

A Review of Coated Conductor Development

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1. Introduction

The discovery of the cuprates high temperature superconductor of $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ (YBCO) at 90 K in 1986 marked the beginning of a new era not only in the theoretical value in solid state physics in general, but also in the aspect of potential applications in industry. Soon after the discovery, the quest for the practical applications led to the investigation on the current carrying capacity of this new material at the boiling point of liquid nitrogen of 77K. The first experiment on the sintered polycrystalline bulk YBCO samples, known as the first generation HTSC, was rather disappointing, because the large angle grain boundaries severely limited supercurrent flow especially in a magnetic field. With the knowledge that textured cuprates films can carry high current density, the “second generation” HTSC materials became promising in practical applications. The quick success on small single crystal substrates introduced the idea of depositing HTSC film on a flexible inexpensive metal tape substrate with a proper texture in long length. The cost consideration highlights the use of a metal tape together with a cheap film making method, which can be scaled up for commercial applications. The traditional UHV (ultra-high vacuum) thin film vapor deposition techniques are not ideal compared with CSD (chemical solution deposition), because in a practical application the engineering critical current density of a final tape is a crucial factor. Compared with other cuprates (especially BSCCO), YBCO seems to be the most promising material because of its current carrying ability in a magnetic field and high current density in self-field. The use of the RABiTS and IBAD makes textured coated conductor applicable. Nowadays long length YBCO coated conductor is on the way, and people can expect their applications before long.

Two major problems limiting the applications of coated conductors are weak intragrain pinning and weak intergrain coupling [1]. As the origin of a weak intragrain pinning is structural in nature, it is important to choose a material that has strong intrinsic flux pinning capability at a high temperature and in a magnetic field. Despite the relative ease of fabrication, Bi-containing compounds have limited applications in a high magnetic field at 77 K due to their intrinsically low irreversibility fields (B_{irr}) as shown in Fig. 1 [1-3]. The superconductor of MgB_2 is getting hot in the past three years, however its current carrying ability has not been comparable with YBCO coated conductor, so the theoretical importance of MgB_2 is more than the possible applications. Other HTS materials, for example Tl- and Hg-containing materials, though have a relatively high T_c and higher B_{irr} than Bi compounds, low J_c values still remain due to the existence of weak-link dominated intergrain coupling problems [1, 2]. In addition, both Tl and Hg are toxic and their productions usually involve a second step for the incorporation of Tl or Hg into the crystal structure. Thus, ReBCO type compounds remain the choice for coated conductor applications because of their high J_c at 77 K in the presence of high magnetic fields usually required for power applications, as well as their relative ease of fabrication among superconductors with higher B_{irr} .

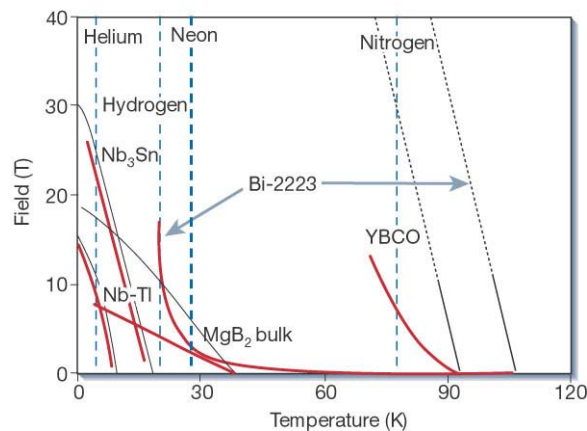


Fig. 1. Magnetic field-temperature diagram for some superconductors (adapted from Larbalestier *et al.* [3]),

black line: upper critical field H_{c2} and red line: irreversibility field B_{irr} .

The driving force for the development of coated conductor is originated from the possible engineering application in magnetic field and theoretical values as well. A high quality and low cost (less than \$50/KA-m) will lead to an industrial-scale commercialization. Factors that must take into account in developing high quality low cost coated conductors are shown in Fig. 2. It is critical that a superconducting layer is deposited on a substrate with both in-plane and out-of-plane texture. Biaxial alignment of the grains is necessary because high angle grain boundaries create weak links that limit current flow as shown in Fig. 3.

Indeed, most of the key techniques in developing ReBCO superconductors are focus on the three critical parts: substrate (as a carrier), buffer layers (as a texture base), and superconductor layer (current carrier). Many traditional techniques such as LPE (liquid phase epi-growth), VD (vapor deposition), and LD (liquid deposition) were introduced both in buffer layer development and YBCO fabrications. Two well established texture generation methods known as the RABiTS (rolling assisted biaxially textured substrate) and IBAD (ion beam assisted deposition) were developed to generating in plane and out-of-plane textures on metallic tape and on the buffer layer itself respectively. The idea in making RABiTS is to form biaxial texture on a metallic tape with cool roll and recrystallization, a thermo-mechanical process. This approach was pioneered by Hitachi on textured Ag tape [4] and by Oak Ridge National Laboratory (ORNL) on textured Ni and Ni-alloy tapes [5, 6]. RABiTS are known as a bulk method because the textured metallic tape can be generated in a fast way and subsequent deposition of textured buffers layer is a simple process of extension of substrate texture to a buffer layer. The architectures of buffer layers have been developed from YSZ single layer to CeO₂-YSZ-CeO₂ multilayers and so on. But in the IBAD approach, an untextured tape requires a thicker buffer layer around 1 μm deposited by a VD method at temperature less than 100°C. Consequently, compared with the RABiTS process, this is a slow process in biaxial texture generating. However, MgO was applied and viewed as a quick way compared with traditional YSZ buffer layers in the IBAD approach [7, 8]. As the RABiTS approach could generate a biaxially textured metallic substrate, a more straightforward method known as *self-oxidation epitaxy* (SOE) was developed by Matsumoto *et al.* [9] in 1998. SOD may have the advantage of easy to generate an oxide layer (buffer layer) with the same texture as the substrate through a controlled oxidation.

Reproducible High quality YBCO film with a current density over 1 MA/cm² can be made routinely through PLD method (an in-situ approach), and through e-beam deposition of Y, BaF₂ and Cu followed by high temperature anneal (an ex-situ method). But one of the non-vacuum MOD approach known as the TFA (trifluoroacetate) method attracts most of the attention recently. The TFA is a CSD approach pioneered by Gupta [10] and optimized by McIntyre and Cima [11]. Recently ASC (American Superconductor Corp.) announced their newly achievement of over 100 Amps per centimeter of width over 10 meters wires by TFA. This is an astonishing news because the wire was made by using a high volume, low-cost manufacturing method and reproducible. This means the TFA method can be expected to scale up for making long length second generation coated conductor wires with excellent electrical performance with a price-performance ratio below that of copper. Another MOD approach, a fluorine-free TMAP[12, 13] method, is being developed and shows promising results on single crystal substrates[14] and on The RABiTS substrates.

Other focus in developing ReBCO coated conductors lie in: thick film development, substrate select, processing monitor, current flow modeling, termination study, buffer layer architecture, buffer layer developing through CSD, grain boundary doping, and other new techniques such as PED *etc.*

2. Substrates for Coated Conductors

2.1 substrate materials and properties

Substrate including buffer layers is the carrier of any form of coated conductors. Lattice match and suitable orientation are prerequisites for the epitaxial growth of a HTSC film. Other fundamental factors that affect the choice of a substrate include thermal expansion coefficients with buffer layer as well as with the superconductor layer[15], chemical compatibility and high-temperature stability, dielectric and magnetic properties, suitable mechanical properties (e.g., ductility and strength), size availability (length, width, and thickness) as well as cost effectiveness for a large-scale production. Substrates of chemistry inert to the HTSC layer are required during the fabrication through a high temperature process.

According to the application, substrates can be classified into two categories: dielectric and metallic substrates. Dissipation factor and dielectric constant as a function of temperature and frequency are of the most critical substrate parameters when coated conductor is applied for the electronic devices in an alternative field. Various dielectric substrate materials are used in microwave, optical, micro-electronic,

and MM-wave components and circuits. Dielectric substrates can be classified as soft and hard. The lowest values of both the dielectric constant and the dissipation factor are necessary to achieve minimum dielectric losses at microwave and MM-wave frequencies. Pure Teflon and PTFE are called soft substrates, which have lower dielectric constant and dissipation factor at room temperature and at low temperature in a wide range of frequency. But these materials are difficult to be used as substrates because they cannot resist a high temperature that is necessary during the fabrication of a ReBCO film. Sapphire, alumina ceramic, fused silica or quartz, strontium titanate (SrTiO_3), magnesium oxide (MgO), yttrium stabilized zirconate (YSZ), and lanthanum aluminate (LaAlO_3) belong to the hard substrate category. These materials have been widely used in the development of superconducting passive and active microwave components and circuits. Also, single crystals like YSZ, SrTiO_3 (STO), LaAlO_3 (LAO), and MgO are used as substrates to demonstrate the properties for various methods. Most of the commonly used substrates are listed in Table 1 and 2.

The magnetic properties of substrates could be important especially in cases where AC losses are of major concern. Metallic substrates with non-magnetic or minimum magnetic hysteresis loss are preferred for the application of superconductor wire in alternative magnetic field. Several factors, notably flexibility and availability in long length, prohibit the use of ceramic substrates for large-scale applications. Metallic substrates have obvious advantages in making long and flexible tapes. Silver (Ag) is an attractive candidate, because textured Ag tape usually does not require a buffer layer for coated conductors. Ag also has non-magnetic and inert and nonpoisonous to ReBCO compounds. However, Ag has its major drawbacks such as its low melting point ($\sim 961^\circ\text{C}$) and extreme softness after annealing. With alloying, higher intensities can be expected, but the trade off is an even lower eutectic temperature. The difficulty to get high quality biaxial textures is another drawback for a silver substrate. Iron and its alloys usually are not considered as the candidate substrates for the coated conductor because most of them belong to a magnetic material and cannot resist oxidation at high temperature during the ReBCO formation as well. Among the metallic materials considered, Ni and Ni-based alloys have very good stability and oxidation resistance at an elevated temperature. High quality biaxial textures have been demonstrated on these materials through the RABiTS technique with cubic percentage even higher than 99%. Different categories of Ni base alloys, such as Ni-Cr and Ni-W, have been developed in the consideration of mechanical and magnetic properties (refers to Table 2). For example, Ni-5at%W alloy though has a Curie temperature of 334K ($>77\text{K}$), the magnetic losses are 0.086W/KA-m at 60Hz, a factor of 5 lower than that of the Ni substrate [16]. For the Ni-Cr alloys, the Curie temperature drops below the boiling point of liquid nitrogen (77K) when the content of Cr is higher than 10at%.

