issue is to develop a technology that result in little or no reaction with iron-pnictides. Subsequently, Wang *et al.* reported Ag-sheathed $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ (Sr-122) conductors with transport J_c of 1200 A/cm^2 (at 4.2 K and self-field)¹⁰. The Sr-122/Ag interface was clear and no observable reaction layer, indicating that Ag sheath is benign in proximity to the compound at high sintering temperature. Fe-sheathed Sr-122 and $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ (Ba-122) tapes were also fabricated by *ex-situ* PIT method^{11–13}. Through optimization of sintering process and rolling texture, J_c of $>10^4 \text{ A/cm}^2$ in 10 T at 4.2 K was reported for Sr-122 tapes¹⁴. Recently, Ag-sheathed Ba-122 and Sr-122 conductors have been investigated intensively, and high J_c exceeding 10^4 A/cm^2 (Maximum $J_c = 1.2 \times 10^5 \text{ A/cm}^2$) at 4.2 K and 10 T has been reported by applying uniaxial pressing, such as hot pressing (HP) and cold pressing (CP)^{15–19}. By far, all pnictide wires and tapes with

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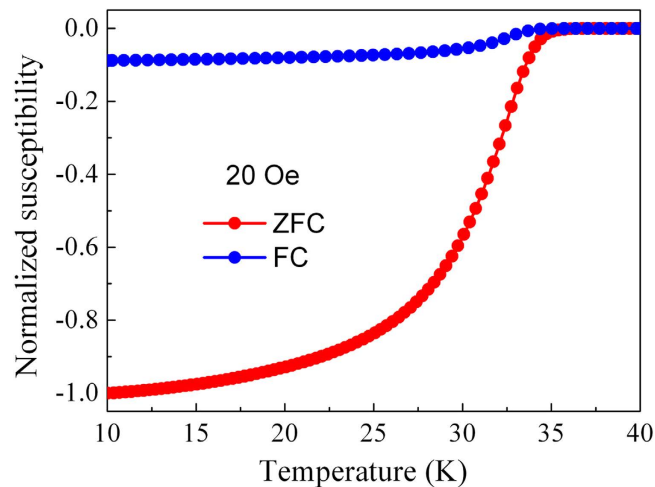


Figure 1. Magnetization versus temperature curves of the precursor powders.

the high J_c - B performance ($>10^4$ A/cm², at 4.2 K and 10 T) are prepared by using the expensive Ag or magnetic Fe sheath, which is similar to Ag-sheathed BSCCO and Fe-sheathed MgB₂ conductors^{20–23}.

For practical applications of high- T_c superconductors including cuprate, pnictide and MgB₂, the copper material is a desirable sheath because of many advantages^{24–26}. Firstly, when compared with common Ag and Fe sheath, Cu is a low-cost and nonmagnetic material. Secondly, Cu sheath has good mechanical properties, which make the coil winding easier in magnet applications. Thirdly, high purity Cu has large residual resistivity ratio (RRR) value, and provides both electromagnetic stabilization against flux jumps and quench protection^{27,28}. It is well known that Cu material has been proved to be an effective sheath in the conventional NbTi and NbSn₃ conductors. Cu is also used as stabilizer in high- T_c conductors, such as Nb/Cu/monel MgB₂ wires^{29,30}, for providing electrical stability of magnets and other devices during transients. However, since the discovery of cuprate superconductors in 1986, no high transport J_c - B performance ($>10^4$ A/cm², at 4.2 K and 10 T) has been reported for Cu-sheathed high- T_c superconductors. Cu is highly reactive to superconducting core at high-temperature sintering^{31–33}. The interfacial reaction layer and composition deviation of superconducting phase can lead to J_c degradation. In worse case, no transport J_c can be detected, because the thick reaction layer apparently prevented electric current from flowing from the sheath material to the superconducting core. Therefore, it is considered a grand challenge to develop a process for Cu-sheathed high- T_c superconductors with superior performance. In the present work, we report successfully fabricated Cu-sheathed Sr-122 tapes by an *ex-situ* PIT method. DC susceptibility of Sr-122 precursor powders was measured, and the result is shown in Fig. 1. The significant shielding currents appear at about 36.0 K and increase as the temperature decreased, which is similar to that reported for high-quality precursors¹⁷. During the final heat treatment, we introduce a hot pressing process with combination of short-time sintering (800 °C/30 min or 700 °C/60 min) and low external pressure (~20 MPa). This rapid fabrication can effectively avert the formation of reaction layer, and therefore result in a high transport J_c of 3.1×10^4 A/cm² at 4.2 K and 10 T.

