Recent Progress on the Development of Iron-based Superconducting Wires for High-field Applications

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Outline

1. Properties & application potential of iron-based superconductors
2. Microstructural defects & weak-link GBs in polycrystalline IBS
3. Improving the $J_c$-performance of IBS wires and tapes
4. Progress on practical wires for high-field applications
5. Summary & prospects
Crystal structures of iron-based superconductors

REFeAsO
[1111]
RE: rare earth

AEFe$_2$As$_2$
[122]
AE: Ba, Sr

AFeAs
[111]
A: Li, Na

FeCh
[11]
Ch: Se, Te, S

SmFeAsO$_{1-x}$F$_x$
$T_c = 55$ K

Ba$_{1-x}$K$_x$Fe$_2$As$_2$
$T_c = 38$ K

LiFeAs
$T_c = 18$ K

Fe(Se,Te)
$T_c = 16$ K

• basically tetragonal with long $c$-axes including a Fe plane ($ab$-direction)
• large structural variation at blocking layer

SmFeAsO$_{1-x}$F$_x$
$T_c = 55$ K
- Exceptionally high $H_{c2}$ for 1111- and 122-type iron-based superconductors
- Small anisotropy gives high vortex stiffness
Upper critical fields of iron-based superconductors

- The conventional low-$T_c$ superconductors (NbTi & Nb$_3$Sn) restrict the magnets with field below 25 T at liquid helium temperature.

- For 1111- and 122-type IBS, the $H_{c2}$ is still above 40 T at 20 K.

- Promising for applications operated at liquid helium temperature and also in moderate temperature around 20 K, which can be obtained by cryocoolers.

Comparative T-H phase diagram for different superconducting materials

Application potential of iron-based superconductors

- $T_c = 38$ and $56$ K in 122 & 1111 system
- Ultrahigh $H_{c2} > 80$ T
- Very small anisotropy $\gamma = 1.5$~2
- Strong vortex pinning

Shimoyama 2014 SuST 27 044002

Promising candidate for:

- NMR
- Accelerator
- MRI
## Application potential of iron-based superconductors

Superconductivity parameters for practical superconductors

<table>
<thead>
<tr>
<th>Material</th>
<th>$T_c$ (K)</th>
<th>$H_{c2,4.2K}$ (T)</th>
<th>Coherence length $\varepsilon_{ab}$ (nm)</th>
<th>Anisotropy $\gamma_H$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nb47wt%Ti</td>
<td>9</td>
<td>11.5</td>
<td>4</td>
<td>Negligible</td>
</tr>
<tr>
<td>Nb$_3$Sn</td>
<td>18</td>
<td>25</td>
<td>3</td>
<td>Negligible</td>
</tr>
<tr>
<td>MgB$_2$</td>
<td>39</td>
<td>25</td>
<td>6.5</td>
<td>2~2.7</td>
</tr>
<tr>
<td>YBCO</td>
<td>92</td>
<td>&gt;100</td>
<td>1.5</td>
<td>7</td>
</tr>
<tr>
<td>Bi-2223</td>
<td>110</td>
<td>&gt;100</td>
<td>1.5</td>
<td>50~100</td>
</tr>
<tr>
<td>Bi-2212</td>
<td>90</td>
<td>&gt;100</td>
<td>1.5</td>
<td>50~100</td>
</tr>
<tr>
<td>Sm-1111</td>
<td>55</td>
<td>&gt;100</td>
<td>1.8~2.3</td>
<td>5~10</td>
</tr>
<tr>
<td>Ba-122</td>
<td>38</td>
<td>&gt;80</td>
<td>1.5~2.4</td>
<td>1.5~2</td>
</tr>
<tr>
<td>Fe(Se,Te)</td>
<td>16</td>
<td>&gt;40</td>
<td>1.2</td>
<td>1.1~1.9</td>
</tr>
</tbody>
</table>
1. Properties & application potential of iron-based superconductors
2. Microstructural defects & weak-link GBs in polycrystalline IBS
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4. Progress on practical wires for high-field applications
5. Summary & prospects
$J_c$ in IBS single crystals and films

- IBS single crystals and films show high in-field $J_c$ above 1 MA
- Very weak field dependence of $J_c$

**Sm-1111 single crystal**
Moll 2010 *Nature Mater.* 9 628

**Ba-122:K single crystal**
Yang 2008 *APL* 93 142506

**Ba-122:Co films** 2.6 MA (9 T, 4.2 K)
Yuan 2017 *SuST* 30 025001

**FeSeTe films** 0.97 MA (9 T, 4.2 K)
Yuan 2015 *SuST* 28 065009
Grain boundary nature of 122 pnictides

- $J_c$ decreases exponentially with increasing GB angle.
- The critical angle $\theta_c$ of Ba-122 GBs is $9^\circ$, larger than YBCO ($\theta_c \sim 5^\circ$).