Another potential substrate is copper. Textured copper with FWHMs of 9° and 8° for the rocking curve and phi-scan respectively has been prepared through the RABiTS approach by Jin *et al.* [17]. Recent research at ORNL shows that copper is a promising substrate for the future development of the coated conductor.

The mechanical properties of substrates are important especially in cases where the engineering critical current density J_e , which is defined as current per total cross sectional area of superconductor and sheath, is of a major concern. It is preferable to use a thin (but strong) substrate in order to provide good mechanical support for handling of the conductor in various processing steps during and after the deposition. Typically, $50\mu\text{m}$ thick substrates are commonly used nowadays [18]. But if chose a high strength material, 10-30 μm or even thinner substrates are more preferable. Reasonable high yield strength is necessary because a un-recoverable deformation (ductile deformation) usually damages the buffer and the ReBCO layer and makes the handling more difficult.

It is important to match ($<10\%$, [15]) the thermal expansion coefficients of the substrate with the buffer layer and the superconducting film. According to the present setup, usually the thickness of substrate is in the range of 50-100 μm , much thicker than the YBCO film or even the total ($<3\mu\text{m}$) of the YBCO and the buffer layer, as no big difference of the Young's modules between the substrate and the film, thus, the substrate thermal expansion properties will determine that of the overall wire. From the mechanical point of view, cracking possibility will reduce if a lightly compress stress generated in the YBCO film as well as in a buffer layer upon cooling from a growth temperature. That means an ideal substrate should have a thermal expansion coefficient that matches or is slightly larger than those of films. If keeping alloy content (especially W) in a low level, the thermal expansion coefficient of the currently used substrate is just in a good range as described above.

Table 1. Room temperature properties of the commonly used materials for substrates and buffer layers

Materials	Crystal structure	Lattice constant (nm)	Plane Space d (nm)	Misfit to YBCO (%)	Dielectric Constant ϵ	α ($10^{-6}/^{\circ}\text{C}$)	<i>m.p.</i> ($^{\circ}\text{C}$)
YBa ₂ Cu ₃ O _{6.88}	Orthorhombic	a=0.3817 b=0.3883 c=1.1633	(a+b)/2 =0.385	0		7.9 (11) 16 (33)	1150
Zr _{0.8} Y _{0.2} O _{1.9}	Cubic	0.5147	0.3640	-5.45	27	10.3	2680
Zr _{0.85} Y _{0.15} O _{1.93}	Cubic	0.5139	0.3636	-5.56			2730
CeO ₂	Cubic (CaF ₂)	0.5411	0.3826	-0.62	15	9.9-13.2	2600
SrTiO ₃	c.p.	0.3905	0.3905	+1.43	300	10.4	2080
LaAlO ₃	r.p.	0.3792	0.3792	-1.51	25.4	9.2	2100
LSAT	c.p.	0.3868	0.3868	+0.47	22	10	1840
LaMnO ₃	c.p.	0.3880	0.3880	+0.78			
La ₂ Zr ₂ O ₇	c.p.	1.0786	0.3813	-0.96			
BaZrO ₃	c.p.	0.4193	0.4193	+8.91			
BaCeO ₃	c.p.	0.4377	0.4377	+13.7			
Y ₂ O ₃	Cubic (Mn ₂ O ₃)	1.055	0.3723	-3.30	13	8.5	2410
MgO	Cubic (Al ₂ MgO ₄)	0.4216	0.4216	+9.35	9.8	8.0-12.8	2852
NdGaO ₃	o.p.	a=0.543 b=0.550 c=0.770	0.384 0.389	-0.26 +1.04	25	7.8	1600
NiO	cF8 (NaCl)	0.4177	0.4177	+8.49	~10		1990
Si	CF8 (C)	0.543	0.384	0.27	11-12	3.12	1410
SiO ₂ (Quartz)		a=0.515 c=1.386			3.8	0.59	1720
Al ₂ O ₃ (Sapphire)	Hexagonal	a=0.4758 c=1.299			11.5-9.4	8.31-9.03	2040

Keys: values at room temperature unless otherwise stated;

α : coefficient of thermal expansion

m.p.: melting point;

o.p.: orthorhombic perovskite;

c.p.: cubic perovskite;

r.p.: rhombohedral perovskite;

f.c.c.: face-centred cubic

Table 2. Properties of some of the commonly used metallic substrates

Materials	Crystal structure	Lattice constant (nm)	d (nm)	Misfit to YBCO (%)	Curie T (K)	α ($10^{-6}/^{\circ}\text{C}$)	σ_y (MPa)	<i>m.p.</i> ($^{\circ}\text{C}$)
YBa ₂ Cu ₃ O _{6.88}	Orthorhombic.	a=0.3817 b=0.3883 c=1.1633	(a+b)/2 =0.385	0		7.9(11) 16(33)		1150
Ag	cF4	0.4086	0.4086	+6.13	0	18.9-25		961
Cu	cF4	0.3615	0.3615	-6.10	0	17	75	1083
Ni	cF4	0.3524	0.3524	-8.57	627	13-17.4	59*	1455
Ni-7 at.%Cr	cF4				250		64	~1430
Ni-9 at.%Cr	cF4				124		87	~1430
Ni-11 at.%Cr	cF4				20		102	~1430
Ni-13 at.%Cr	cF4				0		157*	~1430
Ni-V	cF4	0.3520	0.3520	-8.57		11		~1425
Ni-Fe	cF4	0.3590	0.3590	-6.75		~12		~1450
Ni-5%W	cF4				334		254*	
Ni-2%Fe-3%W	cF4					12.9	183*	
Inconel 601	cF4						337	1384
Hastelloy	cF4						360	1370

Keys: values at room temperature unless otherwise stated;

*: denotes values at 76 K[178]

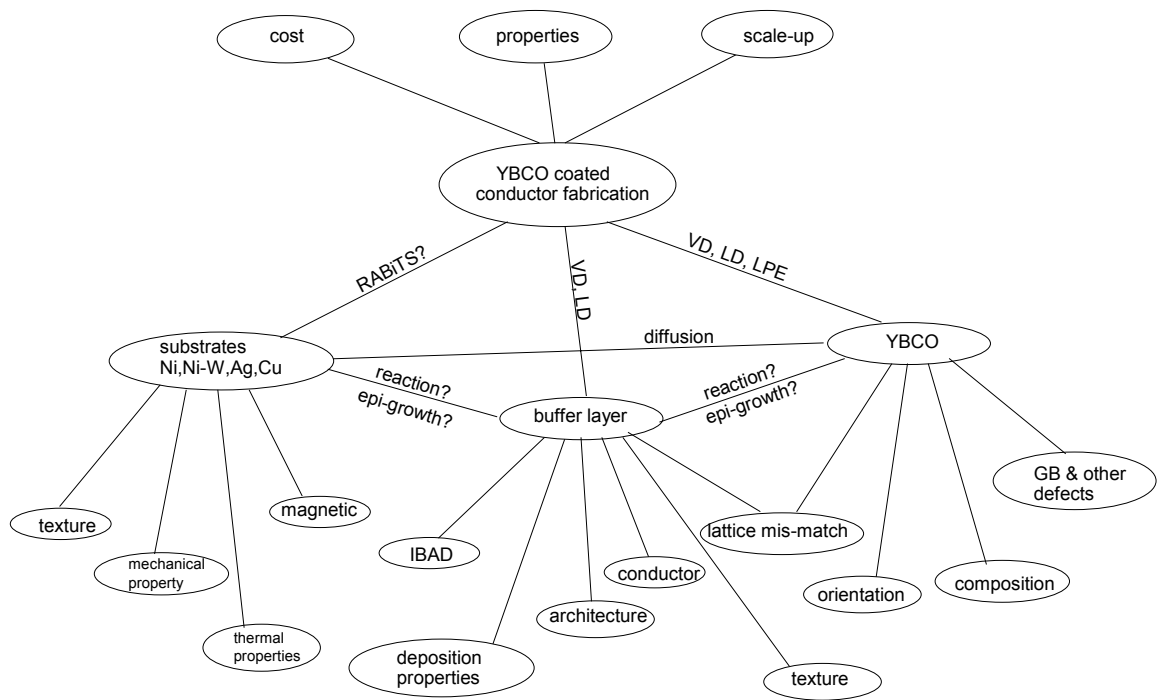


Fig. 2. Factors to be considered in developing coated conductor with high quality and low cost.

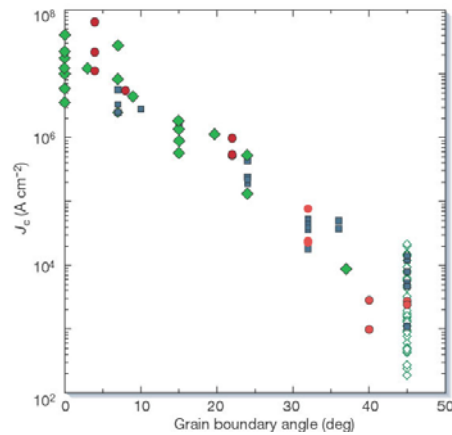


Fig. 3. Transport critical current density measured at 4.2 K or converted from 77 K by multiply a factor of 10.9 in YBCO thin films grown on [001] tilt bicrystal substrates of SrTiO₃ (adapted from reference [2, 3, 20, 21, 174])

Surface quality of substrates such as roughness, cleanliness, and grain boundary grooving play a significant role for the buffer and the YBCO growth. As a Ni-based RABiTS substrate was formed through cool deformation and high temperature anneal (>800°C), high grain boundary energy usually will generate thermal grooving at GB as shown in Fig. 4. Preliminary experimental data have not shown adversely affect of a GB grooving on J_c [18]. The experimental results of Eickemeyer [19] show that micro alloying reduced the susceptibility of a substrate material against grain boundary grooving at a processing temperature. Highly alloyed nickel with 5at% tungsten was not susceptible to thermal etching effects independent of the processing route, i.e. melting or powder metallurgy. Although the surface roughness and

cleanliness effects have not yet been fully studied, it is likely that any surface residue such as lubricant from the rolling process will interfere with the film adhesion or the desired epitaxial growth.

