

TM 5 8/1/97
TOA # 52

02

FABRICATION OF BIAXIALLY-TEXTURED THICK FILM Y-Ba-Cu-O
SUPERCONDUCTOR BY MOCVD ON CUBE-TEXTURED METAL SUBSTRATES

¹V. Selvamanickam, ²A. Ivanova, ³D. B. Fenner, ⁴T. Thurston, ¹M. S. Walker,
²A. E. Kaloyeros, and ¹P. Haldar

¹Intermagnetics General Corporation
Latham, New York 12110

²University at Albany
Albany, NY 12222

³Fenner Engineering
Simsbury, CT 06070

⁴Brookhaven National Lab
Upton, NY 11973

Abstract

YBa₂Cu₃O_x (YBCO) superconducting films have been deposited on cube-textured metal substrates and single crystal substrates by Metal Organic Chemical Vapor Deposition (MOCVD). The cube-textured substrates were fabricated by a conventional metal-working process. A high degree of in-plane texture (5° to 10°) was achieved in these substrates. Buffer layers of Pt, CeO₂, and Ytria-Stabilized Zirconia (YSZ) were deposited on the cube-textured substrates by pulsed laser ablation. These buffer layers were also found to be strongly biaxially-textured, with a spread of about 10° in the in-plane orientation. YBCO films deposited by MOCVD on both cube-textured metal substrates and single crystal YSZ substrates were found to be stoichiometric, dense, and biaxially-textured. A spread of approximately 10° in the in-plane texture has been achieved in the YBCO films deposited on the cube-textured metal substrates.

This work was supported by the Department of Energy (Contract # DE-FG02-95ER82000) and the Air Office of Scientific Research (Contract F49620-95-C-0023).

1/25?

Introduction

Several electric power applications of high temperature superconductors (HTS) require operation in significant magnetic fields at 77 K [1]. Owing to their inherent deficiencies in flux pinning properties, it has been well documented that the Bi-based superconductors cannot be used in high magnetic fields at these temperatures [2]. Conversely, Y-Ba-Cu-O superconductors have been shown to exhibit current densities greater than 10^6 A/cm² at 77 K and operate at magnetic fields as high as 5 T, at this temperature [3]. These properties were however achieved only when Y-Ba-Cu-O was deposited on single crystalline substrates and could not be achieved in randomly aligned polycrystalline substrates. The interest in Y-Ba-Cu-O conductors was renewed in 1991 through the demonstration of high current densities in polycrystalline substrates by the development of a biaxially-textured buffer layer over a randomly aligned polycrystalline metallic substrate [4]. The biaxially-textured buffer layer was deposited by ion beam assisted deposition technique (IBAD). Process optimization over the last few years has resulted in current densities higher than 10^6 A/cm² in IBAD-buffered YBCO tapes that are more than a micron in thickness [5]. These tapes are also found to sustain a high current density even in fields of several Tesla at 77 K, and can meet or exceed the requirements for various power applications if they can be fabricated in long lengths. After the successful demonstration of the viability of Y-Ba-Cu-O conductor fabrication using biaxially-textured buffered substrates by the IBAD technique, alternate processes have been developed to achieve biaxially-textured substrates. Some examples are hot rolling [6], rolling and heat treatment [7], and inclined substrate pulsed laser deposition [8].

The present challenge is to develop a long-length fabrication technology to deposit suitable buffer layers and the superconductor on a biaxially-textured metallic tape. One deposition technology of interest is metal organic chemical vapor deposition (MOCVD). MOCVD has been industrially proven to be a rapid deposition technique for various materials and is the deposition technology of choice for large-scale manufacturing of dielectric, ferroelectric and piezoelectric materials. Even YBCO superconductors have been deposited at rates as high as 0.5 to 1 μ m/min with superior superconducting properties [9]. MOCVD can be used to achieve uniform deposition over large areas and is not limited by line-of-sight deposition and is thus amenable to large-scale fabrication. In this study, MOCVD has been examined as a deposition technique to fabricate Y-Ba-Cu-O superconductor on cube-textured metal substrates.

Experimental

The biaxially-textured metal substrates were fabricated by a metal working process based on techniques well-established for achieving cube texture in Ni alloys [10] and other metals [11]. As demonstrated as early as in 1927 [12], in this technique, polycrystalline metals are subjected to heavy deformation by rolling followed by a suitable recrystallization process which results in texturing along the (100) orientation in the rolling plane and along the [100] orientation in the rolling direction. Such a biaxial texture has not found much use in conventional metal working except in the fabrication of cube-textured Fe-Si alloys for transformer applications where directional properties are needed to achieve low losses [13]. Nickel and copper substrates were fabricated in this study by the conventional cube texturing technique. The texture of over 20 metal substrates were examined by reflective X-ray measurements at the University at Albany and by transmission X-ray measurements at the Synchrotron Light Source at Brookhaven National Laboratory (BNL).