Results

Cu-sheathed Sr-122 tapes were finally hot pressed at 700 °C (HP700 tapes) and 800 °C (HP800 tapes). Fig. 2(a) shows a typical transverse cross-sectional optical image of HP800 tapes. After hot-pressing, the tape thickness of HP800 samples decreased from ~0.40 mm to ~0.29 mm. Fig. 2(b) displays a longitudinal cross-sectional optical microstructure of HP800 tapes. A uniform deformation of both superconducting core and Cu sheath along the length can be obviously seen, which is essential for the achievement of high transport J_c ^{34,35}. This uniformity is attributable to the good mechanical properties of Cu sheath.

As shown in Fig. 3, the XRD analysis was performed on the planar surfaces of superconducting cores after peeling off Cu sheath. For comparison, the data for randomly orientated precursor powders is also included. The XRD patterns on the surfaces clearly exhibit a ThCr₂Si₂-type structure, ensuring that Sr-122 is the main phase for both HP700 and HP800 samples. Using a final short-time hot-pressing process, the formation of non-superconducting reaction layer at the interface seems to be prevented. More importantly, the transport critical current I_c may be measured and obtained in these Sr-122 tapes³¹. However, the impurity peaks are detected on the core surface, especially for HP800 samples. Some Cu reacted with Sr-122 phase, producing SrCuAs and Cu_{9.5}As₄ phases. This is consistent with large FWHM (full width at half-maximum) of (002) and (103) peaks for Sr-122 phase. On the other hand, the XRD patterns for the central planar sections of HP tapes after carefully polishing are also exhibited in Fig. 3. The diffraction peaks have some differences compared to those of the surfaces. The XRD patterns of central parts exhibit pure Sr-122 phase without detectable impurities. No Cu element can be detected in

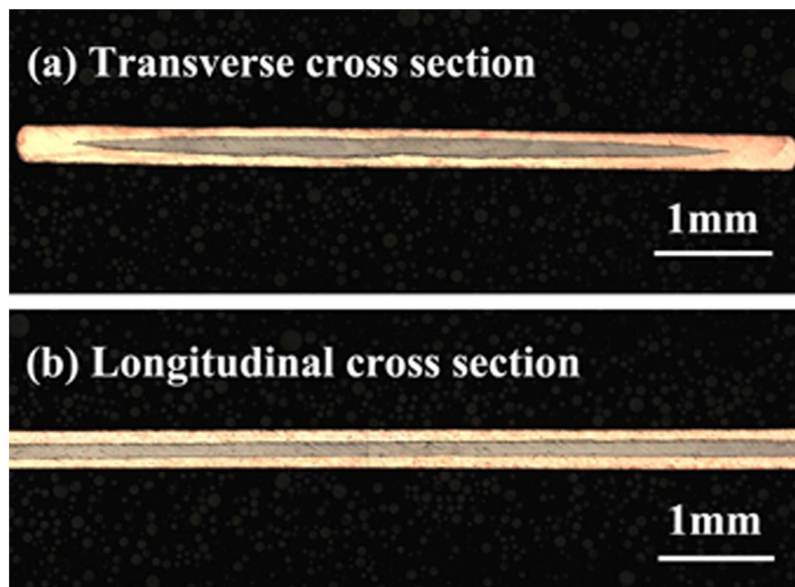


Figure 2. Optical images of Cu-sheathed Sr-122 tapes: (a) the transverse cross section of HP800 tapes; (b) the longitudinal cross section of HP800 tapes.