In following the idea in the BSCCO wire, conductive buffer layers are preferred as they can provide a current by-pass in case a local failure in a HTS film. This would improve electrical stability because current would be able to transfer from the superconductor to the conductor layer, hence, avoiding the creation of hot spots that could be detrimental. However most of the commonly used buffer layers are oxide and insulator, very limited current can be transferred through the buffer layer to the metallic substrate, as an alternative, a silver layer was coated on the HTS film, which provides a measure of stability, and protect the ReBCO layer as well.

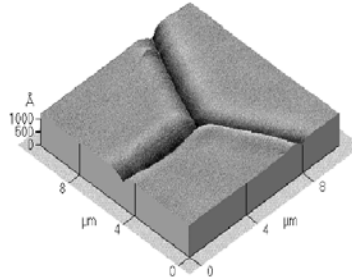


Fig. 4. AFM image showing the a thermal grooving in Ni grain boundary annealed at 800 °C (adapted from Goyal [18])

2.2 Textured metallic substrate by RABiTS

The shorter coherence length in nature for ReBCO HTSC needs a “perfect texture like single crystal” for the high current density, which bring in the practical difficulties especially for a metal polycrystalline substrate for long length applications. According to the early experimental data of YBCO on bicrystal STO [20, 21], when misorientation angles are less than 4-6°, the influence of the grain boundaries on the current density is minor, however when the misorientation is higher than ten degrees, a drop of current density exponentially with the angles between grains has been observed (see Fig. 3). As ReBCO belongs to the perovskite with a layered structure, the transport current in *a-b* plane is much higher than that in the *c*-axis. To make high current density films, *c*-axis oriented film (out of plane texture) with *a-b* in plane texture is a must. In order to deposit ReBCO with (001) orientation, the most suitable texture for a substrate with a cubic symmetry is the cube texture $\{100\}\langle 001\rangle$. The RABiTS technique involves a deformation of metal ingot via a rolling and followed by an annealing to develop the desired texture. Hence, the basic requirement for the RABiTS materials is that a metal can generate a desired texture by the rolling and recrystallization process. Pure Ni is easily deformed and has a higher oxidation resistance, other properties like crystallographic, physical and chemical properties make Ni and its alloys suitable substrates. The RABiTS Ni was firstly developed by Goyal *et al.* at ORNL [22] with a sharp cubic texture. Biaxially textured Ni tapes are now available in hundreds of meters in length. However, Ni is ferromagnetic at 77K ($T_c=627K$) and also its tensile strength is low (< 60 MPa), these problems have to be overcome for Ni be used as a coated conductor substrate. de Boer and Reger *et al.* [23] studied most of the possible alloying elements based on their solid solubility in Ni, costs, ability to reduce the Curie temperatures below 77 K, as well as the ability to form sharp cubic textures and retain a FCC structure. They concluded that Cr, V, and Cu were the three alloying elements that would form a cube texture as sharp as pure Ni. In addition, they also showed that the V and especially Cr alloying could reduce the Curie temperature and improve the tensile strength. Hence, these alloys remain the popular substrates under investigations [24, 25]. Ni-W and Ni-Cr textured tapes with improved yield strength and reduced magnetism have also been explored by ORNL [18, 26] and by Eickmeyer *et al.* [27]. Ni-Fe alloy has been reported with a sharp cube texture and kilometers long tape is now commercially available [28-33]. However, Ni-Fe alloy is not commonly used as a substrate for a coated conductor because of its poor oxidation resistance. A ternary alloy, Ni-2%Fe-3%W, with improved yield strength, magnetic properties, and oxygen resistance, have been developed and used in ORNL [34]. Other Ni-based ternary alloys like Ni-Cr-W and Ni-Cr-V were also studied by Tuissi *et al.* [35].

Another design of the RABiTS, known as a clad-tape composite, has been developed in Japan by inserting a core metal rod into a Ni pipe or sandwiching a core metal sheet between two Ag plates and subsequently cold rolled and annealed as in the RABiTS process. The core metal is usually stronger and/or

has better magnetic properties in order to improve the performance without losing the biaxial texture of the original tape on the outer layer [36].

Because silver has the advantages of being a good electrical conductor and non-poisonous to HTS materials (which may result in the elimination of the additional buffer layers deposition and cut down the total cost), though with several drawbacks including low melting point, high raw material cost, and poor mechanical properties, Ag is still one of the most promising substrates. An intensive research on Ag has been conducted recently [37-41] including a 100-m long of Ag-0.1wt%Cu/Ag-10wt%Ni/Ag-0.1wt%Cu clad tape with improved strength developed in Toshiba Corp [42, 43]. YBCO was deposited by PLD on a non-buffered 1 m of Ag tape has been reported with an in-plane FWHM of 23° and an average transport J_c 0.12 MA/cm². Suo *et al.* [44] used AgMg, Ni, or Ni alloys as a reinforced core for Ag tapes and succeeded in producing {110}<001> Ag tapes with a Ni core having in-plane FWHM of 10-15°.

The RABiTS of Ag usually has the texture {100}<001>, {100}<011>, {110}<110>, and {110}<001>. As the lattice constant of Ag (0.4086nm) is larger than that of YBCO, a mismatch of 6.13% for YBCO grown on {100}<001> Ag texture. For other textures the mismatch may even larger. However, Doi *et al.* [45] reported that out of the four possible cubic Ag textures, NdBCO would only grow on {100}<001> texture while YBCO would only grow on {110}<110>.

Cu-based tapes have been proposed as substrates for coated conductors [22, 46]. Such tapes could also have the potential advantage over Ni of being inexpensive and non-magnetic. But the primary disadvantage is that Cu is much more susceptible to oxidation. Jin *et al.* [17] investigated {001}<001> texture development during annealing for a 50µm thick cool rolled copper tape. Rutter *et al.* studied the Ni overlayers on biaxially textured Cu substrate by D.C. sputtering [47] and found a good texture of pure copper was extended to the overlayer of Ni with 98% cubic phase when annealed at 400-500°C. Such a method has been shown to reduce the oxidation rate by as much as a factor of 20 for relatively thick nickel films plated onto copper tapes [48].

Table 3. Composition and mechanical properties of some of the commercially available alloys used for IBAD substrate[179] (materials are in the cold-worked state except where specified otherwise) [180]

Alloy	Composition (wt%)						σ_y (MPa)	σ_{UTS} (MPa)
	Ni	Fe	Cr	Mo	Co	Al		
Hastelloy C-275	60	5	15	16			360	790
Hastelloy X	52	18	21	9			350	800
Inconel	60.5	15.1	23			1.4	440	760
SS 304	8	72	18					550
Rene 41	55.4		19	11	11	1.5	1069	1420
Inconel 625	66		21.5	9			520	930

2.3 Untextured metallic substrates

The IBAD technique can generate texture on a no textured polycrystalline metallic substrate or even on an amorphous substrate. The range of potential materials is rather large, because there is not requirement for the substrate to have a lattice match and neither a textured substrate with proper orientation, however highly oxidation resist materials are necessary. Iijima initially started inclined substrate deposition on Hastelloy [49] and it became one of the popular choices of substrate [50, 51]. Commercially available polycrystalline metallic substrates including various Ni-based alloys, Hastelloy X, Inconel 601, Rene 41, and stainless steel SS304 were investigated by Yin *et al.* [37], other commercial alloys such as Inconel 625 [8, 52] was also studied. The compositions of these materials are listed in Table 3.

3. Buffer Layers

Buffer layers are functioned as a texture base and a reaction barrier between the YBCO and the substrates. The nature of a buffer layer will depend on whether the route is the RABiTS or the IBAD. In the RABiTS route, the texture are formed on a metallic substrate, hence the buffer layer act more as a barrier: preventing diffusion of metal atoms up into the superconductor, as well as prevent oxygen diffusing down through to the substrate to cause oxidation of the metal. In order to be an effective barrier, it is important that the buffer layer is free of cracks, which usually caused by stress induced through differential thermal contractions. Thus, a buffer material having a closely matched thermal expansion coefficient with the substrate and superconductor is desired. The basic requirements for a buffer layer are to be biaxially

textured with suitable lattice parameters and non-reactive with a HTS layer. The crystallographic orientation of a buffer generally requires a {100} cubic texture. Both metallic (e.g., Ag, Pd) and ceramic (e.g., YSZ, CeO₂) materials have been used as buffer layers. In the IBAD approach, one of the key functions of a buffer layer is to generate a texture on a randomly oriented polycrystalline metallic substrate. The buffer layer should also be a good barrier, and defect free as well. An additional property, which may be desirable, is for a buffer to have a high electrical conductivity, so that currents may be transferred through it into the metallic substrate.