Buffer layers of Pt, CeO₂, and YSZ were deposited by pulsed laser ablation on cube-textured metal substrates. The substrates were polished, degreased, and cleaned prior to deposition of buffer layers. Over 20 different deposition conditions were studied. The deposition temperature was varied over a range of 75°C to 800°C. The thickness of the buffer layers ranged from 250 to 5000 Angstroms.

YBCO was deposited on the buffered metal substrates as well as on single crystal (unbuffered) YSZ substrates by MOCVD. The MOCVD experiments were conducted in a research reactor shown schematically in fig. 1. As shown in the figure, solid sources of Y-

tmhd, Ba-tmhd, and Cu-tmhd (tmhd = tetramethyl heptanedionate) were sublimated at appropriate temperatures and accurately metered using mass flow controllers, in a carrier gas of nitrous oxide. The vapors were mixed in a manifold and then sprayed from a shower head in the reactor. All gas lines were heated above the sublimation temperature of Ba-tmhd so as to avoid condensation. The substrate was placed beneath the shower head on a heater block. Steady state conditions were first achieved in the precursor delivery system and in the reactor prior to deposition. The deposition temperature was varied from 750°C to 810°C. The deposition was performed in a reactor pressure of 1 to 10 Torr. After deposition, the samples were cooled in a high oxygen partial pressure and subjected to oxygen annealing at 500°C.

The texture of the buffer layers and YBCO superconductor was examined by 2-theta, rocking curve, and polefigure XRD measurements. The overall composition of the YBCO films was analyzed by Rutherford Backscattering Spectroscopy (RBS).

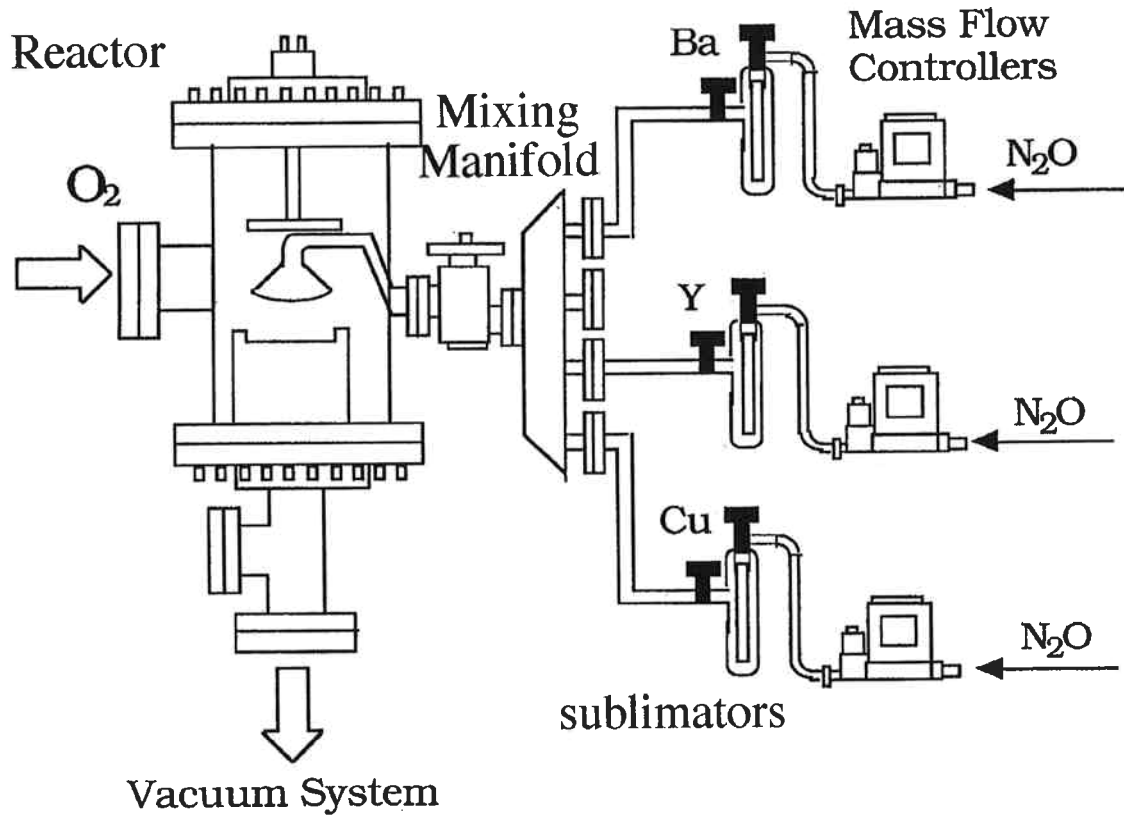


Figure 1 Schematic of MOCVD Research Reactor

Results and Discussion

Cube-textured Metal Substrates

The metal working and heat treatment process parameters were optimized to achieve a high degree of cube texture in both Ni and Cu substrates. Results from a 2-theta XRD measurement performed on a Ni substrate is shown in fig. 2. A high degree of texturing along the (200) plane can be observed. Figure 3 exhibits a polefigure of the (111) peak of the Ni substrate which reveals a high degree of in-plane texture. Results from azimuthal scan (χ -scan) transmission X-ray measurements performed at BNL on a similar substrate is shown in fig. 4 and indicate that the texture is present through the thickness of the substrate. The spread in the in-plane texture in over 20 substrates was found to be in the range of 5 to 10°.