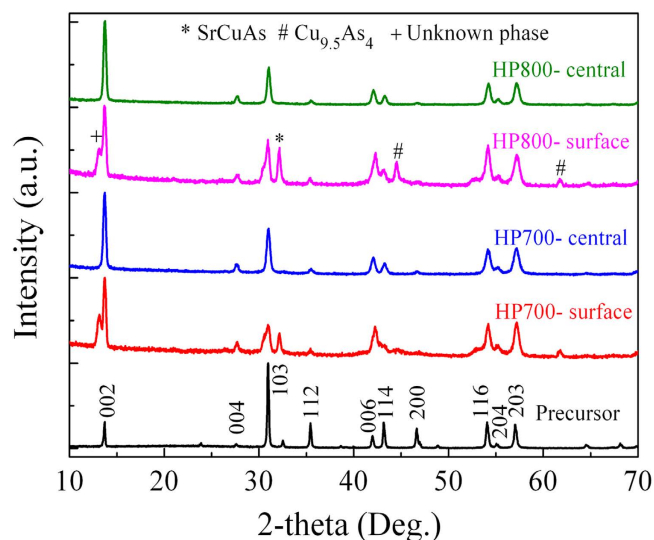


Figure 3. XRD patterns for the planar surfaces and central sections of the superconducting cores of both HP700 and HP800 samples. As a reference, the data for randomly orientated precursor powders is also included. The peaks of $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ phase are indexed, while the peaks of impurity phases are also marked.

the central parts by further EDX identification. The peak characteristics are similar to those of textured Sr-122 tapes^{17–19}, which have high transport J_c -B properties. We quantify the c-axis texture parameter F according to the Lotgering method³⁶ with $F = (\rho - \rho_0)/(1 - \rho_0)$, where $\rho = \Sigma I(00l)/I(hkl)$ and $\rho_0 = \Sigma I_0(00l)/I_0(hkl)$. I and I_0 are the intensities of corresponding XRD peaks measured for the textured and randomly oriented samples, respectively. F values of 0.41 and 0.44 were obtained for HP700 and HP800 tapes, demonstrating that c-axis oriented grains have been achieved in Cu-sheathed tapes. The larger F value in HP800 samples is in agreement with the previous reports confirming that the higher HP temperature, the larger degree of grain alignment¹⁸.

DC susceptibility measurements were conducted on HP700 and HP800 samples. Fig. 4(a) depicts two typical groups of the susceptibility curves under a 20 Oe magnetic field parallel to the tape plane. The superconducting transition of HP700 tapes begins at about 33.0 K. It is evident from the zero-field cooled (ZFC) signal that the susceptibility starts to decrease slowly and full shielding is reached at about 15 K.

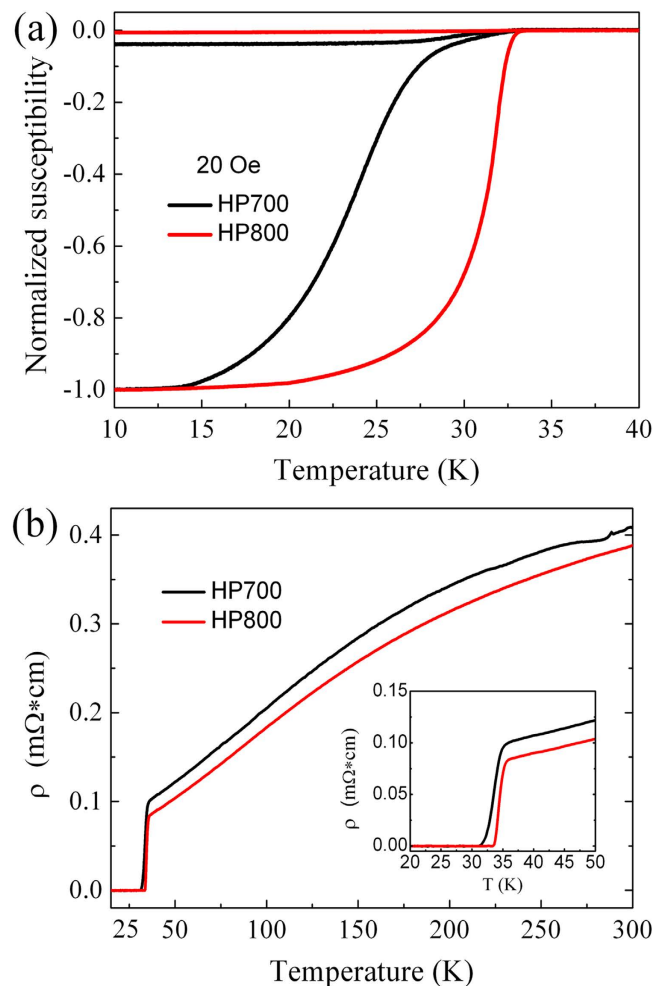


Figure 4. (a) Temperature dependence of the DC magnetic susceptibility curves of HP700 and HP800 samples. (b) Resistivity versus temperature curves of HP700 and HP800 samples; Inset showing the enlarged part near the superconducting transition. All data were obtained after peeling off the Cu sheath.