3.1 IBAD buffer

The IBAD is a method characterized by an assisting ion beam concurrently bombarding films during growth. The technique involves the use of O and Ar ion beam to control the orientation of a film growth. When the incident beam axis is tilted from the substrate normal, the assisting ions should have a planar component of momentum, which could impart azimuthal anisotropy to the growing film. In 1985, Yu *et al.* first demonstrated Nb thin films biaxially aligned on a fused silica, with [110] fiber texture and azimuthal ordering, by using off-normal IBAD [53, 54]. Bradley *et al.* constructed the theory of biaxial alignment and expected the azimuthal ordering to be improved by adjusting the beam axis on an ion-channeling axis of the film [55]. In 1991, Iijima *et al.* found a sharp biaxially aligned structure of YSZ thin films on polycrystalline Ni-based alloy can be developed by an off-normal IBAD method [49, 56]. It was successfully applied for in-plane texturing control of YBCO films, and demonstrated high J_c values [57]. YSZ was the focus in the early studies of the IBAD buffer, since its lattice parameters have a reasonable match with YBCO. The IBAD YSZ process develops texture by an evolutionary grain growth competition. This process requires between 500 and 1000 nm of material to achieve a good in-plane texture (ϕ scan full-width-at-halfmaximum (FWHM) of 12°) [50, 58]. The group in LANL (Las Alamos National Laboratory) optimized the IBAD YSZ process and made meter-long coated conductors with critical current densities, J_c , of 1 MA/cm² (75 K, SF), or better, for YBCO films thicker than 1 μm [59]. However, it usually took 20 h to deposit 1 μm for one-meter long tape. So measures must be taken to accelerate this process. Wang *et al.* [7] found that a comparable in-plane texture could be achieved by using magnesium oxide (MgO) as the source material in the IBAD process. They showed that a IBAD MgO requires only ~10 nm to develop the texture comparable to that of 1 μm thick IBAD YSZ layer. In essence, this translates to a process, which is about 100 times faster than the IBAD YSZ. It was believed that the thin MgO IBAD layer was not a sufficient barrier to prevent diffusion of cations from the substrate into the YBCO where chemical reactions can degrade superconducting performances. The architectures of IBAD YSZ and MgO, interface structure, and epitaxial growth feature of YBCO film are shown in Fig. 5. Groves *et al.* [60] reported a further YSZ layer on MgO by PLD to increase the superconducting performance of YBCO films on IBAD-MgO templates. More recently a 100 nm MgO buffer by magnetron sputtering was applied on a 10 nm IBAD MgO template. A combination of improvements in processing of these small area samples resulted in a J_c over 1 MA/cm² for a YBCO film with a thickness over 1.5 μm .

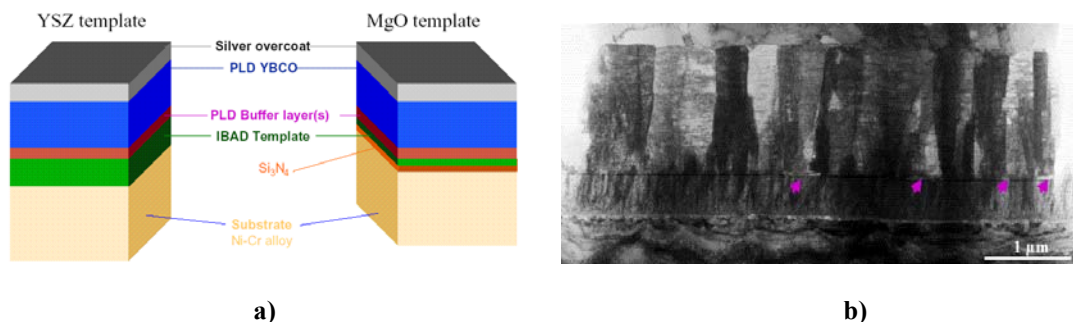


Fig. 5. a) Architecture of IBAD YSZ/MgO and PLD YBCO on Inconel substrate, b) Microstructure demonstrate the epitaxial growth of YBCO film on IBAD YSZ buffer with CeO₂ cap layer (adapted from LANL web-site)

Due to its close lattice matching with YBCO (<1%), CeO₂ has been investigated via the IBAD route but without much success [61]. Nevertheless, a more successful route is using CeO₂ as a cap layer to improve the lattice matching between IBAD-YSZ and YBCO. Holesinger *et al.* [62] showed that by adding

a 30 nm CeO₂ cap layer onto 700 nm IBADYSZ, the J_c improved from 0.6 to 1 MA/cm² with an overall critical current improvement from 100 A to 200 A. They also demonstrated that a cap layer that is too thick would reduce J_c , as CeO₂ grows thicker, it becomes rougher and more porous. Despite the intensive research and positive results via the IBAD route, the technique has a major drawback as being a vacuum process that may not be cost effective for an industrial scale production.

3.2 Inclined substrate deposition

Similar to IBAD, a method by the name of *inclined substrate deposition* (ISD) was firstly reported in 1996 [63]. By using an off-axis pulsed laser ablation, ISD leads to in-plane alignment of YSZ films on untextured substrates without ion beam assistance. Compared with IBAD, ISD can have a deposition rate of 0.5 μm/min faster than the IBAD approach. Metzger *et al.* [64] reported the MgO buffer by the ISD route with a deposition rate of 120~150 nm/min on Hastelloy C276 substrates. A 7 degrees FWHM of YBCO in plane texture and a critical current of 84 A for a 8 mm width and 1.8 μm YBCO film, which means a current density of 0.58 MA/cm². Sumitomo Electric[65] recently reported a 10-m long YSZ-buffered Ni-alloy tape prepared by reel-to-reel ISD with a J_c of 0.05-0.2 MA/cm² throughout the tape. The tape moving speed was 1.2 m/h and the HTS deposition rate reportedly could be raised to 3 μm/min by using an industrial scale 200 W, 660 mJ laser at 150 Hz. However, the J_c of the 10-m long tape dropped below 10⁵ A/cm² at positions corresponding to target changing, thus, reducing the "effective" length to about 4-m pieces. So far the YBCO films on the ISD buffered metallic tape with a critical current density over 1 MA/cm² have not been reported, But it offers the potential for rapid production of high-quality biaxially textured buffer layers suitable for YBCO coated conductors [66].

3.3 Ceramic buffer on RABiTS

Because of the reaction and mismatch between a YBCO film and a Ni-based substrate, a buffer layer is an effect way to bypass these difficulties. In the early days notable metal buffers attracted a lot attentions because they are easy to deposit and have a good resistance to oxidation. But the lattice mismatch and poor texture had driven the research on the direction of ceramic buffer with a better lattice match and a high resistance to oxidation. As in the IBAD and ISD, YSZ was firstly selected as the ceramic buffer. Then in 1991 Wu *et al.* [67] reported a better lattice match material CeO₂ as the buffer layer epitaxially grown on single crystal of LaAlO₃, sapphire, and yttria-stabilized zirconia using pulsed laser deposition. It is demonstrated that a CeO₂ film are chemically and structurally compatible to the high-temperature superconductor YBCO film. In 1996 Goyal *et al.* [5] demonstrated for the first time a high current density (0.9MA/cm²) YBCO film deposited by laser ablation on Ni RABiTS substrates with CeO₂ and YSZ buffer layers. The strong biaxial texture of the metal (in-plane 6-7 degrees FWHM) is conferred to the superconductor by deposition of intermediate oxide layers, which serve both as a chemical and as a structural buffer. Since then CeO₂ has been widely used as a buffer layer in various single crystalline and metallic substrates. In 1997, Paranthaman *et al.* [68] and He *et al.* [69] firstly showed that an epitaxial CeO₂ layer cracked extensively when its thickness exceed 50 nm. Nevertheless, problems associated with CeO₂ cracking may be overcome by depositing a layer of YSZ on top [6]. Thus Ni/CeO₂/YSZ/YBCO became a standard architecture in early-coated conductor studies. Since YSZ does not have good lattice match with YBCO, a second CeO₂ layer, the cap layer, is added on top of the YSZ. The topmost CeO₂ layer is sometimes substituted with a Re₂O₃ layer, which has relatively good lattice match with YBCO. The preferable buffer architecture now appears to be Ni-alloy/CeO₂/YSZ/CeO₂/YBCO. In addition, Takahashi *et al.* [70] showed that CeO₂ could also be grown on a pure Ag sample almost without any (111) growth component.

On the set of RE oxides (Re₂O₃) buffer candidates, ORNL leads the studies where they have investigated a wide range of oxides including Y₂O₃, Yb₂O₃, Gd₂O₃, and Eu₂O₃ [71-73]. They deposited the Re₂O₃ layer via either vacuum or solution techniques such as sol-gel and spin or dip coating and capped it with either YSZ and/or CeO₂ layers via vacuum process. The best RE oxide for buffer layers reported is Yb₂O₃, possibly due to its close lattice match with Ni. Buffer layers deposited via a sol-gel process tend to produce lower J_c values (0.2 MA/cm²) than those made by vacuum techniques (over 1 MA/cm²) due to high porosity.

Another promising buffer layer is Y₂O₃, firstly reported in 1992 by Bardal *et al.* [74] and Prusseit *et al.* [75] by reactive thermal co-evaporation, grown on Si substrates with an architecture of Si/YSZ/Y₂O₃/YBCO. Recently, Tomov *et al.* [76] had successfully deposited the CeO₂/YSZ/Y₂O₃ buffer architecture without the need of a forming gas on a newly developed NiCrW ternary alloy tape. The in-

plane FWHM was 10° and they could completely eliminate the (111) growth component. Ideally, the best process would be one that requires minimum or no buffer layers. With a lattice parameter in between Ni and YBCO, Y_2O_3 has been studied as a possible single-buffer material by Ichinose *et al.* [77-79] using electron-beam (e-beam) evaporation. Well-textured Y_2O_3 with in-plane and out-of-plane FWHM values of 11° and 3° respectively are reported. A relatively smooth film was produced but there is the evidence of the (111) growth component present at grain boundaries, which could weaken the intergrain coupling.

In 2000, Chirayil and Paranthaman *et al.* [80] reported the YBCO films deposited on the sol-gel LZO-buffered Ni substrates with sputtered YSZ and CeO_2 top layers had a critical current density of $480,000 A/cm^2$ at 77 K and self-field. Two years later, Sathyamurthy and Paranthaman [34] reported Critical current densities, J_c , of $1.9 MA/cm^2$ at 77 K and self-field and $0.34 MA/cm^2$ at 77 K and 0.5 T obtained on these films for the same set of buffer layer. Paranthaman also reported Gd_2O_3 buffer on which a current density of $1.2 \times 10^6 MA/cm^2$ of a YBCO film had been reached. But the problem is in these processes an extra buffer layer like YSZ and/or CeO_2 were needed which were fabricated through a high cost vacuum approach. Single, epitaxial buffer layers of insulating $LaMnO_3$ (LMO) or conductive $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) have been grown through sputter deposition on biaxially textured Ni and Ni-alloy substrates by Aytug *et al.* [81]. A critical current density, J_c , exceeding $1 \times 10^6 A/cm^2$ at 77 K has been demonstrated on a 200 nm YBCO film on LMO-buffered Ni-RABiTS. This result offers a prospect for the use of single LMO-buffered metal tapes in the development of YBCO-coated conductors. There are many other ceramic buffer materials being considered including $LaNiO_3$ [82-85], $Ni/LaNiO_3/SrRuO_3$ [86, 87], and $NdGaO_3$ [88]; however, no significant breakthrough has been made in terms of quality, speed, and scalability. An ideal single buffer through MOD approach (non-vacuum) with a good electrical properties of the YBCO films had not been reported. The technique difficulties including the porosity, rough surface, and biaxial texture may be the main frustration for the development of single-buffers. During the fabrication of buffers, multi-layers and multiple steps including vacuum and/or heat treatment processes would inevitably slow down the overall production speed.