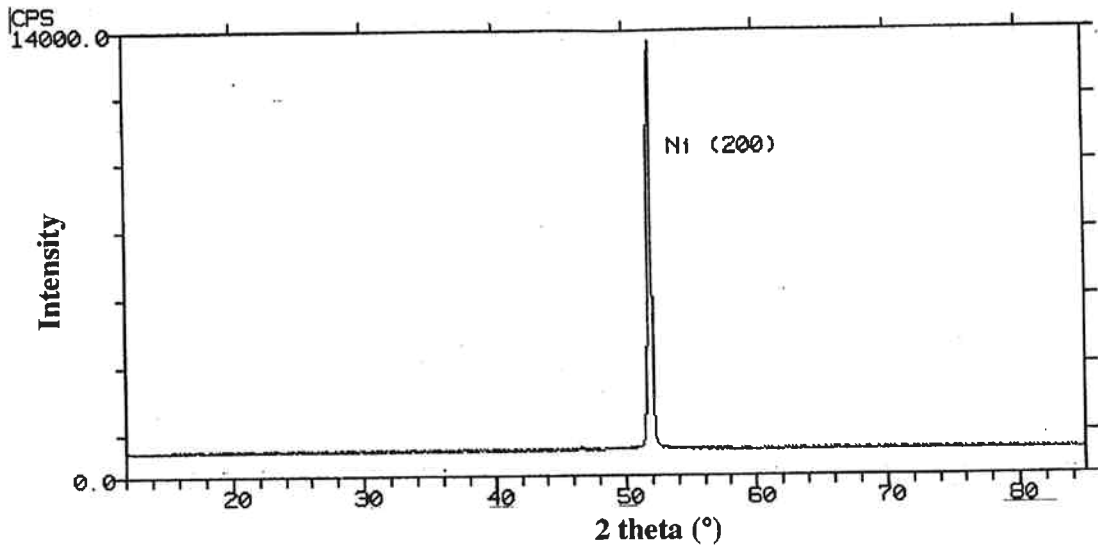


Figure 2 2-theta XRD pattern of a cube-textured Ni substrate showing an intense (200) texture.

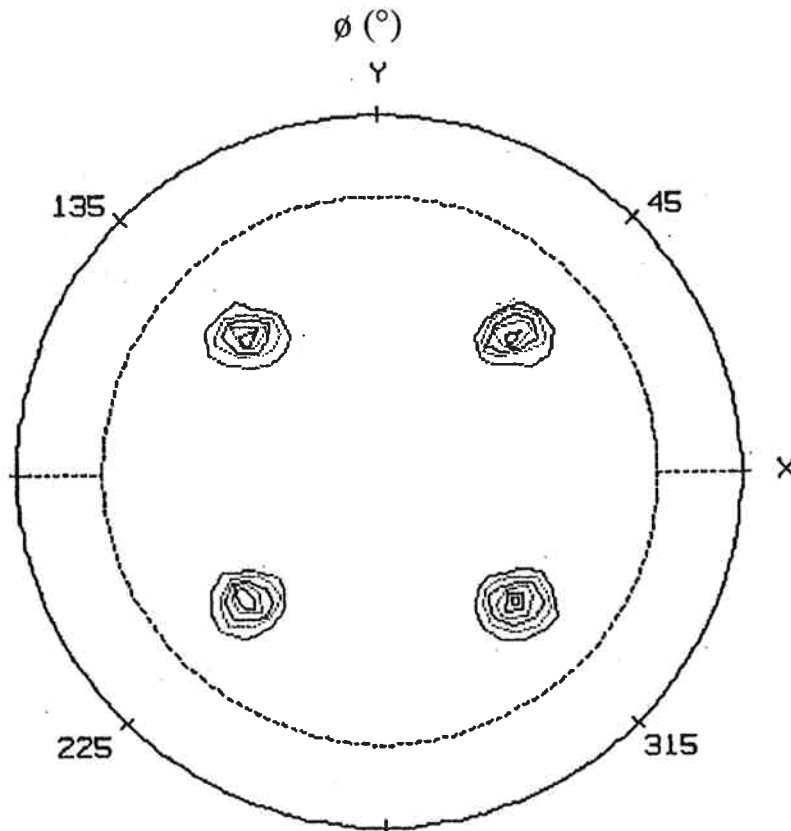


Figure 3 Polefigure of the (111) peak of a cube-textured Ni substrate showing a high degree of in-plane texture.

