IMAGE PROCESSING PLUME FLUENCE FOR PROCESS CONTROL OF PULSED-LASER THIN-FILM DEPOSITIONS

J.G. Jones', R.R. Biggers', N.C. Boss', J.D. Busbee', D.V. Dempsey', G. Kozlowski$^{12}$

$^1$Air Force Research Laboratory, Materials Directorate
Wright-Patterson AFB, OH 45433-7916
University of Dayton Research Institute, Dayton, OH 45409

$^2$Wright-State University, Dayton, OH

Abstract: Process control is a crucial element in all deposition techniques. It is especially elusive in the versatile and efficient deposition technique known as pulsed-laser-deposition (PLD). Image processed emissions as well as signal processed intensity measurements from the plume of a $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) target are monitored in situ to determine two dimensional spatial and time-of-flight (TOF) information about plume components. Manual and fuzzy-logic based regulation of laser energy and pressure, simultaneously, based on TOF feedback have resulted in improved film quality and repeatability of the PLD thin-film depositions. The plume was imaged under various deposition conditions, including in situ changing of beam focus relative to the target surface, chamber pressure, and laser beam energy. Analysis of the collected images will provide an increased understanding of the effect of changing environmental conditions in order to improve the deposited thin film quality. Copyright © 2000 IFAC

Keywords: Fuzzy Control, Image Processing, Image Sensors, Process Control, Spectroscopy

1. INTRODUCTION

Process control is being applied to the growth of superconducting YBCO thin-films to improve the quality of the films grown. Each superconducting thin-film has a unique critical temperature, $T_c$, below which the film can conduct electricity without loss, and a current density, $J_c$. Useful $T_c$ values are usually well above 77 K, a temperature which can be maintained using readily available liquid nitrogen. A useful minimum desired $J_c$ is $1 \times 10^6$ A/cm$^2$. These properties, $T_c$ and $J_c$ can be measured ex situ for each film to quantify the quality of individual films.

These films grown by pulsed laser deposition (PLD) were affected by environmental chamber conditions which include chamber pressure, substrate temperature, target rotation rate, beam footprint area, target to substrate distance, movement of laser beam across the target, laser voltage, laser beam pulse length, and repetition rate (Biggers, 1997). Some of these characteristics of the deposition system cannot be adjusted and will vary with each PLD apparatus (Geotech, 1994).
However, a few of these process parameters can be adjusted in situ to regulate dynamics of the plume that are generated by each pulse of the laser. Minimization of process drift can be achieved by application of real-time adjustments to process variables (Buggers, 1998).

A fuzzy logic based controller has been implemented on a Wintel computer to continuously monitor the deposition process and adjust both the laser excitation voltage (kV) and chamber pressure (mTorr) to maintain a desired time of flight (TOF) (Jones, 1998). The TOF is the time calculated from initial target impact of the laser pulse to the time of maximum emission of a particular species as the components pass the viewing position. Fig 1. For YBCO, barium (553 nm) and copper (327 nm) plume species are monitored at a point half way between target and substrate, and also simultaneously at some chosen point closer to the substrate. The operator may designate any one of these four channels as the control or feedback channel. A TOF is specified for the component monitored by this designated channel. The control system then adjusts the laser voltage and chamber pressure simultaneously to maintain this specified TOF setpoint. Experimental data will show the effect of changing the position of a beam focusing optic (energy density distribution in beam footprint) and whether it can be used in situ as a third control variable. Using this method of process control, variability from deposition to deposition can be reduced. However, the operator must know a priori what TOF settings correspond to growth conditions correlated to deposited films having high quality as exemplified by ex situ Tg and Jc measurements.