3.4 Metallic buffer on RABiTS

The ductility of the buffer layer could be important especially in the case where susceptible to cracking upon thermal cycling or mechanical deformations are of the major concern. Metallic buffer layers are preferred as they can provide a current by-pass in case a local hot spot in the HTS film, hence, avoiding the failure in the system. For the oxidation resistance and non-magnetic consideration, noble metals such as Au, Ag, Pt, Pd *et al.* are attractive candidates. Compare with other noble metals Ag has the advantages of least expensive, inert to oxygen, and nonpoisonous to ReBCO compounds. Due to the large lattice mismatch ($\sim 6.13\%$), an intermediate layer with a lattice parameter between that of Ag and Ni (for example: Pt or Pd) may be necessary for the texture development of the Ag layer [28, 89]. Recently, Ag as a buffer layer deposited on a Ni-based RABiTS had been reported by Lee *et al.* [90]. Goodall reported an easily scalable technique of electro-epitaxial technique for a cube-textured silver buffer deposition [91]. Continuous 150 nm thick silver films with in-plane FWHM values of 8 degrees was epitaxially deposited directly onto cube-textured Ni, Ni-10 wt% Cr and Ni-Fe substrates using no intermediate buffer layer and no post-deposition heat treatment.

3.5 Self-oxidation epitaxy

How to make the ceramic buffer layer simpler is a key for the scale-up and cost effective. An ideal solution to simplify a buffer would be generating an oxide buffer through a controlled oxidation of a textured substrate and form a native oxide layer with a proper texture that functions as a buffer layer. In 1991, Ginsbach *et al.* [92] tested this idea by oxidize Ni sheets to form NiO. Nevertheless, they failed in demonstrating that the YBCO could be grown on NiO single crystals and estimated that a NiO layer of $\sim 10 \mu m$ thick is required to prevent Ni diffusion through the layer. In 1999, Matsumoto *et al.* [9] successfully grew an epitaxial oxide on a rolled textured Ni substrate, a procedure they termed as *self-oxidation epitaxy* (SOE) buffer. It was found that (100) NiO become dominant at an elevated temperature above $1000^\circ C$ in air. The preferred growth orientations of SOE NiO on (100) Ni are (100) NiO and (111) NiO [93]. The Initial experiments had not shown reasonable J_c (less than $0.1 MA/cm^2$) until relatively recently, when Matsumoto *et al.* added a thin oxide cap layer (MgO , YSZ or CeO_2) between the NiO and YBCO and improved the J_c to $0.3 MA/cm^2$. It was suggested that the relatively low J_c might be caused by Ni inter-diffusions[9] or perhaps because of the rough nature of the NiO surface. Usually the texture of the SOE NiO buffer layer is somewhat poorer than that of the underlying substrate, having an in-plane FWHM of

14°, and cannot comparable with those ceramic buffer deposited by PVD. Watanabe *et al.* [36] reported a 30-m long SOE NiO buffer on their clad-tape Ni/NiCr and Ni/SS tapes with the NiO in-plane FWHM of 19°; however, only a low J_c value of 10^4 A/cm² was obtained. Boffa *et al.* [94] have developed a cube textured NiO layer by oxidizing NiV in a 10 mTorr pure O₂ at 700°C for 30 mins. The J_c of the YBCO deposited on a CeO₂ cap layer was 0.6 MA/cm². Rutter *et al.* [95] used an additional step where the uncontrolled surface oxide on the NiFe tape was first removed before SOE was subsequently carried out, yielding a cube textured iron nickel oxide layer with a FWHM of 8°. Watanabe[36], used the SOE technique and made A 50-m long, 3µm, biaxially textured NiO buffer layer at a speed of 15 m/h with in-plane texture 13 to 19 degrees in the whole length, by using thin MgO cap layer with the thickness of about 100 nm, a J_c of 0.3 MA/cm² at 77K and self field was obtained in short samples of YBCO films on NiO/Ni tapes.

Recently, Matsumoto [96] focused his research on The surface quality by polishing the NiO buffer produced by the SOE, a critical current density, J_c , for the YBCO film directly deposited on the SOE-NiO buffer by pulsed laser deposition method was 0.17 MA/cm² at 77 K. it was believed that the number of superconductive weak coupling in the YBCO film was reduced by the flattening of the NiO surface. In addition, Matsumoto deposited BaSnO₃ as a cap layer to improve the crystal orientation of the YBCO film grown on SOE-NiO. As a result, J_c of the YBCO film formed on BaSnO₃/NiO/Ni substrate reached 0.45 MA/cm² at 77 K.

Further work is required for SOE buffer layer to reach the level of performance of other buffer materials. Several researchers have also been working on Ni-alloy for the SOE buffer development.

4. Superconducting Layer

A textured substrate obtained from either the RABiTS process or the IBAD process provides a starting material over which the epitaxial layer of YBCO (HTS) can be applied using various candidate options. Among these options, PLD, RF-sputter, PED, e-beam based deposition and electrophoresis are physical deposition processes, and MOCVD, sol-gel, CVD, aerosol/spray pyrolysis and MOD are chemical deposition methods. In addition, PLD, PED, sputter and e-beam based deposition are the processes that can result in a thin/thick film of YBCO in the substrate and require no additional post thermal treatment, called in-situ methods; and The other processes such as e-beam Y-BaF₂-Cu, MOCVD, sol-gel, CVD, aerosol/spray pyrolysis, MOD, electrode-position and electro-phoresis all require post thermal treatment to complete the crystallization and oxidation of the deposited films to the desired YBCO phase, called ex-situ methods. Among these methods, PLD, e-beam Y-BaF₂-Cu, and MOD/sol-gel are most prominent and popular for the ReBCO coated conductor development.

4.1 In-situ methods

Most of the in-situ methods are originally from thin film depositions usually utilize physical vapor deposition (PVD) techniques where the film grows at an atomic-scale increment. Examples of these processes include pulsed laser ablation, sputter, and e-beam evaporation *etc.* The early work was well established on the optimizing of the deposition conditions such as the deposition temperature, oxygen partial pressure, as well as the setting up of the YBCO phase diagram [97]. High quality YBCO films can now be produced. Based on thin film technologies (PVD), very high critical current densities over 1000,000 A/cm² (77K self-field) of YBCO coated conductors have been achieved routinely in many groups in the world. However, several problems frustrate these in-situ techniques to be used for the coated conductor applications. First, there is a widespread concern that such vapor phase techniques may not be financially viable for many applications mainly due to its high capital cost of high vacuum system used in these processes; second, in-situ routes are of slow processing rates due to the atomic-scale increment in thickness. The deposition is confined to very limited area because of the chamber size and local conditions (T and PO₂) *etc.*, this may cause difficulties in scaling-up for a production. Another problem is that there exists an upper limit to the film thickness over which the desired *c*-axis oriented epitaxial growth cannot be properly maintained [98, 99], though there are occasionally report on thick film with current density over 10^6 A/cm². Other concerns like only a small fraction of target materials be used for the film deposition because of the uniform evaporation in the chamber may result in an over consumption of target materials, consequently, the change of target may introduce extra problems for a long length tape as well. Though with the difficulties, big progress have been made recently, Peterson [100] from Los Alamos National Laboratory reported a 600A/cm for shorted sample and an over 200A/cm for 1 meter long tape by PLD YBCO on IBAD MgO substrates. Freyhardt [101] from University of Goettingen (Germany) got a critical

current of 223 A/cm over a 10-meter length with a current density exceed $2 \times 10^6/\text{cm}^2$ by the PLD YBCO on the IBAD-YSZ tape.

4.2 Ex-situ methods

Ex-situ methods attract a lot of attentions recently. Compare with in-situ approaches, ex-situ methods have only been developed in recent years, it was originally developed in the consideration of simple operation, low cost and easy to scale up. Most of the ex-situ approaches are viewed as thick film processes and are not very well established. However, there are breakthroughs on ex-situ BaF_2 process, MOD-TFA and MOCVD method, other routes are booming for example, ultrasonic spray pyrolysis and non-fluorine sol-gel approaches[14] *etc.* The LPE had been viewed as one of the most promising routes for high-rate growth of ReBCO, as it is a one-step process capable of growing films at high growth rates. While the relatively low current density and high processing temperatures, which may result in over oxidation, interface reaction and ruin the buffer layer, make this method step out in the competition. Usually ex-situ routes involve two processing steps whereby a precursor layer is first deposited either by ultrasonic spray, spin or dip coating, or the *ex-situ* BaF_2 process *etc.* The precursor layer is subsequently heat treated to convert to a crystallized ReBCO layer in order to obtain the desired biaxial texture. In the *ex-situ* BaF_2 process, the precursor film is co-evaporated by e-beam from Y, Cu, and BaF_2 sources, followed by an *ex-situ* heat treatment under a controlled humidified atmosphere. The exact mechanism of the heat treatment process is still under investigation; however, Solovyov *et al.* [102] proposed that the metal-oxifluoride precursor decomposes as the temperature increase in the presence of H_2O vapor while releasing HF, and the decomposed precursor product is converted into a Y123 phase. Hence, it is important for the conversion process to go to a completion, which is generally a straightforward process for short samples. However, problems arise for longer samples where non-uniform YBCO conversion is reported [103]; this is thought to be due to the built-up of HF to a saturation level, leaving part of the precursor film un-reacted. Recently, Lee [103] from ORNL reported 90A/cm, one meter long with $0.82 \mu\text{m}$ YBCO film on RABiTS and Feenstra *et al.* [104] reported 270A/cm-w for 3-meter long and $3 \mu\text{m}$ YBCO film on RABiTS.