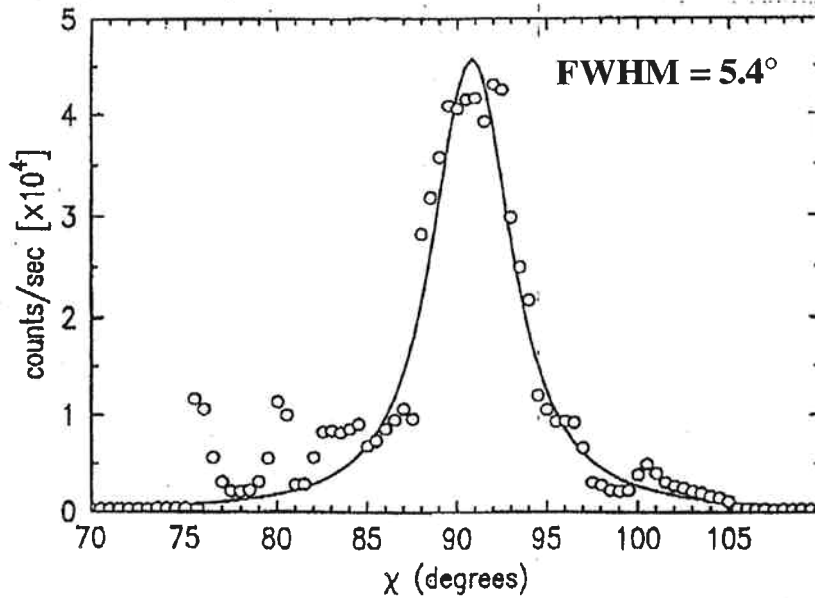


Figure 4 Azimuthal scan (χ -scan) of a cube-textured Ni substrate examined by Transmission X-ray measurements at the Synchrotron Light Source at BNL. The substrate is found to exhibit a strong in-plane texture with a spread less than 6° .

Buffer Layer deposition

2-theta XRD patterns obtained from two samples after buffer layer deposition are shown in figures 5 and 6. Figure 5 represents data obtained from a cube-textured Ni substrate with Pt and YSZ buffer layers. As seen in the figure, both buffer layers are textured in the (200) orientation. A minor fraction of YSZ (111) is still present. Data obtained from a cube-textured Ni substrate with CeO_2 and YSZ buffer layers is shown in fig. 6. Evidence of texturing in the (200) orientation in both buffer layers is seen in the figure. Further, polefigure X-ray measurements revealed strong in-plane texture in the YSZ layer with a spread of about 10° , as shown in figure 7.

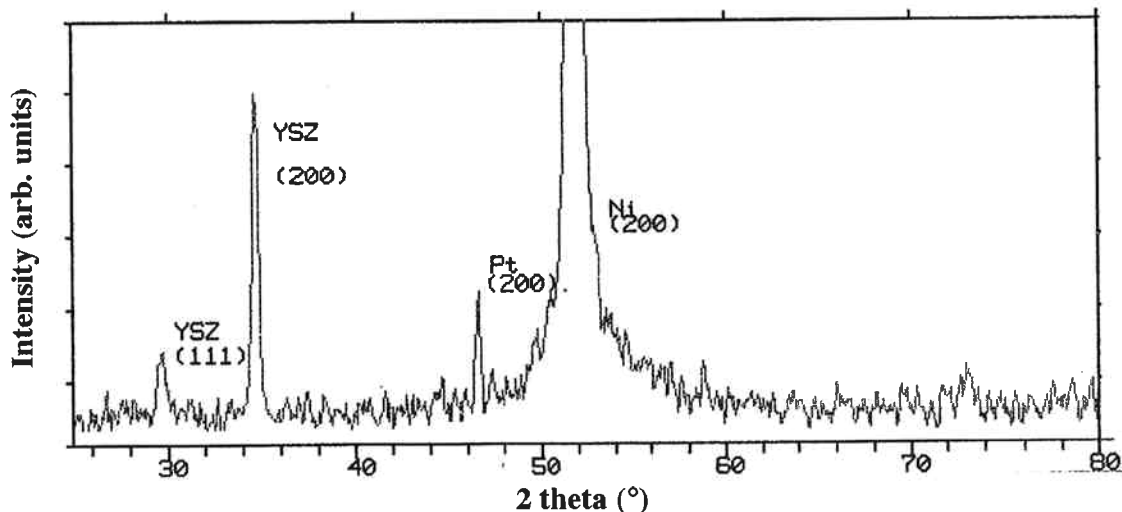


Figure 5 2-theta XRD pattern of a cube-textured Ni substrate with textured Pt and YSZ buffer layers.