The traditional **powder-in-tube (PIT) method**, which has been utilized in commercial $\text{Nb}_3\text{Sn}$, Bi-2223 and MgB$_2$ wires, is promising for the large-scale manufacture of IBS conductors.

Katase T et al. 2011 *Nat. Commun.* 2 409

Co doped Ba-122 IBS thin films on bicrystals
Structural defects in polycrystal pnictides

- **cracks** and low density (**porosity**) always lead to poor grain connection
- **impurity phases** (such as Fe-As) that wet the grain boundaries
- inter-grain $J_c$ in polycrystalline IBS was largely suppressed

*Low Temperature Laser Scanning Microscopy (LTLSM) + SEM*

Katase T et al. 2009 *APL* 95 142502

1111-type polycrystal IBS bulks
Grain boundaries in the Sr122 polycrystals are usually coated by impurity amorphous layers (10-30 nm), which show significant oxygen enrichment. These oxygen-rich layers undoubtedly obstructed many grain boundaries, consequently resulting in a poor grain connection.

**STEM-EELS study of 122-type polycrystal IBS bulks**

Wang et al. 2011 *APL* 98 222504

- An amorphous layer
- EELS: electron energy loss spectroscopy
- A high level of oxygen at the boundaries.
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Powder-in-tube method for IBS wires and tapes

Starting powders

- Metall tube
- Packing
- Low density & impurities

Drawing / Groove rolling

- Cracks induced in mechanical deformation

Flat Rolling

- Heat treatment
  - Loss of volatile elements
  - Pores
  - Chemical reaction with metal sheath
The first IBS wire developed in IEECAS

The first 1111-IBS wire in 2008
SmFeAsO$_{1-x}$F$_x$ wire sheathed with Ta
T$_c$ = 52 K, H$_{c2}$ = 120 T
But the transport current can not be measured

Gao 2008 Sust 21 112001

The 122-IBS wire and tape in 2010
Sr$_x$K$_{1-x}$Fe$_2$As$_2$ wire sheath with Ag/Fe
J$_c$, self field = 1200 A/cm$^2$
Using silver sheath, we obtained transport current for the first time.

Wang 2010 Physica C 470 183

- At present, Ag is the most widely used sheath materials for high-J$_c$ IBS wires and tapes since it does not react with IBS cores during heat treatment.
Improve the microstructure of 122-IBS wires and tapes

**in-situ**

![Image](a) Pure

Wang 2010 *Physica C* 470 183

\[ J_c (4.2 \text{ K}, 0 \text{ T}) = 1200 \text{ A/cm}^2 \]

**VS**

**ex-situ**

![Image](a)

Qi 2010 *SuST* 23 055009

\[ J_c (4.2 \text{ K}, 0 \text{ T}) = 3750 \text{ A/cm}^2 \]

Sr-122 wires

- fewer impurity phases
- higher mass density
- better crystallinity

after the *ex-situ* synthesis was proposed, the transport \( J_c \) of 122-IBS increased much more rapidly than 1111-IBS, which still suffers from low purity precursor.
Improve the microstructure of 122-IBS wires and tapes

rolling induced c-axis texture

Sr-122 tape

$J_c (4.2 \text{ K}, 0 \text{ T}) = 5400 \text{ A/cm}^2$

Wang L 2011 *Physica C* 471 1689

rolling texture + Sn addition

$J_c (4.2 \text{ K}, 10 \text{ T}) = 1.7 \times 10^4 \text{ A/cm}^2$

Gao 2012 *Sci.Rep.* 2 998

- grain texture can reduce the high-angle GBs, and suppress the influence of weak-link effect for inter-grain currents
**Improve the microstructure of 122-IBS wires and tapes**

**hot isostatic press (HIP)**

<table>
<thead>
<tr>
<th>Wire Type</th>
<th>Origin</th>
<th>Property Details</th>
<th>Conditions</th>
<th>Additional Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ba-122 round wire</td>
<td>National High Magnetic Field Laboratory, Florida State University</td>
<td>$J_c (4.2 \text{ K}, 10 \text{ T}) = 1 \times 10^4 \text{ A/cm}^2$</td>
<td>192 MPa, 600 °C</td>
<td>highly dense superconducting core with mass density near 100%</td>
</tr>
<tr>
<td>Ba-122 wire</td>
<td>University of Tokyo</td>
<td>$J_c (4.2 \text{ K}, 10 \text{ T}) = 2 \times 10^4 \text{ A/cm}^2$</td>
<td>175 MPa, 700 °C</td>
<td>highly dense superconducting core with mass density near 100%</td>
</tr>
<tr>
<td>Ba-122 wire</td>
<td>IEECAS</td>
<td>$J_c (4.2 \text{ K}, 10 \text{ T}) = 1 \times 10^4 \text{ A/cm}^2$</td>
<td>200 MPa, 700 °C</td>
<td>almost no grain orientation (texture)</td>
</tr>
</tbody>
</table>
Improve the microstructure of 122-IBS wires and tapes

