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*Title:* High Current Y-Ba-Cu-O Coated Conductor Using Metal Organic Chemical Vapor Deposition and Ion Beam Assisted Deposition

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# High Current Y-Ba-Cu-O Coated Conductor using Metal Organic Chemical Vapor Deposition and Ion Beam Assisted Deposition

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***Abstract* --- $YBa_2Cu_3O_x$ (YBCO) films have been deposited on buffered metal substrates by Metal Organic Chemical Vapor Deposition (MOCVD). Nickel alloy substrates with biaxially-textured Yttria-Stabilized Zirconia (YSZ) buffer layers deposited by Ion Beam Assisted Deposition (IBAD) were used. A Liquid Precursor Delivery system was designed, constructed and used in the MOCVD facility at Intermagnetics for the reported work. A critical current ($I_c$) of 97.5 A corresponding to a critical current density ($J_c$) of 1.3 MA/cm$^2$ and 130 A/width of tape was achieved at 77 K in self-field conditions in YBCO film grown by MOCVD on IBAD substrate.**

***Index Terms* --- MOCVD, IBAD, current density, biaxial-texture**

## I. INTRODUCTION

Intermagnetics has been developing a Metal Organic Chemical Vapor Deposition (MOCVD) process for the fabrication of high current $YBa_2Cu_3O_x$ (YBCO) conductor using substrates prepared by Ion Beam Assisted Deposition (IBAD). MOCVD has several special features such as non line-of-sight coating, separation of precursors from deposition chamber, high deposition rates, and no target fabrication expense, all of which can be important for production of long-lengths of YBCO conductor for electric-power applications. The viability of YBCO for producing high-current tapes was first demonstrated using IBAD substrates [1]. Only IBAD substrates have been used to demonstrate a 100-A YBCO tape in meter-long lengths [2]. In an earlier report, we had published achievement of both high critical currents (50 A) and critical current densities (> 1 MA/cm$^2$ at 77 K) in YBCO films fabricated by MOCVD with IBAD substrates [3]. The performance of the MOCVD-prepared YBCO films in the presence of high magnetic fields was shown to be comparable to that of Pulsed-Laser Deposition (PLD)-prepared YBCO films. In this publication, we report the use of a liquid precursor delivery system, a doubling of the critical current to almost 100 A and microstructure analysis of MOCVD-prepared YBCO films.

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## II. EXPERIMENTAL

The metal substrates used for MOCVD of YBCO consisted of biaxially-textured buffer layers of Yttria-Stabilized-Zirconia (YSZ) deposited by IBAD at Argonne National Lab (ANL) and Los Alamos National Lab (LANL). Polished Hastelloy-C and Inconel 625 metal substrates, about 100 µm in thickness were used. The thickness of the YSZ buffer layers was typically 0.5 µm to 1 µm thick. In the process used at ANL, an 8-cm Kaufmann-type DC ion source was used to produce a 300-eV assisting ion beam of Ar/10%$O_2$ that was directed on the substrate surface at an angle of 35°. Electron-beam evaporation was used for YSZ deposition. The atomic deposition rate was 1.6 Å/s and the ion flux was 200 µA/cm$^2$ to yield an ion-to-atom flux ratio of 2.8. The details of the IBAD process used LANL have been published elsewhere [4]. Briefly, Inconel 625 substrates were polished to a surface roughness of 3±1 nm. The assist ion source was a linear dc Kaufmann-type, with beam dimensions of 2.5 x 23 cm. The atomic deposition rate was 0.5 Å /s, the assist ion energy was 200 eV, and the ion flux was 190 µA/cm$^2$. The YSZ-buffered metal substrates were cut into 1 cm long, 4 to 8 mm wide segments for YBCO deposition by MOCVD. In addition to the IBAD substrates, single crystal YSZ substrates and $SrTiO_3$ substrates were also used for deposition to serve as a comparison.

MOCVD of YBCO was conducted in a custom-built facility at Intermagnetics described previously [3], [5]. Tetramethyl heptanedionate (thd) precursors were used for Y, Ba, and Cu. YBCO films, 1 micron in thickness, were deposited within a temperature regime of 700 to 800°C in a reactor pressure of 1 to 5 Torr and an oxygen partial pressure of 0.3 to 0.8 Torr. After deposition, YBCO films were coated with silver pads, annealed around 400°C in oxygen.