This behavior suggests the presence of inhomogeneity^{14,37}. For HP800 samples, the shielding current occurs at 33.5 K, which may be ascribed to improvement in crystallization. When compared to HP700 samples, the HP800 samples exhibit sharper superconducting transition and reach full shielding at higher temperature (≈ 20 K). Obviously, enhanced uniformity in superconducting phase has been achieved in HP800 samples³⁷. Fig. 4(b) shows resistivity versus temperature curves. We measured onset T_c values of 34.6 and 35.1 K for HP700 and HP800 tapes, respectively, which are comparable to Fe-sheathed and Ag-sheathed tapes^{17,19,38}, but slightly smaller than those reported in ref. 18. The impurity of copper compound in present work does not significantly affect the superconducting transition. In addition, the resistivity of HP700 and HP800 tapes drops to zero T_c at 32.3 and 33.8 K, respectively. The larger onset T_c and smaller transition width for HP800 samples are consistent with the above magnetic results.

From the viewpoint of practical applications, superconducting wires must be able to carry large transport current density in high magnetic fields. We determined the transport I_c by the standard four-probe method. Fig. 5 displays the J_c - B properties of HP700 and HP800 tapes at 4.2 K. For HP700 tapes, the J_c values of 3.5×10^4 A/cm² and 4.2×10^3 A/cm² are obtained in self-field and 10 T, respectively. The striking result is that HP800 tapes show a great enhancement of J_c values in the whole field up to 14 T. Such an improvement can be attributed to improved texture, better homogeneity and crystallization. For HP800 tapes, the J_c data in self-field is not given because the transport I_c is too large to be measured by the measurement system we used. Excitingly, the transport J_c reaches 3.1×10^4 A/cm² at 10 T. To our knowledge, this is by far the highest critical current density under high field ever reported for Cu-sheathed high- T_c superconductors. Importantly, due to its extremely small magnetic field dependence, the transport J_c still maintains a high value of 2.7×10^4 A/cm² in 14 T. It is convincible that the Cu-sheathed Sr-122 tapes have a very promising future for use in high-field superconducting magnets.

We conducted SEM/EDX to investigate the influence of hot pressing process on the microstructure of Cu-sheathed Sr-122 tapes. As shown in Figs. 6(a,b), both HP700 and HP800 samples exhibit dense

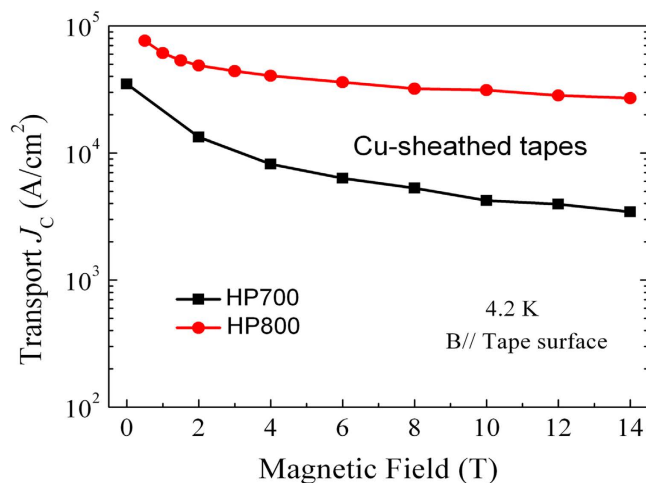


Figure 5. Magnetic field dependence of the transport J_c at 4.2 K for both HP700 and HP800 tapes. The applied fields up to 14 T were parallel to the tape surface.

Sample name	Fabrication conditions	Onset T_c (K)	J_c (kA/cm ²) (4.2 K, 10 T)
R700	700 °C/1 h	34.3	1.3
R800	800 °C/0.5 h	34.5	3.0
HP700	700 °C/1 h/20 MPa	34.6	4.2
HP800	800 °C/0.5 h/20 MPa	35.1	31.0

Table 1. The onset T_c and transport J_c for Cu-sheathed Sr-122 tapes with and without hot pressing.