Cold press process

- Cold pressing can largely increase the mass density of 122-IBS phase
- Cracks cannot be completely healed by subsequent heat treatment.

\[ J_c (4.2 \text{ K}, 10 \text{ T}) = 8.6 \times 10^4 \text{ A/cm}^2 \]

Ba-122 tapes made by NIMS, Japan

- Cold pressing can largely increase the mass density of 122-IBS phase
- Cracks cannot be completely healed by subsequent heat treatment.

Gao 2014 Sci. Rep. 4 4465
Improve the microstructure of 122-IBS wires and tapes

hot press process

(Sr-122 tapes by IEECAS)

$J_c (4.2 \text{ K}, 10 \text{ T}) = 1.0 \times 10^5 \text{ A/cm}^2$

$J_c (4.2 \text{ K}, 10 \text{ T}) = 1.2 \times 10^5 \text{ A/cm}^2$

strongly improved c-axis texture and core density, thus greatly improving transport $J_c$

30 MPa, 850~900 °C
Continuously increased $J_c$ for 122-IBS wires and tapes

- **The first IBS wire**
- **Ag sheath**
- **Ex-situ & metal addition**
- **Rolling texture**
- **Hot press**

$J_c$ reached $10^5 \text{A/cm}^2$ for the first time

Practical level desired for application

$10^5 \text{A/cm}^2$ for 122-IBS tape in IEECAS
Recently in IEECAS, a new $J_c$ record was achieved in Ba-122 tapes

- $I_c$ (4.2 K, 10 T) = 437 A
- $J_c$ (4.2 K, 10 T) = $1.5 \times 10^5$ A/cm²
- $J_c$ (4.2 K, 27 T) = $5.5 \times 10^4$ A/cm²
- $J_c$ (20 K, 5 T) = $5.4 \times 10^4$ A/cm²
- $J_c$ anisotropy (4.2 K, 10 T) = 1.37
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Challenges in practical applications

**challenges**

- magnetic flux jumps
- thermal quenching
- AC loss
- device winding damage
- thermal stress
- electromagnetic stress
- material cost
- large-scale production

**strategies**

- Multifilament structure
- Composite sheath instead of silver single sheath
- PIT method

Bi-based wires: Ag/Ag-alloy sheath
IBS wires: Ag/various metal composite sheath is possible

inner sheath: chemical stability
outer sheath: mechanical strength & reduce Ag ratio
Fabrication process for multifilament wires and tapes

The first 122 iron-pnictide multifilamentary wire

Yao et al. 2013 APL 102 082602
7-, 19- & 114-filament Sr-122 wires with Ag/Fe sheath

When increasing the number of filaments and reducing the filament diameter:

- degraded uniformity of mass density for Sr-122 filaments;
- degraded uniformity of interface between Sr-122 filaments and Ag sheath;
7-, 19- & 114-filament Sr-122 wires with Ag/Fe sheath

Gauss fits of particle size inside the Sr-122 filaments

$J_c$ (4.2 K, 10 T): 7-fil: $1.4 \times 10^4$ A/cm²
19-fil: $8.4 \times 10^3$ A/cm²
114-fil: $6.3 \times 10^3$ A/cm²

The refined grains can increase the density of grain boundaries and reduce the degree of grain texture, which are not beneficial to the $J_c$ improvement.

Yao et al. 2015 JAP 118 203909
Advantages of Ag/Monel composite sheath

Monel, any of a group of nickel-copper alloys, first developed in 1905, containing about 66% nickel and 31.5% copper, with small amounts of iron, manganese, carbon, and silicon.

Advantages:
- a melting range of 1300-1350 °C;
- It also has good ductility and thermal conductivity.
- excellent mechanical properties at subzero temperatures, does not undergo a ductile-to-brittle transition even when cooled to the temperature of liquid hydrogen. This is in marked contrast to many ferrous materials which are brittle at low temperatures despite their increased strength.

Typical values of Vickers hardness after annealed at 800~900 °C:

- Pure silver: 30~40; Iron: 90~100; Monel: 150~180

Yao et al. 2015 JAP 118 203909; Yao et al. 2017 SuST 30 075010
Heat treatment temperature up to 850 °C is safe for Ag/Monel sheath, higher than 770 °C for Ag/Cu sheath.