The out-of-plane-and in-plane texture of the YSZ buffer layers were examined by X-ray Diffraction including polefigure measurements. The surface morphology of the YBCO films was examined by Scanning Electron Microscopy (SEM) followed by compositional analysis by Energy Dispersive X-ray Spectroscopy (EDS). The thickness of the films was measured by surface profilometry. The current density of the films was tested in liquid nitrogen in zero applied magnetic field. The samples were not patterned and the $J_c$ measurements were conducted across the entire width of the sample. Continuous D.C. currents up to 100 A were used in the measurements.

## III. LIQUID PRECURSOR DELIVERY

The schematic in fig. 1 shows the configuration of the MOCVD system at Intermagnetics. The process consists of 3 steps related to precursor delivery. In the first step, a liquid precursor is pumped at a constant flow rate using a liquid delivery pump. In the second step, the liquid precursor is flash-vaporized in a vaporizer maintained at a constant temperature. The vaporized precursor is then flowed in a carrier gas such as Ar through heated lines and then injected on a hot substrate through a showerhead. All components of the precursor delivery system have been designed and developed at Intermagnetics.

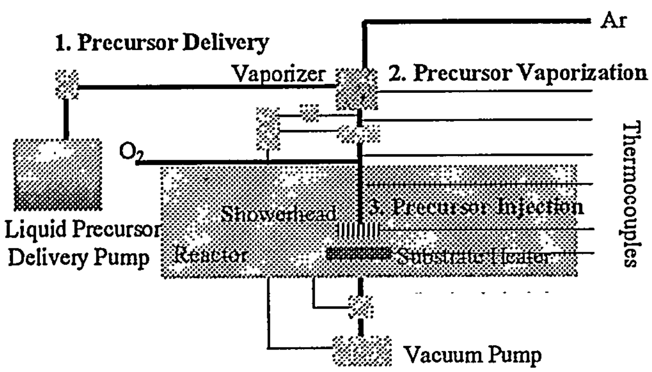


Fig. 1. Schematic of the MOCVD system used at Intermagnetics

There are several advantages to the liquid precursor delivery system compared to solid precursor delivery. The key points are listed in Table I.

The composition of YBCO films prepared using the Liquid Precursor Delivery System over a course of dozen deposition runs showed that the film composition was reproducible within 5% of either side of the target composition.

## IV. CRITICAL CURRENT MEASUREMENTS

It was reported previously that critical currents ($I_c$) of 50 A were achieved in YBCO films grown by MOCVD on IBAD substrates at 77 K in self-field conditions [3]. High values of critical currents were found to be maintained also in the presence of a magnetic field. At 75 K, with the magnetic field perpendicular to the c-axis ($B \perp c$), critical current density ($J_c$) values of 570,000 A/cm$^2$ and 240,000 A/cm$^2$ were achieved in fields of 1 and 5 T respectively. With the field parallel to the c-axis ($B \parallel c$), $J_c$ values of 215,000 A/cm$^2$ and 100,000 A/cm$^2$ were achieved in fields of 1 and 2.5 T respectively. At 64 K, the $J_c$ values were 1.2 MA/cm$^2$ and 770,000 A/cm$^2$ at 2 and 5 T respectively in the $B \perp c$ orientation. In the $B \parallel c$ orientation, the $J_c$ values were 680,000 A/cm$^2$, 520,000 A/cm$^2$ and 300,000 A/cm$^2$ at 1, 2, and 5 T respectively. Since that report, the critical current of YBCO films made by MOCVD on IBAD YSZ substrates has been doubled to almost 100 A as shown in the Current-Voltage (I-V) curve in fig. 2. The critical current of 97.5 A exhibited in fig. 2 corresponds to a critical current density of 1.3 MA/cm$^2$ and translates to 130 A/cm width of film. These values compare very well with those achieved in YBCO films deposited on buffered-metal substrates using Pulsed Laser Deposition.