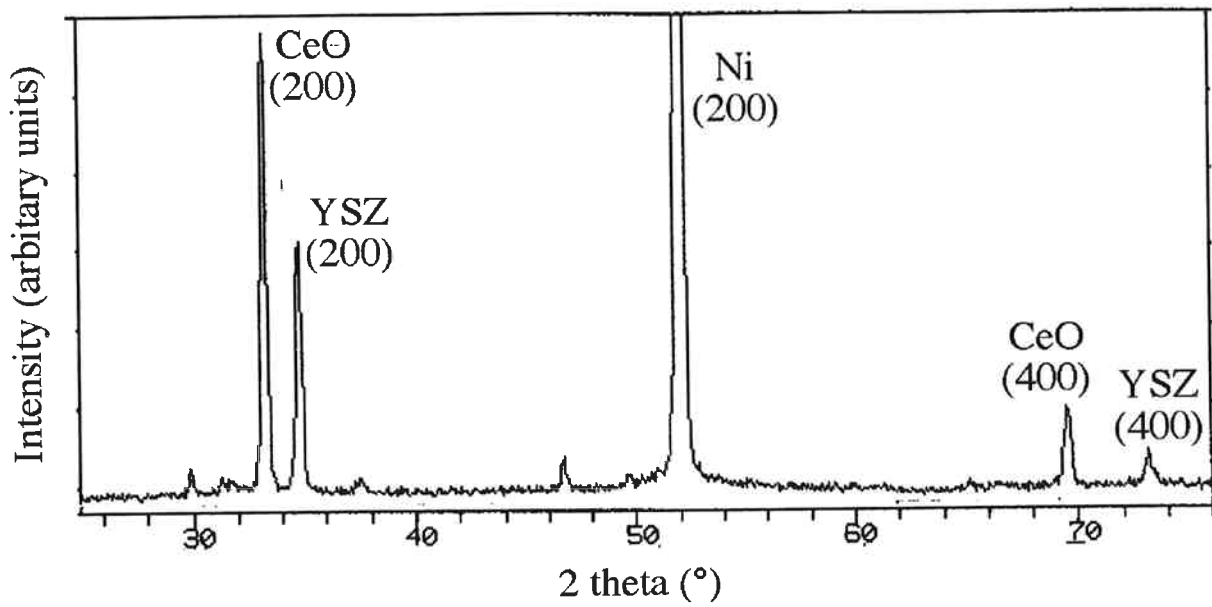


Figure 6 2-theta XRD pattern of a cube-textured Ni substrate with textured CeO_2 and YSZ buffer layers

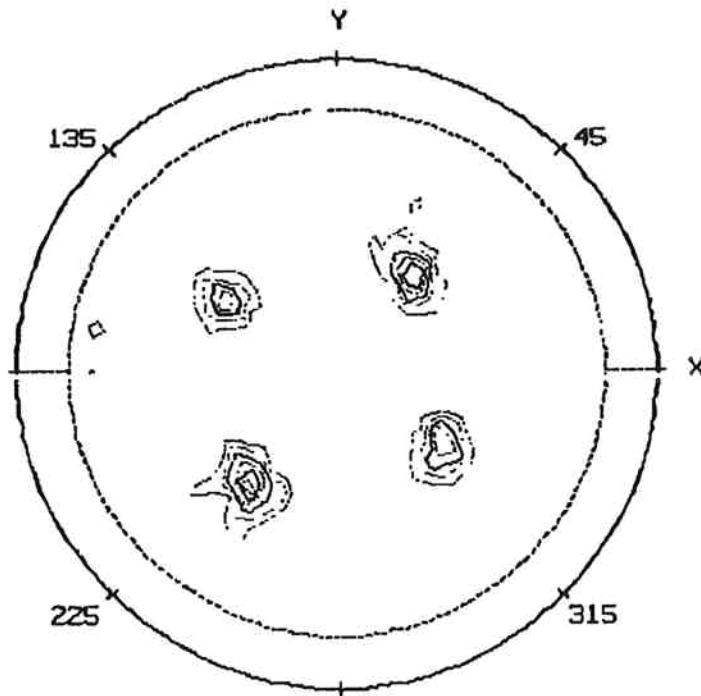


Figure 7 Polefigure of (220) peak of YSZ buffer layer described in fig. 6. A strong in-plane texture can be seen.

YBCO Deposition by MOCVD

YBCO deposition on Single Crystal YSZ Substrates

The substrate temperature was found to influence the stoichiometry of YBCO films deposited on YSZ single crystals. Table 1 summarizes the overall composition of seven YBCO films deposited over a temperature range of 770°C to 810°C and under similar conditions of reactor pressure, sublimation pressure, carrier gas flow rate, oxygen flow rate and sublimation temperatures. It can be seen from Table I that films deposited at a lower temperature were Ba deficient. The film deposited at 810°C was close to stoichiometric composition. 2-theta and rocking curve measurements obtained from one of the films is shown in fig. 8. A high degree of c-axis alignment, with a spread in the c-axis texture of 0.7° can be observed.