Actually, Ex-situ CSD approaches were developed soon after the discovery of YBCO THSC in 1986. In the early time, there were three commonly used solutions applied for the ReBCO deposition[105-108]: 1) sol-gel processes that used 2-methoxyethanol as a reactant and solvent; 2) hybrid processes that use chelating agents such as acetylacetonates or diethanolamine to reduce alkoxide reactivity and 3) metal organic decomposition (MOD) techniques that use high-molecular-weight precursors and water-insensitive carboxylates, 2-ethyl-hexanoates, *etc.* These CSD routes have been used to grow both oxide buffer layers and superconductor films. The early research failed in developing high current density films and people believe that the carbon-contained precursors might result in the formation of stable BaCO_3 at the grain boundaries [109]. In 1988, Gupta [10] firstly reported a method by using metal trifluoroacetate precursors, called TFA method, to generate textured YBCO films on crystal substrates, McIntyre and Cima[11] further developed this method and high current density were achieved on thin YBCO films. The use of TFA salts appears to avoid the formation of the BaCO_3 because the stability of barium fluoride is believed greater than that of barium carbonate and fluorine can be removed during the high temperature anneal ($>650^\circ\text{C}$) in a humid, low oxygen partial pressure environment [110]. Now Metalorganic deposition using trifluoroacetate salt (TFA-MOD) is a better known and well established sol-gel route attempted by several groups towards coated conductor fabrication [10, 111-113]. Spin or dip coating is usually used to deposit the precursors that are then calcined under various O_2 partial pressures. High J_c values ranging from 6.7 MA/cm^2 to 7.5 MA/cm^2 on single crystalline LaAlO_3 substrates and $1.7\text{-}2.5 \text{ MA/cm}^2$ on the IBAD-YSZ and on Hastelloy [114] have been reported. Nonetheless, all these high J_c films were very thin, ranging from 85 nm to 180 nm, resulting in a low J_e . Even with a recent result on a 3-cm long Hastelloy tape with J_c of 2.5 MA/cm^2 , where the YBCO film was only 230 nm thick. A real breakthrough attributed to the researchers in American Superconductor Corporation (ASC), Malozemoff *et al.* [115] reported one meter long YBCO film with thickness over $1 \mu\text{m}$ and a current density over 1 MA/cm^2 . More recently ASC announced more than 10 meters long reproducible YBCO coated conductor wire with over 100 Amps per centimeter of width with $1 \mu\text{m}$ thick YBCO film. These results are comparable with some vapor deposition approaches on metallic substrates.

MOCVD is another promising ex-situ approach; it offers several advantages for the large-volume production of coated conductors. Precursors are maintained outside the vacuum chamber and therefore refill is simple during a long deposition cycle. The deposition zone can be unlimited size since it can be configured to be as long and as wide as the showerhead, which is extremely beneficial for a high output.

Recently Lee *et al.* [116] reported a meter long YBCO coated conductor wire by MOCVD with over 90 Amps per centimeter of width and a current density over 1.2 MA/cm^2 on a IBAD substrate.

Recently xu *et al.* [12, 14] and Shi *et al.* [13, 117] restarted the non-fluorine MOD research for the YBCO coated conductor development, preliminary results show that in a humid low oxygen atmosphere, carbon contain precursor can be converted to high quality YBCO film with ideal textures and good electrical properties ($T_c=90\text{K}$, $J_c=1.6 \text{ MA/cm}^2$) on single crystal substrates. The results show importance both in theoretical and in practical applications, because a re-assess of the function of carbon in precursor and/or in a YBCO film bring out the topic: how much is the carbon contaminatate in a YBCO film, the compare of the function of C with F during the conversion of precursor films; and also a most important being that the removal of fluorine in the form of HF at high temperatures in TFA approach is a non-trivial process, There appear to be many issues related to fluid-flow and complicated reactor designs may be required for the scale-up.

Ultrasonic spray pyrolysis is a method whereby a nitrate or a carboxylate precursor solution of the superconducting constituents is sprayed onto a substrate at an elevated temperature [118-120]. Film composition is controlled empirically by adjusting the cation ratios in the solution, flow rate, and the substrate temperature. Further heat treatment is required to improve the film crystallinity and to optimize the microstructure. The growth rate of this process is in the order of $0.7\text{-}1 \mu\text{m/min}$ [120]. This technique was primarily used for $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$ (Tl-1223) [4, 121] and $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_8$ (Hg-1223) [122] compounds. Hitachi [121] reported a one-meter long Tl-1223 film on Ag tape with a J_c of $9 \times 10^4 \text{ A/cm}^2$ at 77 K , 0 T . Initial attempts of deposit YBCO on a Ag tape have been reported to have a large FWHM value of about 20° [119] even though results on single crystalline LaAlO_3 showed a sharp texture with in-plane FWHM of $\sim 3^\circ$ [120]. The main problems associated with spray pyrolysis are high porosity and rough surfaces on the YBCO layers.

LPE was one of the promising routes for ReBCO coated conductor fabrications as it has a very high growth rate, typically one to two orders higher than those vapor-phase deposition techniques, and being a non-vacuum process made it an attractive candidate for possible industrial implementation. However, due to the incongruent melting, ReBCOs are predominantly grown from high-temperature solutions. Growing HTS compounds via the LPE technique started back in the late 1980s and early 1990s. Scheel *et al.* [123] transferred the LPE technique from semiconductor and optoelectronic films to ReBCO system and first presented details of the essential problems, difficulties, and possibility of growing HTS materials via LPE method in 1991. Since then, there had been intensive researches worldwide on LPE of ReBCO, first in Europe and later followed by the Japanese group at ISTEC. The LPE method has the potential for growth of thick films near the thermodynamic equilibrium. But one of the challenges is the high growth temperature, which may over oxidize substrates and ruin the buffer for metallic based tapes. Substrate reactivity causes serious problems in the formation of LPE films on metallic substrates for long-length coated conductor production. Most metals will react strongly with, or dissolve in, the highly corrosive Ba-Cu-O flux at the growth temperature of $970\text{-}1000^\circ\text{C}$ in air. In fact, some ceramic materials including YSZ and SrTiO_3 are attacked by the flux at the growth temperature. Though some effort has been made towards lowering flux temperature, for example Yamada *et al.* [124] used fluoride into the flux to decrease the peritectic temperature of the YBCO system by $\sim 30^\circ\text{C}$ and the eutectic temperature of more than 50°C by the addition of BaF_2 into a Ba-Cu-O flux. The temperature for YBCO films growth at $900\text{-}925^\circ\text{C}$ is still high. Another challenge is the low solubility of RE elements in the flux, which resulted in difficulties compare with the traditional LPE of semiconductor and garnet films. Attempts to grow YBCO films on metallic substrates by LPE have had limited success, with the most promising results by Yamada *et al.* [125] on NiCr tapes grown at reduced temperature of $\sim 820^\circ\text{C}$ from a Ba-Cu-O-F-Ag flux having zero-field transport J_c at 77 K as high as 1.9 MA/cm^2 . However, the Ni-Cr tape used had a rather complex buffer and seed layers prepared by PVD techniques that may cause major concerns towards large-scale industrial implementation. More recently, Shiohara *et al.* [126] reported $1 \mu\text{m}$ YBCO film with current density over 100A/cm on the SOE buffer, however, as of the difficulties, they decided to abandon LPE for now.

5. Studies of Grain Boundaries in HTSC

5.1 Grain boundaries in high T_c superconductor

Since the first day of HTSC superconductivity, the material science and the physics of grain boundary in superconducting compounds have developed into a fascinating field of research. This is because after the enthusiasm following the discovery of high T_c superconductivity, it was realized that applications of these materials are extremely difficult to achieve. The reason for these difficulties rooted in the ultra-short

coherence length of a few angstroms seemed to preclude the current carrying ability of polycrystalline samples. These unique electronic properties, different from those of the grain boundaries in conventional metallic superconductors, have made grain boundaries in a high temperature superconductor an important tool for basic science. They are moreover a key issue for large-scale applications of high T_c cuprates. With the issue of intergrain weak link in grain boundaries, highly textured films and even single crystalline are essential for the application of coated conductors.

The properties of polycrystalline high T_c superconductors are influenced in a complex manner by a large variety of interfaces, such as grain boundaries and twin boundaries. To understand the behavior of bulk polycrystalline, information about the properties of individual grains and well-defined interfaces is required. According to the displacement and the rotation of the abutting crystals, grain boundaries are usually classified into tilt and twist two categories. Normally tilt refers to a rotation around an axis in the plane of the grain boundary, and twist to a rotation of the crystal grains around the axis perpendicular to the grain boundary plane. Grain boundaries with identical misorientations of the grains with respect to the grain-boundary interface are called symmetric and otherwise they are asymmetric. For high temperature superconductors with their layered structure, 90° boundaries are particularly noteworthy. Fig. 6 shows the a -oriented grains in an ill developed YBCO film grown on cerium oxide buffered YSZ substrate. In addition to 90° boundaries formed by the rotation of the crystals, different angles boundaries may also be formed during the crystal growth on a polycrystalline substrate. For example, when YBCO grown on a RABiTS substrate with a proper buffer layer, the misorientation transferred from the substrate to the YBCO film, though the FWHM may be getting smaller. The electric properties are directly related to the misorientation in the layered cuprates high temperature superconductors.