Table I
COMPARISON OF SOLID- AND LIQUID-PRECURSOR DELIVERY APPROACHES FOR MOCVD

| | Solid-state Precursor | Liquid-state Precursor |
|---|---|---|
| Technique | Precursors are individually sublimed separately from 3 separate ovens | Precursor is flash vaporized at a single location |
| Temperature Control | Difficult (film composition is sensitive to even 1°C change) | Single-point of control. Easier |
| Precursor Stability | Bad for long deposition periods | More stable in solution form |
| Precursor Vapor Pressure | Changes as precursor is depleted | Constant volume of precursor is vaporized. Vapor pressure is constant |
| Deposition Rate | Difficult to increase due to turbulence of carrier gas at high rates | Easily increased by increasing precursor flow rate. |

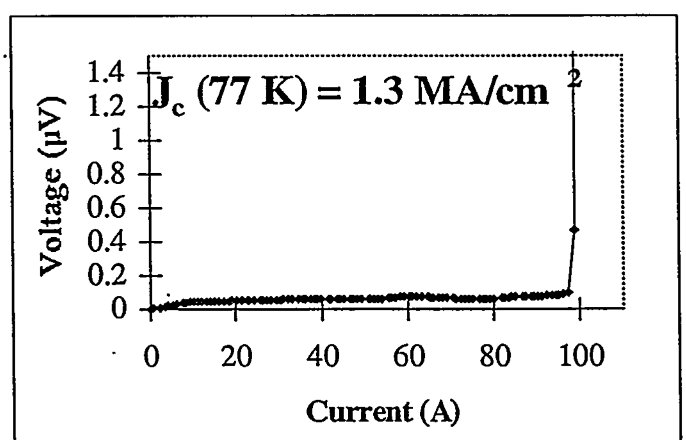

Fig. 2. I-V curve obtained from a YBCO film deposited by MOCVD on an IBAD substrate. The critical current is 97.5 A that corresponds to 130 A/cm width of tape.

## V. MICROSTRUCTURE ANALYSIS

The surface morphology of the films was examined by SEM. It was found that the surface morphology of the films became rougher with increasing thickness from 0.1 to 1 micron. A puzzling finding was that the rougher films almost always yielded a higher $J_c$ than the smoother films. Figures 3 and 4 show a relatively smooth film and a very rough film, both 1 micron in thickness. Contrary to expectations, the smooth film sustained a critical current of only 1 A while the rough film exhibited a critical current of almost 100 A.

The cause of surface roughness in films of the same thickness as seen in figures 3 and 4 could be due to that fact that the rough films are usually off-stoichometric. Off-stoichiometric compositions were deliberately chosen to achieve higher $J_c$.

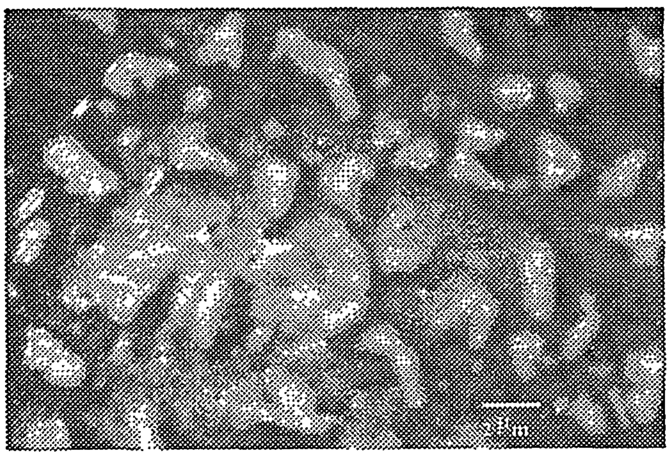

Fig. 3. Microstructure of an 1 micron YBCO film deposited by MOCVD on an IBAD substrate In spite of the relatively smooth film surface, the film sustained a critical current of only 1.3 A.

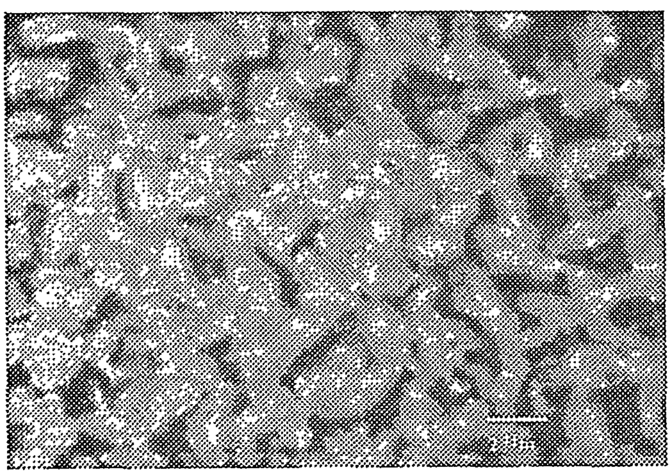

Fig. 4. Microstructure of an 1 micron YBCO film deposited by MOCVD on an IBAD substrate In spite of the rough film surface, the film sustained a critical current of 97.5 A (data shown in fig. 2).