layered structure, which is similar to that of Bi-2223 tapes. HP700 samples have smaller grain size than that of HP800 samples. Fig. 6c exhibits a typical SEM micrograph of polished cross section of HP800 samples. It is noted that the boundary between Cu sheath and Sr-122 core is clear, further suggesting that there is no apparent reaction layer after hot pressing¹³. The corresponding EDX element mappings of HP800 tapes are presented in Figs. 6(d–i). From the Cu mapping, we observe a diffusion of Cu into Sr-122 area, and the diffusion width is approximately 8 μm . This indicates that Cu element interdiffuses into Sr-122 core and reacts with it during heat treatment. For the elements of Sr-122 phase, Sr, K, Fe and As are detected locally in the core area, disappear almost completely at the border of the core area. Comparing with recent Ag-sheathed Sr-122 tapes¹⁸, we conclude that the slight depression of superconducting properties in this work is mainly due to the diffusion of Cu. At the same time, the diffusion also causes the inhomogeneous distribution of the superconducting elements in Sr-122 area, particularly in the diffusion region. In addition, the EDX mapping of HP700 samples is showed in Fig. 6(j). For each element, the content has a dramatic change at the border of Sr-122 core. Further analysis reveals that the diffusion width of Cu element is smaller than 3 μm in HP700 samples. Although the sintering time of 60 min is longer than that used for HP800 tapes (30 min), the width is much smaller.

The diffusion of Cu and the composition deviation of superconducting phase easily induce severe porosity at the interface, and apparently break the electrical contact between Cu sheath and Sr-122 core^{39,40}. This disadvantageous phenomenon can be avoided by the simple HP method, because it can greatly reduce the pores and cracks by combining the deformation and heat treatment in a single step. As shown in Fig. 6c, the Cu sheath and Sr-122 core are tightly connected. As a result, high transport J_c values have been measured in our Cu-sheathed tapes.

For comparison, Cu-sheathed Sr-122 tapes were also sintered without hot-pressing, and the detailed information is exhibited in Table 1. The transport J_c values for both HP tapes are much larger than those of corresponding tapes without hot-pressing. For example, the J_c value of HP800 tapes ($3.1 \times 10^4 \text{ A/cm}^2$) is an order of magnitude higher than that of R800 tapes ($3.0 \times 10^3 \text{ A/cm}^2$), indicating the great J_c enhancement by the hot-pressing method.

Discussion

Using copper sheath for superconducting tapes with large transport J_c is highly desirable for practical applications. By a modified hot-pressing method with combination of final short-time sintering and low external pressure, we successfully prepared Cu-sheathed Sr-122 conductors with large transport current.