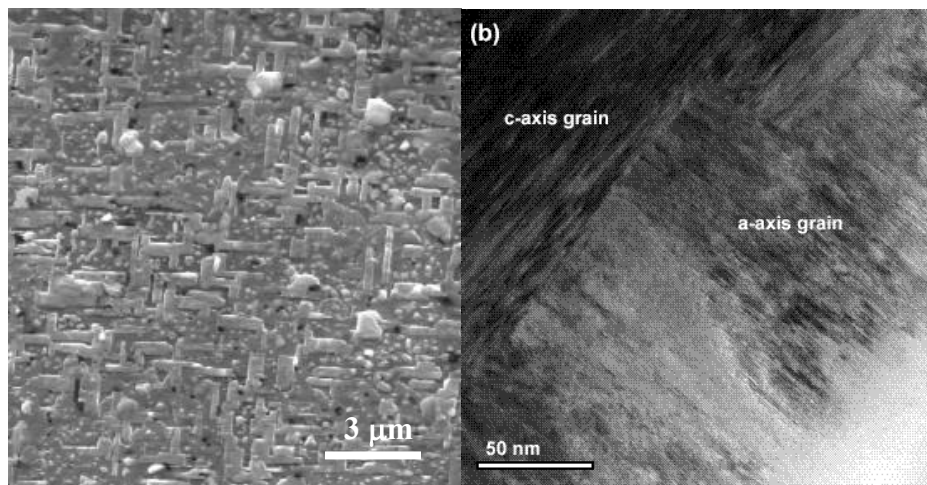


Fig. 6. a -oriented grains in the ill developed YBCO film on cerium oxide buffered YSZ substrate.

5.2 The bicrystal technique

Many attempts have been made to explain the decrease in J_c with increasing the misorientation between grains. The first experiment on a bicrystal grain boundary was carried out on the sample prepared by fusing two SrTiO_3 crystals with a predetermined misorientation between them. The YBCO film was deposited epitaxially on this bicrystal substrate, so that the resulting epitaxial film had a single grain boundary with a prescribed misorientation. Many groups have measured the current-carrying abilities of grain boundaries prepared by the bicrystal technique [127-130]. Fig. 3 shows representative recent data of the variation in J_c as a function of the misorientation angle θ [3]. Critical current density decreases exponentially by three to four orders of magnitude as θ increases to 45° . In addition, there are substantial scatters in J_c for a given θ . The reasons why the decrease is exponential and why the scatter is present are topics of ongoing research. On the other hand, except of the relative misorientation angle of the adjoining grains, the facet plane of a boundary may be important, it has been pointed out recently [131-133] that J_c through different facets may vary dramatically. Hence, a single facet GB is necessary to obtain the intrinsic transport property dependence on the misorientation angle. However, It is thought that the meandering of GBs in PVD films

was due to the initial 3-dimensional (3-D) island nucleation and could be reduced by lowering the growth rate by means of using a smaller supersaturation (thermodynamic driving force) such that 2-D nucleation will take place [134, 135].

5.3 Theories of GB in HTSC

Investigations show that an insulating layer with a resistivity of 10^{-9} – 10^{-7} Ω -cm exists at a grain boundary, which causes the characteristic boundary, and the strongly angular dependent J_c . Models related to GB electrical behaviors had been set up to explain the mechanism in GB. Grain boundaries are structural defects, which, by definition, interrupt the lattice structure of the adjacent crystals and thereby affect most of the properties of the correlated electron system. Based on experiments, many mechanisms have been proposed to explain the weak link in grain boundaries. The popular explanations are described as follows [2].

1). Mechanism based on dislocation array in grain boundary

Low-angle grain boundaries are composed of an array of alternating superconducting and nonsuperconducting regions[3], because low-angle grain boundaries are accommodated by arrays of separated, periodic dislocations as described in the so-called Dayem bridge model and related theories [127, 136, 137]. Small decreases in J_c have been observed for low-angle grain boundaries.

2). Mechanism based on deviations from an ideal stoichiometry

The deviations from an ideal stoichiometry at grain boundaries or formation of second phases have been regarded as a possible cause of the weak-link behavior. Such effects may be true for the case of bismuthates, but may not be the controlling factor for the ReBCO, because weak links exist for the grain boundaries with excellent cation composition [136, 138].

3). Order-parameter symmetry-based mechanism

High- T_c cuprates are characterized by a predominant dx^2-y^2 -wave pairing symmetry [139] and the order-parameter symmetry affects the transport properties of grain boundaries in the high- T_c cuprates in various ways. The dx^2-y^2 -wave-dominated symmetry together with the microstructure of the grain boundaries has been found to cause a depression of J_c by one to two orders of magnitude, as the [001]-tilt angle is increased from 0° to 45° [131, 140, 141].

4). Interface charging and band bending

The high resistivity of the normal-state demonstrates an insulating layer in the grain boundary; this is in accordance with EELS studies, which have revealed a reduction of mobile charge carriers at grain boundaries [132, 142]. Recently, Mannhart and Hilgenkamp pointed out [143, 144] that bending of the electronic band structure could occur at interfaces in the high- T_c cuprates, causing depletion or enhancement layers [129, 145, 146] next to the interface.

5). Mechanism based on direct suppression of the pairing mechanism

The misorientation and the interruption of the periodic lattice structure may depress the pairing interaction and result in a low intergrain current density. the interrupting of antiferromagnetic order of the CuO_2 planes is a good example described by Chaudhari *et al.* [147].

The mechanisms described above are not mutually exclusive but that several interacting mechanisms control the grain-boundary transport simultaneously. The knowledge of the grain boundary at present is far from complete; research will lead to the deep understanding in the near future.

6. Highlights of some research directions in ReBCO coated conductors

6.1 ReBCO thick films

Since the first demonstrations of YBCO films with a current density over 1 MA/cm² at 77 K [5, 22, 148, 149], great efforts have been devoted to achieve such performance over long lengths and thick films. The driving force for the development of a thick film is to increase the J_e for coated conductor applications. The current-carrying capacity is exclusively determined by the thickness of a YBCO film for a determined surface area/width and the engineering critical current density J_e , which is determined by the total thickness of the YBCO coating and substrate. J_e will increase for a thick YBCO film if the critical current density keeps at high level. However, it has been reported that the J_c decreases rapidly as the thickness of an epitaxial YBCO film increases either on a single-crystal substrate [150-155] or on a metal substrate [156, 157]. While the trend of an exponential drop of J_c with YBCO thickness seems to be universal regardless of the deposition technique, the cause of this drop is not yet clear. In literature, a transition to *a*-axis orientation [150, 152], a loss of in-plane and/or out-of-plane texture [151, 153], a decrease in the number of pinning sites [154], and a change in the microstructure [156] have been reported as possible causes for the

J_c drops. Hence it is important to determine whether thick YBCO films with good transport properties can be made on textured metal substrates. In the early time people believed that the super-current can only be carried by a thin YBCO layer near the interface. However, recently, the experiment performed in Oak Ridge National Laboratory showed that high current densities could be achieved on thick films up to 6.4 μm . The questions are is there a limit for thickness, how to get high current density and what is ideal thickness for a coated conductor. It is both theoretical and practical importance to answer these questions.

6.2 PED approach

By using similar principle of PLD, a new technique called PED (pulsed electron deposition) was developed recently [158]. It was estimated that the investment of PED is only 1/10 of the cost of PLD and the energy efficiency is higher than the PLD method as well. The preliminary experiment results showed that good current density (34A/cm and 0.85 MA/cm²) could be made with the PED method by depositing the YBCO film on a RABiTS substrate with a thickness of 0.4 μm . By using the PED method, Zhai *et al.* [159] deposited a 300nm YBCO film on a STO single crystal and demonstrated $T_c(\text{onset}) = 89.9\text{K}$ and $J_c > 1.5 \text{ MA/cm}^2$. However, Zhai's result showed the presence of impurity phases. A study of the plume dynamics (using an ion probe as an in-situ measurement technique) shows strong differences between the PED and PLD plumes, possibly indicating the origin of the different growth behaviors. At present no high J_c comparable with the PLD method has been demonstrated for the PED approach and because this is a newly developed technique, it will take some time for a bettering understanding of the potential scale-up for this PVD process.

6.3 Grain boundary doping

Recently, some results reported by Mannhart's group [130] raise hopes for increasing the critical current density of high- T_c wire. It suggests a way to improve the weak coupling between the polycrystalline grains while preserving the critical current density within each grain [160]. Because divalent calcium is roughly the same size as a trivalent yttrium and so readily takes its place within the crystal lattice. It is believed that if calcium diffuses into GBs in a polycrystalline YBCO film, the critical current density might be improved. In 1999, Mannhart's group examined this possibility[161] by doping a 24° GB with 30% Ca to add more hole charge carriers to the material. A significant increase in the critical current density of the GB was observed by as much as an order of magnitude at 4 K. But this improvement came at a price: with so much Ca in the material (at grain boundaries and in grains), T_c fell below 80 K. So how to dop GBs only is a challenge for high current density with less cost in T_c loss. Recently, Mannhart's group seems found a promising way to dop only the GBs to achieve good GB coupling while maintaining a high T_c within grains. They designed a bilayers and superlattice structure as illustrate in Fig. 7 [160], by depositing an undoped YBCO alternatively capped with a thin layer of 30% Ca-doped YBCO. This cap layer doesn't significantly contribute to the current flow through the film at 77 K, but it does provide a source for Ca that can migrate down into the undoped layers. As expected, at a temperature of 77K, the critical current density of the 24° grain boundaries in the average doping bilayer and in the trilayer YBCO films have already reached $1.2 \times 10^5 \text{ A cm}^{-2}$ and $3.3 \times 10^5 \text{ A cm}^{-2}$, greatly exceeding the J_c value of the undoped sample of $5.5 \times 10^4 \text{ A cm}^{-2}$.

Daniels *et al.* examined the effect of Ca doping on small-angle GBs[162], looking at uniformly Ca-doped YBCO films with GB angles of 5° and 7°. Their experiment, too, found a decrease in T_c , and increase in the GB critical current density for a 30% Ca doped YBCO film on STO bi-crystals. Holzapfel *et al.* [163] studied Ca- or Ag-doped YBCO thin films prepared by pulsed laser deposition on symmetric low angle SrTiO₃ bicrystals with misorientation angles of 4°, 8°, 12°, and 16°. Their results show that, for the 10 % Ca substitution, no significant J_c enhancement at 77K in zero field but a strongly decreased J_c at higher magnetic fields due to a reduce in irreversibility field at 77K. Ag-doped Y123 bicrystal films showed increases of critical current densities at a higher field and a slight shift of transition from a strong to a weak link compared with the undoped Y123 bicrystal film. It is clearly that there are many issues related to the GB doping to be solved before any practical applications availed for this technique.