2. EXPERIMENTAL

A Neocera PLD chamber, shown in Fig. 1, is used to grow YBCO superconducting thin films on nickel substrates. The substrate and corresponding heater assembly were heated to 760°C prior to deposition, in an ambient background pressure of 150 mTorr of oxygen. The Neocera PLD chamber provides for up to five 5.08 cm targets to be held in a carousel, with the desired target selected as needed. A Lambda Physik 305 cm3 laser was used at a 4 Hz repetition rate, with laser voltage settings from 15.5 to 23 kV. When operated at 4 Hz, the laser produces pulses of 248 nm photons with nominal energies of ~330 mJ pulse in a 15 mm2 footprint at the YBCO target. An optical train delivers beam energy to the YBCO target and includes an adjustable lens for variable beam focusing and an aperture to make the laser beam fit on the beam steering mirror. The lens can be adjusted over a 20 cm range, numbered from 60 to 80 cm, with the calculated focus occurring at the target surface when the lens is at the 74.2 cm position. At the nominal optical position of 67 cm, the target is positioned 7.2 cm ahead of beam focus.

A Newport two axis motor controller was used to continuously scan the laser beam in a rectangular fashion across the 5.08 cm diameter target. Monitoring of the YBCO plume generated from each laser pulse is performed using two types of in situ sensors. A CCD camera with a filter for barium (553 nm) is used to monitor the two-dimensional spatial intensity of the plume from an overhead position. Fig. 2, while four photomultiplier tubes and corresponding filters are used to simultaneously monitor the line-of-sight intensity of the plume at four separate positions along the horizontal. Fig. 3. A computer continuously captures the CCD images and four channel PMT waveforms, as well as facilitating the operation of the process through a graphical user interface (Jones, 2000).

Plume images captured using a CCD based monochrome NTSC camera were collected using a Windows NT 4.0 dual processor 450 MHz Pentium II computer with a National Instruments PCI-1408

![Fig. 1. Pulsed laser deposition research chamber. Plume generated by each laser pulse travels orthogonal from the target surface.](image1)

![Fig. 2. Plume image taken with a chamber background environment of 150 mTorr of oxygen, 760°C, 67 cm focus, and a laser voltage setting of 20.5 kV.](image2)
monochrome acquisition board and LabVIEW 5.x. Each image collected represents one NTSC frame, which is composed of two interleaved fields each lasting 16.67 ms in duration. Since the NTSC CCD camera is continuously transmitting plume images, a trigger signal that is generated when the laser fires is used to indicate which image should be captured. Since each field represents 16.67 ms and the plume emits for only 50 ms, software was written to linearly interpolate the even or odd blank field that will occur with each captured frame.

A 553 nm ±5 nm filter was used in conjunction with a polarizer and CCD camera to image essentially only the excited barium emissions which are emitted from the plume. A polarizer reduces saturation of the CCD by reducing the light reaching the camera by more than 50%. Using software that was developed in LabVIEW, the images can be collected, processed, and archived to a file at approximately 2 Hz. Each experimental setting consists of collecting 60 seconds of data and results in more than 100 images that are 640 x 480 pixels each.

A Photomultiplier tube, a filter for barium (553 nm) or copper (327 nm) and a collimating tube makes up a fixed wavelength emission sensor (ES) that can monitor a line-of-sight intensity of the YBCO plume at a fixed position with very high resolution. Four of these sensors, two colinear pairs, were constructed to provide four simultaneous measurements, two for barium and two for copper, of each plume. One copper and barium ES colinear pair is mounted halfway between the target and substrate, while the other copper and barium ES colinear pair is mounted on a Nikon Biostation x-y stepper motor controlled stage. This allows for remote adjustment of the sensors, horizontally and vertically, to precisely monitor the much weaker plume emission that occurs near the substrate.

These four sensors are simultaneously monitored and recorded using a Tektronix four-channel digital phosphor oscilloscope with a 500 MHz bandwidth. A PCI GPIB data acquisition card facilitates high-speed transfer of the collected waveforms from the oscilloscope to the lab computer on a continuous basis during depositions. With this configuration all four ES waveforms can be continuously recorded using a 15,000 point per channel resolution and can be remotely collected by the computer at approximately a 1.5 Hz rate.

During a typical deposition several collected sets of waveforms are recorded, but five key parameters about the waveforms are continuously calculated and recorded. The parameters calculated are: time of the fireball that occurs when the laser beam impinges on the target, time of peak emission, time difference (TOF) between the first two parameters, the peak intensity of the plume species, and the average intensity of area under the emission signal of the plume species.