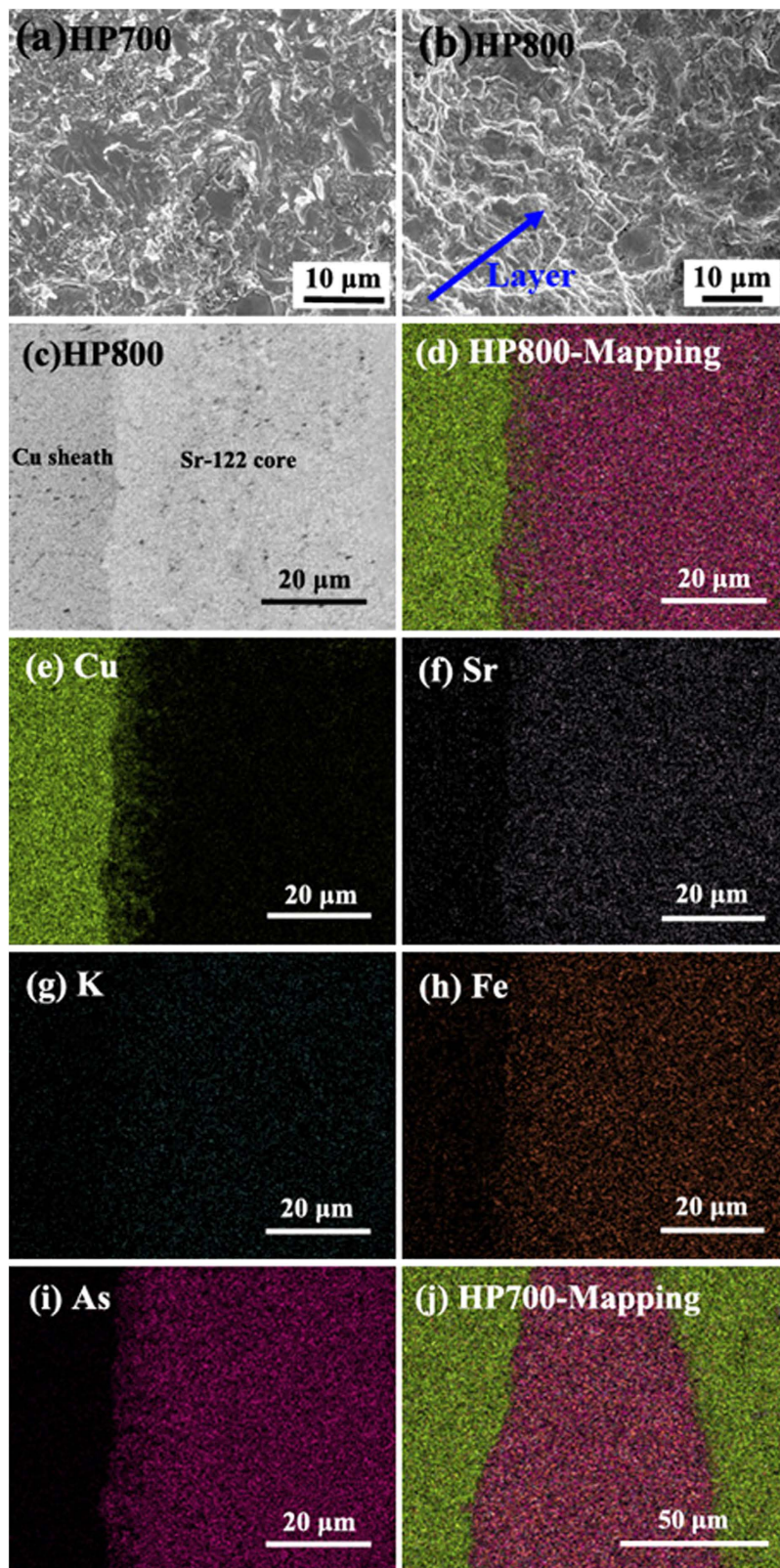


Figure 6. SEM microstructures of Sr-122 tapes: (a) HP700 and (b) HP800 samples. (c) SEM image showing the interface between Cu sheath and Sr-122 core for HP800 tapes. (d) The corresponding EDX mapping image for HP800 samples; (e–i) Area mappings of Cu, Sr, K, Fe and As element, respectively. (j) The EDX mapping image for HP700 samples.

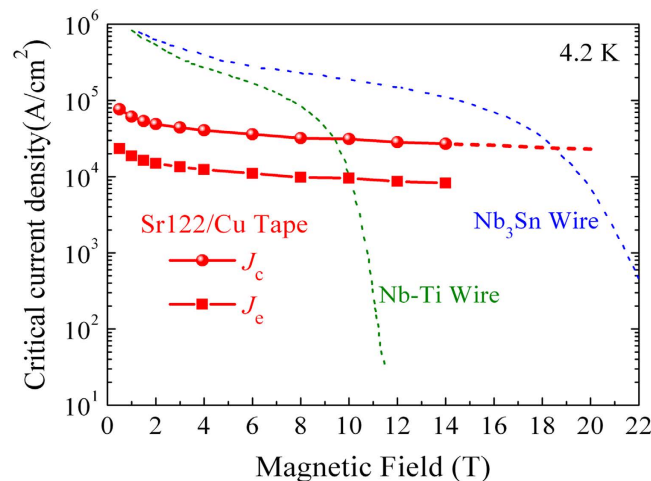


Figure 7. Magnetic field dependence of the transport critical current density J_c and the engineering current density J_e for our best Sr122/Cu tapes. The J_c - B curves of PIT processed NbTi and Nb₃Sn wires are also shown for comparison.

We demonstrated that the fabricating method developed in our lab can produce high-performance Cu-sheathed superconductors. First, a short-time hot-pressing process can form high-quality Sr-122 phase, which is supported by XRD and resistivity characterizations. For HP800 tapes, the resistivity data demonstrates that the onset T_c is 35.1 K with a transition width of about 1.5 K. Second, this fast fabrication does not give rise to the reaction layer even though the Cu sheath is used. As discussed by above EDX mappings, only a little bit of Cu diffuses into polycrystalline Sr-122 phase. Earlier studies reveal that the thick reaction layer induces the contamination of the superconducting phase to decrease T_c , and prevents electric current from flowing from the sheath material to the superconducting core^{26,33}. Third, the Cu sheath and Sr-122 core are tightly connected under external pressure, and thus the current path can be enlarged. Meanwhile, the hot pressure can not only considerably increase the core density, but also effectively promote complete reaction of Sr-122 phase, which in return to solve the problem that is low sintering temperature (800 or 700 °C) and short-time reaction (30 or 60 min) yield poor re-crystallization and ordinary superconducting performance^{18,38}. In summary, the simple hot pressing method ensures high-quality Sr-122 phase and inhibit the formation of reaction layer in Cu-sheathed Sr-122 tapes.