Flat rolled tapes with a thickness down to 0.4 mm can be made.
- For the rolled tapes, the transport $J_c$ gradually grows with the reduction of tape thickness from 0.9 to 0.45 mm.
- For the hot-pressed tapes, a high transport $J_c$ of $3.6 \times 10^4$ A cm$^{-2}$ was achieved at 4.2 K and 10 T.
- For the 0.6 mm thick tapes, the transport $J_c$ decreases with the decline of heat treatment temperature.
- low annealing temperature or large deforming ratio is possible to cause inhomogeneous microstructure for the Sr-122 filaments
- a well-fitted positive semi-logarithmic correlation between the Sr-122 hardness and transport $J_c$
the microstructure of the Sr-122 filaments is well in accordance with their $J_c$ performance
$J_c$-strain relationship of Sr-122/Ag tapes

- The irreversible strain $\varepsilon = 0.25\%$ under tensile stress, comparable to Bi-2212 wire.
- Reversible critical currents under a large compressive strain of $\varepsilon = -0.6\%$ observed for Sr-122/Ag wire; when the applied strain exceeds the irreversible tensile strain limit, the critical current drops rapidly, and a significant crack is found along the sample width.

$I_c$ - tensile strain measurement
Kovac 2015 SuST 28 035007

$I_c$ – strain measurement
Liu 2017 SuST 30 07LT01
The compressive strain dependence of transport $J_c$ and $n$-values for the 0.75 mm tapes is shown in the graph. The graph illustrates that there is almost no $J_c$ degradation under a large compressive strain of 0.6% for Sr-122/Ag/Monel conductors. This finding is promising for large-scale applications in which conductors are usually designed to work under compressive state for safety.

Yao et al. 2017 SuST 30 075010

Almost no $J_c$ degradation under a large compressive strain of 0.6%

Zhou et al. 2014 SuST 27 0750002

Cooperate with Prof. Huajun Liu group in Institute of Plasma Physics, CAS
low-cost copper as sheath for 122-IBS tapes

single copper sheath

copper and thin silver double sheath

\[ J_c (4.2 \text{ K}, 10 \text{ T}) = 3.5 \times 10^4 \text{ A/cm}^2 \]
\[ J_e (4.2 \text{ K}, 10 \text{ T}) = > 10^4 \text{ A/cm}^2 \]

Lin 2016 SuST 29 095006

\[ J_c (4.2 \text{ K}, 10 \text{ T}) = 4.4 \times 10^4 \text{ A/cm}^2 \]
\[ J_c (20 \text{ K}, 10 \text{ T}) = 3.6 \times 10^3 \text{ A/cm}^2 \]

Liu 2017 SuST 30 115007
The first 10-meter class iron-based superconducting wire by scalable rolling process in IEECAS

Ma 2016 Physica C17 516

The average $J_c$ is $1.84 \times 10^4$ A/cm$^2$ for the 11 m long Sr122/Ag wire

The fluctuations of the $J_c$ is $\sim$5%
The first 100-meter class iron-based superconducting wire

made in IEECAS

<table>
<thead>
<tr>
<th>Items</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Structure</td>
<td>Ag-sheathed Sr122 tape</td>
</tr>
<tr>
<td>Length</td>
<td>10 m</td>
</tr>
<tr>
<td>Width</td>
<td>5 mm</td>
</tr>
<tr>
<td>Thickness</td>
<td>0.35 mm</td>
</tr>
<tr>
<td>Matrix</td>
<td>Ag</td>
</tr>
<tr>
<td>Number of filament</td>
<td>1</td>
</tr>
<tr>
<td>Insulation</td>
<td>Mica tape</td>
</tr>
<tr>
<td>Ic @ 4.2 K, 10 T</td>
<td>&gt;100 A</td>
</tr>
</tbody>
</table>

showing a high property and good uniformity

4.2 K, 10 T

$J_c > 12000 \text{A/cm}^2$

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● **Further improving transport $J_c$ for multifilamentary 122 IBS wires**
  find optimized conditions for cold work process and heat treatment
  improve the interface uniformity between IBS filaments and sheath
  improve the microstructure of IBS phase inside filaments

● **Improving the architecture of multifilamentary 122 IBS wires**
  increase the filament number for high-field applications
  increase the engineering critical current density $J_E$

● **Developing long-length 122 IBS wires with composite sheath**
  employ intermediate annealing in the cold-work process to alleviating the deformation hardening effect of sheath
The transport $J_c$ of 122-type iron-based superconducting wire is rapidly increasing, and has surpassed the practical level at 4.2 K and 10 T with a maximum of $1.5 \times 10^5$ A/cm$^2$.

Composite sheath is quite promising for developing high-strength, high-$J_c$ performance and low cost multifilamentary iron-based superconducting wires, which can be strong candidates for high-field application such as IMR, NMR and accelerator.

The world’s first 100-meter class iron-based superconducting wire was achieved in IEECAS, demonstrating the great potential for large-scale manufacture.
Thank you for your attention!