Table I Composition of YBCO films deposited by MOCVD on single crystal YSZ substrates, at different temperatures

| Substrate Temperature (°C) | Composition | | |
|-------------------------------|-------------|-----|-----|
| | Y | Ba | Cu |
| 770 | 1.0 | 1.7 | 3.0 |
| 777 | 1.0 | 1.8 | 3.0 |
| 785 | 1.0 | 1.5 | 3.0 |
| 790 | 1.0 | 1.7 | 3.0 |
| 798 | 1.2 | 1.9 | 3.0 |
| 800 | 1.0 | 1.9 | 3.0 |
| 810 | 1.0 | 2.0 | 3.0 |

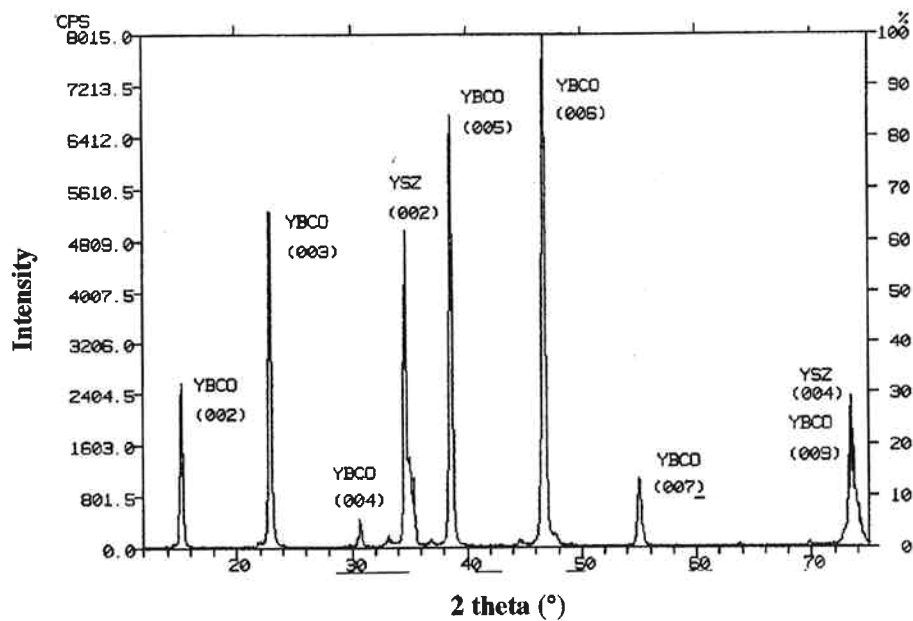


Figure 8(a) 2-theta XRD pattern of a YBCO film deposited by MOCVD on a single crystal YSZ substrate. A high degree of c-axis texture is seen.

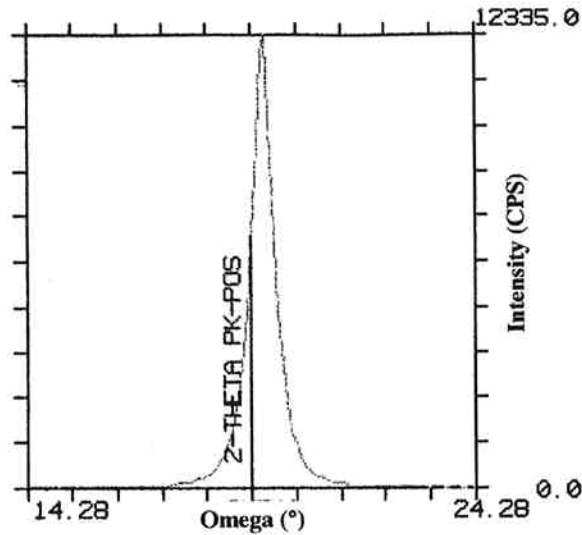


Figure 8(b) Rocking curve of (005) peak of a YBCO film deposited by MOCVD on a single crystal YSZ substrate. A high degree of c-axis texture is seen.

YBCO deposition on Cube-textured Metal Substrates

The substrate temperature had to be optimized again for MOCVD on cube-textured metal substrates due to the difference in thermal conductivity between the metal and single crystal YSZ substrate. Further, the precursor sublimation conditions had to be modified to achieve a stoichiometric composition of the YBCO film. Results from 2-theta measurements from a YBCO film deposited on a cube-textured Ni substrate with Pt and YSZ buffer layers are shown in fig. 9. The YBCO film can be seen to be c-axis aligned. Figure 10 exhibits the polefigures of the (005) and (013) peaks of the film. These measurements show a high degree of biaxial texture in the film. The spread in the in-plane texture is found to be about 10°. RBS analysis showed the composition of the film to be stoichiometric.