The information obtained from the analysis of the surface morphology of the films does not provide a complete detail of the microstructure and its influence on the critical current. Therefore, steps have been taken to examine the cross sectional microstructure of MOCVD-prepared YBCO films. Samples were fractured and the cross sections were examined using SEM. Figure 5 shows a microstructure of a YBCO film on $SrTiO_3$ substrate. The microstructure shows that the film is dense with a columnar structure. EDS spectra were obtained from various sections of the sample and the YBCO and $SrTiO_3$ substrate regions were confirmed.

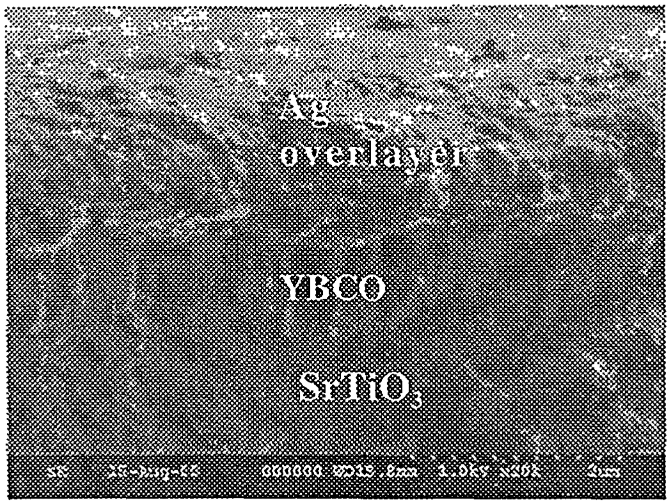


Fig. 5 Fracture surface of a YBCO film on $SrTiO_3$ substrate. A columnar grain growth is seen. The film appears to be dense.

More detailed analyses will be performed including on YBCO on metal substrates to understand the relationship between the microstructure and $J_c$.

## V. CONCLUSIONS

YBCO films have been deposited by MOCVD on biaxially-textured YSZ buffer deposited by IBAD. A Liquid-precursor delivery system was designed and constructed for use in the MOCVD facility. Through improvement to the MOCVD equipment and optimization of process conditions, critical current as high as 97.5 A corresponding a $J_c$ of 1.3 $MA/cm^2$ was achieved at 77 K in self-field conditions. This value corresponds to 130 A/cm width of tape. Microstructure analysis of the tapes reveal a very rough surface morphology in the high $J_c$ films.

## REFERENCES

[1] Y. Iijima, N. Tanabe, and O. Kohno, *Adv. in Supercond. IV*, ed. Hayakawa and Koshizuka Springer Verlag, Tokyo, pp. 679-682 (1992).

[2] S. R. Foltyn, P. N. Arendt, P. C. Dowden, R. F. DePaula, J. R. Groves, J. Y. Coulter, Q. Xia, M. P. Maley, and D. E. Peterson, *IEEE Trans. Appl. Superconductivity* vol. 9, 1519 (1999). also 1999 Wire Development Workshop Proceedings (U.S. Dept. of Energy), Cocoa Beach, FL, January 12-13, 1999, pp. 215-228.

[3] V. Selvamanickam G. Galinski, G. Carota, J. DeFrank, C. Trautwein, P. Haldar, U. Balachandran, and M. Chudzik, J. Y. Coulter, P. N. Arendt, J. R. Groves, R. F. DePaula, B. E. Newnam, and D. E. Peterson *Physica C* vol. 333 pp. 155-162 (2000).

[4] P.N. Arendt, S.R. Foltyn, J.R. Groves, R.F. DePaula, P.C. Dowden, J.M. Roper, and J.Y. Coulter, *Appl. Superconductivity* vol. 4, 429. (1996).

[5] V. Selvamanickam, G. Galinski, J. DeFrank, C. Trautwein, P. Haldar, U. Balachandran, and M. Chudzik, *IEEE Trans. Appl. Superconductivity* vol. 9, 1523 (1999).