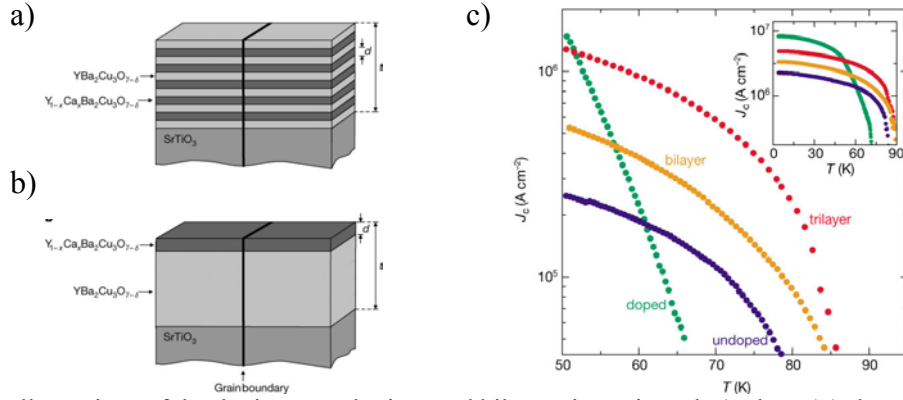


Fig. 7. a), b) Illustrations of the doping superlattices and bilayers investigated. c) The $J_c(T)$ -dependencies of four samples: a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film, a homogeneously doped $\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ film, a bilayer consisting of a 130-nm $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ layer and a 20-nm-thick $\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ layer on top as illustrated in b), and a 20-nm $\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ /130-nm $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ / 20-nm $\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ trilayer. (Adapted from Mannhart *et al.* [160])

6.4 Termination study on single crystal and on buffer layer surface

The termination of the (001) SrTiO_3 substrate is expected to influence both the nucleation and the dislocation formation for a YBCO film. Kawasaki *et al.* [164] developed a method to fabricate atomically smooth SrTiO_3 (100) with steps one unit cell in height by treating the crystal surface with a pH-controlled NH_4F -HF solution. TiO_2 atomic plane terminated the as-treated clean surface and the terminating atomic layer could be tuned to the SrO atomic plane by a homoepitaxial growth. This perfect layer-by-layer mode was verified by reflection high-energy electron diffraction and atomic force microscopy. This technology provides a well-defined substrate surface for atomically regulated epitaxial growth of such perovskite oxide films.

To investigate substrate-film interactions, Huijbregtse *et al.* [165] developed another ex-situ method to prepare SrO-terminated substrates in addition to the well-known TiO_2 termination by high temperature anneal followed by PLD depositions of SrO/TiO and YBCO. It is shown that: preferential precipitation occurs when depositing submonolayer of Y123 films on SrO-terminated SrTiO_3 , and such precipitates are absent on the TiO_2 terminated sample. It appears that the substrate termination determines both the starting and terminating layer of the Y123 phase. Similar effects are observed in submonolayer thick films with cation stoichiometries 122, 124 and 133. When growing several 123 monolayers on SrO-terminated substrates, no precipitates are formed. Instead, to incorporate the off stoichiometry, defects are introduced. Surprisingly, the density of threading dislocations does not depend on the substrate termination.

6.5 Modeling of current flow path in coated conductors

Inter- and intragrain studies have demonstrated that for polycrystalline YBCO films, the critical current density J_c of a grain network is likely to be limited by dissipation at the low-angle grain boundaries, which generally have less current carrying capability than the grains [166]. Hence, the current flow in polycrystalline coated conductors thus relies on the transfer of current across the grain boundaries. A number of models have been proposed to study current percolation in networks of low angle grain boundaries [167, 168].

The grain shape is the one of the most important factor in modeling the current flow. In Specht [169] and Rutter's models [31], an array of hexagons were used to simulate the real case in grain boundaries, while Nakamura *et al.* [170] uses squares, and Leitenmeier *et al.* [171] and Hammerl *et al.* [172] use rectangles. In Holzapfel's model, unlike others, uses different shape grains measured by EBSD. Another issue addressed in these models was the grain boundary misorientation. Early models [169] simply assigned 0 or 1 to either conducting or non-conducting grain boundaries. Subsequent models revised it by simulating critical current flows based on grain misorientations measured by EBSD. Some people also used Gaussian distributions to assign grain orientations. When considering the misorientation, an equation that

phenomenological quantifies the effect of misorientation angle θ on J_c for the weakly coupled regime is described as follows:[173]

$$J_c(\theta) = J_c(0) \exp\left(-\frac{\theta}{\alpha}\right); \quad [\theta \geq 4^\circ] \dots\dots\dots(1)$$

Normally, the plateau region angle α falls into the range of $3.2^\circ \sim 5.5^\circ$ and $J_c(0) = 3.2 \text{ MA/cm}^2$ [144, 174, 175]. In a real case, considering a path along the grain boundaries, which runs across the complete width of the sample. The maximum current, which can cross this path, is equal to the sum of the critical currents of the grain boundary segments, which comprise the path. When all such paths, which cur the samples cross the width, are considered, the minimum of these is the one that will limit the current flowing along the sample. This is a property of networks known as the maximum-flow/minimum-cut theory, proposed by Ford and Fulkerson [176].

In the consideration of limitations of existing models, recently, Rutter *et al.* developed a new model by using a 2-D array of square pixels. Initially each pixel is attributed an index number or letter, which identifies it as a grain and hence at the start of the simulation, the material consist of a large number of very small, square grains. The algorithm used to simulate grain growth first randomly selects an individual pixel and evaluated its overall energy based on the number of neighbors, which are in different grains. The algorithm then evaluated the effect on the energy of changing the index number of the pixel such that it is in the same grain as one of its 4 neighbors (linked by edges). If this energy change is favorable (i.e. negative) or neutral (zero), then the change is make with probability (P)=1. If however, the energy change would be positive, then the change is made with the equation:

$$P = \exp\left(\frac{-\Delta G}{kT}\right) \dots\dots\dots(2)$$

The factor kT is a thermodynamic variable and has been set equal to 1 for the purposed of this model. With this assumption Rutter *et al.* derived a new equation as follows:

$$J_c \approx \left(\frac{1}{N_l}\right)^{\frac{1}{N_w}} \dots\dots\dots(3)$$

Where N_l and N_w represent the number of grains in length and in width respectively.

This equation simulates most of practical coated conductors and predicts the width and length dependence when the width and length are rather high and the tape is highly aspected.

As point out in 1987, big aspect ratio grain boundaries can be realized by stacking in a brickwall-type manner plateletlike grains on top of each other [177]. The Bi-based high- T_c superconductors fabricated with the powder-in-tube technology is viewed as the example of the enhancement of large critical currents. Hammerl *et al.* [172] further developed this idea: they found ways to use large effective grain-boundary areas to enhance J_c of coated conductors that consisted of two- or three-dimensional grain-boundary networks. Hammerl's calculation was also based on Dijkstra's shortest path algorithm for undirected graphs, the grains acting as vertices, the grain boundaries, weighted by their critical currents, as edges. The calculation results showed that doping can double the critical current density. For lower and higher angle GBs, J_c is taken to be exponentially reduced to the undoped values. To consider the effects of stacking coated conductors in multilayer configurations, the intergrain critical currents flowing in the c direction were modeled by reducing $J_c(\theta)$ by an additional c -axis coupling factor f_c . For bilayers, an enhancement of the aspect ratio significantly increases I_c further, and a tape with misorientation of 45° can have the same J_c as a conventional tape with an alignment of 6° . According to Hammerl's calculations, current densities of $4 \times 10^6 \text{ A/cm}^2$ can be achieved for an average grain alignment of 10° at 77 K. Based on this approach, a road to competitive high- T_c cables is proposed.

7. Summary of Coated Conductor Development

The development of coated conductor technology has been reviewed. It is shown that the critical current density of high- T_c wires can be greatly enhanced by using three-fold approaches, which consist of grain alignment, doping, and optimization of the grain architecture. Clearly, there are still a lot of scientific and technological barriers to be overcome before long lengths of high J_c coated conductor be produced

commercially. Cost is still the main concern for scale up of the more reliable and reproducible techniques developed nowadays, especially the approach through vapor depositions such as the PLD method. TFA-MOD or other CSD methods may be the trend to overcome cost and speed consideration during the scale-up. However high quality and reproducibility will be the new focus for the development of these techniques.

Major advances have been made in the last 14 years in coated conductor development mainly in the three aspects: substrate, buffer layer and YBCO layer. Ni-alloy tapes seem to have advantages over pure Ni in terms of mechanical strength and oxidation resistance. However, the sharpness of the texture in Ni alloys appears to be less superior than to pure Ni. One can bypass this disadvantage by depositing a pure Ni layer on top of Ni-based alloys (such as Ni-Cr and Ni-W alloys), which resulted in a good texture as pure Ni. If the poor mechanical properties and low melting point could be overcome, Ag could be a good candidate because it has a major advantage over Ni or Ni-alloy tapes: the possibility of eliminating the need of a buffer layer.

In the area of buffer development, the optimum buffer layer(s) also remains unclear. There are many buffer materials being investigated and all have potential advantages and disadvantages in terms of the ease of fabrication and fulfilling the functions of a buffer layer. For the case where a buffer layer is unavoidable, a non-vacuum processes would be suitable for low cost and scale up. However there is none of the processes through CSD has been demonstrated results good enough for long length coated conductor applications. Whilst a high J_c superconducting layer can be reliably produced by various PVD techniques, there are major difficulties in transferring these technologies to large-scale commercial productions. High-speed, non-vacuum processes capable of fabricating thick films seem to be the most hopeful option for potential industrial implementations and the results of TFA-MOD have demonstrated the possibility and bright future. New technological and theoretical analyses show the potential for developing better-coated conductor wires in the near future.

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