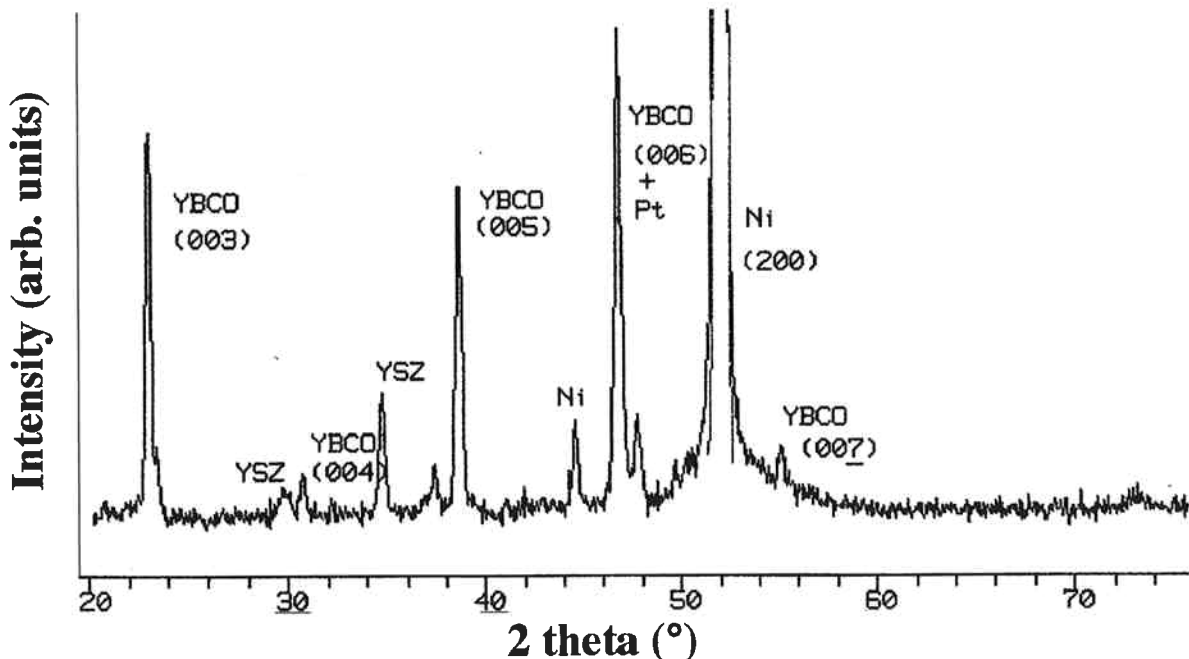


Fig. 9 2-theta XRD pattern obtained from a YBCO film deposited by MOCVD on a cube-textured Ni substrate with Pt and YSZ buffer layers.

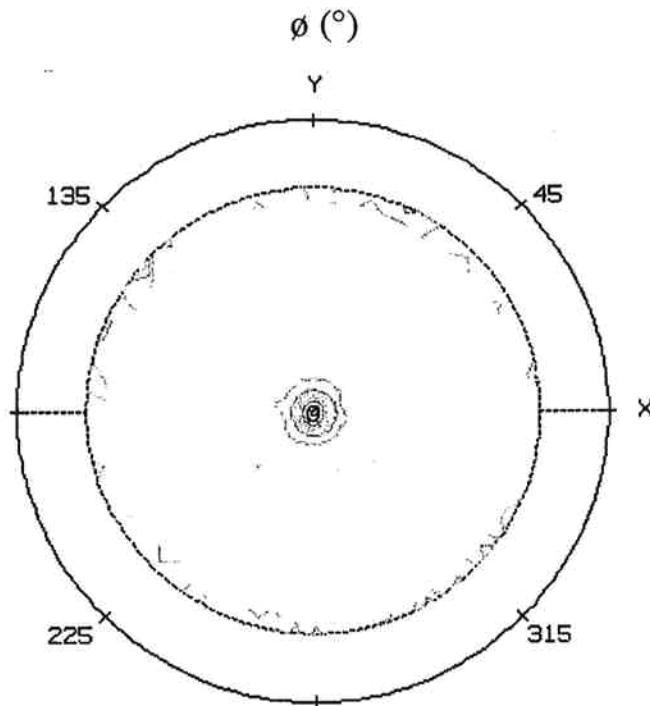


Fig. 10(a) Polefigure of the (005) peak of a YBCO film deposited by MOCVD on a cube-textured Ni substrate with Pt and YSZ buffer layers showing strong c-axis orientation.