It is fascinating that the largest J_c value of 3.1×10^4 A/cm² in 10 T has been obtained in our best Cu-sheathed tapes. Moreover, the J_c of 122-type pnictides have very weak field dependence in strong fields up to 28 T⁴¹, in accordance with ultrahigh H_{c2} values⁵. Thus, the J_c data above 14 T is given by extrapolating from low fields, as presented in Fig. 7. The curve tendency shows that the crossovers with Cu-sheathed NbTi and Nb₃Sn wires are around 9.5 and 18.5 T, respectively. This clearly strengthens the position of pnictide conductors as a competitor to the conventional superconductors for high-field applications. On the other hand, researchers are usually more concerned with the engineering (total cross section) current density J_e in practical applications. As showed in Fig. 7, a high J_e of about 1.0×10^4 A/cm² at 10 T has been achieved in our Cu-sheathed Sr-122 tapes, which has reached the widely accepted practical level for applications²⁸. This achievement is a significant technical breakthrough for the practical applications of Cu-sheathed high- T_c conductors. In the future, if the HP process can be properly adjusted to match the balance between the well re-crystalline reaction and little impurity phase, an even higher J_e can be expected.

The specific cost (\$/kA·m) of superconducting wires and tapes must be considered in practical applications^{25,28}. The price (\$/kg) of Cu metal is 1-2 order of magnitude lower than that of expensive Ag metal. Tape conductors with high J_e (10^4 A/cm²) sheathed in comparatively cheap copper have the strong potential for low specific cost. Moreover, Cu-sheathed conductors do not need additional stabilization or mechanical reinforcement. In contrast, the Ag sheathed wires usually need mechanical reinforcement. For example, the stainless steel or Ag0.5 wt%Al alloy sheath have been used in superconducting wires⁴¹⁻⁴³, which decrease the engineering J_e or increase the complexity and cost of fabrication process. From these view points, we can conclude that the comprehensive performances of our Cu-sheathed Sr-122 tapes are much more attractive for applications than the reported Ag sheathed tapes¹⁶⁻¹⁹, demonstrating that Cu is a very promising sheath for the pnictide wires and tapes.

Methods

Sample preparation. We fabricated Cu-sheathed Sr_{0.6}K_{0.4}Fe₂As₂ tapes by *ex-situ* PIT method. Sr fillings, K pieces, and Fe and As powder with a ratio of Sr:K:Fe:As = 0.6: 0.5: 2: 2.05 were mixed for 12 hours

by ball-milling method. The milled powders were packed into Nb tubes and then sintered at 900 °C for 35 h. As prepared Sr-122 superconducting powders were packed into Cu tubes with OD 6 mm and ID 4 mm. These tubes were sealed and then cold worked into tapes (~0.4 mm thickness) by swaging, drawing and flat rolling. Finally, hot pressing was performed on the 60 mm long tapes under ~20 MPa at two different sintering processes of 800 °C/30 min and 700 °C/60 min. These tapes are defined as HP800 and HP700 tapes, respectively.

Measurements. Phase identification of samples was characterized by X-ray diffraction (XRD) analysis with Cu K α radiation. Magnetization versus temperature curves and resistivity measurements of the superconducting cores were carried out using a PPMS system. The cross sections were polished and then observed by optical images. Microstructure characterization was analyzed using SEM images and EDX scanning. The transport critical current I_c was measured at 4.2 K using short tape samples of 3 cm in length with the standard four-probe method and evaluated by the criterion of 1 μ V/cm. The applied fields up to 14 T in transport I_c measurement were parallel to the tape surface.

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Author Contributions

Y.W.M. directed the research. H.L. designed the experimental plan and fabricated the tape samples. H.L. carried out XRD, magnetization, resistivity, and microstructure characterizations. X.P.Z. and D.L.W. did the high-field I_c measurement. Q.J.Z. and C.H.D. helped with the tape preparation. C.Y. and H.T.Z. contributed to manuscript preparation. H.L. and Y.W.M. wrote the manuscript. All the authors contributed to discussion on the results for the manuscript.

Additional Information

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