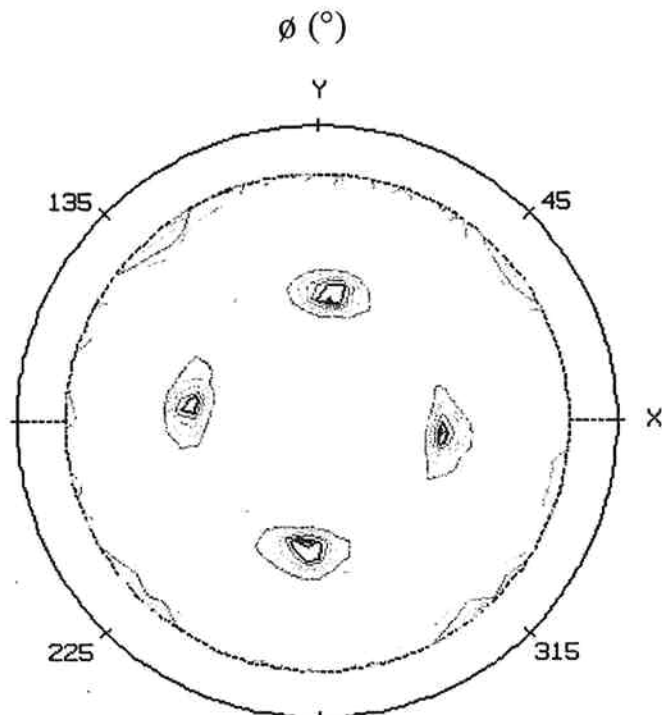


Fig. 10(b) Polefigure of the (013) peak of a YBCO film deposited by MOCVD on a cube-textured Ni substrate with Pt and YSZ buffer layers. The film is found to be biaxially textured.

Conclusions

Metal Organic Chemical Vapor Deposition has been shown to be a viable technique to fabricate biaxially-textured YBCO on polycrystalline metal substrates. An in-plane texture of better than 10° has been achieved in the cube-textured metal substrate, buffer layer and YBCO superconductor. The achievement of biaxially-textured YBCO along with the proven advantages of uniform deposition over large areas and high deposition rates suggest that MOCVD could be a promising candidate for long-length fabrication of surface-coated YBCO conductors.

References

1. U.S. DOE, Office of Energy Management, Superconductivity Program for Electric Power Systems, FY 1994-1998 Multi-Year Plan, 1994
2. D. H. Kim, K. E. Gray, R. T. Kampwirth, J. C. Smith, D. S. Richeson, T. J. Marks, J. H. Kang, J. Talvacchio, and M. Eddy, Physica C, 177, (1991) 431.
3. B. Roas, L. Schultz, and G. Saemann-Ischenko, Phys. Rev. Lett. 40, (1989) 829.
4. Y. Iijima, N. Tanabe, and O. Kohno, Adv. in Supercond. IV, ed. Hayakawa and Koshizuka (Springer Verlag, Tokyo, 1992) 679-682.
5. X. D. Wu, S. R. Foltyn, P. Arendt, J. Townsend, C. Adams, I. H. Campbell, P. Tiwari, Y. Coulter, and D. E. Peterson, Appl. Phys. Lett. 65, 1961 (1994).
6. T. J. Doi, N. Sugiyama, T. Yuasa, T. Ozawa, K. Higashiyama, S. Kikuchi, and K. Osamura, Proc. of Eighth International Symposium on Superconductivity (ISS'95), Hamamatsu, Shizuoka, Japan, Oct. 30-Nov. 2, 1995.
7. A. Goyal, D. P. Norton, J. D. Budai, M. Paranthamna, E. D. Specht, D. M. Kroeger, D. K. Christen, Q. He, B. Saffian, F. A. List, D. F. Lee, P. M. Martin, C. E. Klabunde, E. Hatfield, and V. K. Sikka, Appl. Phys. Lett. 69, (1996) 1795.
8. K. Hasegawa, N. Yoshida, K. Fujino, H. Mukai, K. Hayashi, K. Sato, S. Honjo, Y. Sato, T. Ohkuma, H. Ishii, Y. Iwata, and T. Hara, Proc. Ninth Int. Symp. Supercond. Sapporo, Japan, Oct. 21-24, 1996.
9. A. Ignatiev, Q. Zhong, P. C. Chou, X. Zhang, J. R. Liu, and W. K. Chu, 'Thick Film YBCO Growth by Photo-Assisted MOCVD', Proc. Tenth Anniversary HTS Workshop, Houston, Texas, March 1996.
10. W. E. Seymour and D. Harker, 'Recrystallization Reaction Kinetics and Texture Studies of a 50 Iron 50 Nickel Alloy', Trans. AIME, 188, (1950) 1001.
11. H. Hsu, R. S. Cline, and S. R. Goodman, 'Annealing Textures in High-Purity Silver', Trans. AIME, 224, (1962) 96.
12. v. Göler and G. Sachs Ztsch Phys. 41, (1927) 827.
13. K. Deter, Acta Met. 7, (1959) 589.