Using this sensor suite two experiments were performed in which the laser excitation voltage and laser beam focus position relative to the target position were independently varied. The laser excitation voltage was varied between 15.5 and 25.0 kV in steps of 0.5 kV under nominal deposition conditions of 760°C substrate temperature, laser beam focus position of 67 cm, and a chamber pressure of 150 mTorr of oxygen. For each laser voltage setting, deposition conditions were maintained for 60 seconds while data was collected using the CCD camera and ES suite. This procedure was repeated for laser beam focus positions of 60 to 80 cm with steps of 1 cm, while having the laser voltage set to 22.5 kV, the substrate temperature at 760°C, and the chamber pressure at 150 mTorr of oxygen.

3. RESULTS AND DISCUSSION

Shown in Fig. 4 is a plume image taken with typical deposition conditions of substrate temperature, chamber oxygen pressure, laser cavity voltage, and lens focus position: 760°C, 150 mTorr, 20.5 kV, and 67 cm respectively. Average row intensities, R, were calculated for each captured image horizontally along the most intense row from just next to the discernable edge of the target holder to the horizontal edge of the image. Due to extreme variations of the plume with changing pressures, vertical averages were taken at the most intense column, C, located between just in front of the target holder edge and the edge of the image. Since the laser beam is swept across the target in an x-y fashion, the plume will be randomly offset.
vertical due to the laser beam interacting with the YBCO target at different vertical positions, relative to the camera image. Similarly, there will be variation in the location of the most intense column due to variations in the pulse energy of the laser, as well as changes in pressure.

The average intensity, for both row and column intensity calculations, was computed by averaging all of the R and C values that were determined for each image. Before averaging, all of the images that were collected at approximately 2 Hz for 60 seconds during each laser voltage setting and laser beam focus position, were processed to remove any images that were significantly different due to laser beam variations, or other factors. The graphs shown in Fig. 4 show the effect of changing laser voltage and focus position on the calculated R and C parameters. Each graph shows increasing average intensity for the laser voltage, and also for the laser beam lens position. Clearly changes in the deposition parameters can be monitored. But also these measurements represent the integrated spatial intensity of the plume emissions, but with an arbitrary scale for intensity. For the image information to be useful, it must be correlated to deposition conditions, which produce good films.

Shown in Fig. 5 are average barium emissions from one of the four PMT ES measurements taken at 0.5 Hz rates and averaged for each 60 second deposition condition. This ES information was collected simultaneously with the images that were used for generating the plots in Fig. 4. Comparison of Fig. 4 and Fig. 5 show that the CCD camera approach to collecting plume emission information

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Fig. 4. Average intensity of captured image along most intense row (R) and most intense column (C) with (a) varying Laser kV (top) and (b) varying focus of laser beam on target (bottom).

Fig. 5. Average intensity of barium emission waveforms captured using ES located halfway between target and substrate for (a) varying Laser kV (top) and (b) varying focus of laser beam on target (bottom).

Fig. 6. Time of flight (TOF) of barium species (553 nm) measured halfway between target and substrate with (a) variations in laser voltage (top) and (b) variations in laser beam focus (bottom).
of a particular component can in fact provide a reliable measure of the average intensity. Since the camera is unshielded, it cannot provide any information as to the velocity of the emissions, such as can be provided by measuring the TOF.

The TOF graph for laser voltage, Fig. 6, continuously decreases with increasing voltage from 15.3 to 22.5 kV, with the exception of the 23.0 kV measurement which shows a slight increase in TOF. This is expected to some extent because this is the absolute maximum excitation voltage of the laser cavity. Similarly, the TOF continuously decreases for changes in focus from 60 to 80 cm. Since the TOF represents the time to peak emission of a particular emission species, the speed of emissions from the YBCO goes up with either increasing laser cavity voltage or increasing focal position. Fig. 6 shows that the TOF can be made to be significantly smaller by moving the focal position to between 75 or 80 cm, which is from near focus at the target surface to focus approximately 5 cm in front of the target. The beam footprint undergoes a narrowing and increase in energy density in the central region as seen in stationary target ablation at these lens positions. The closer to 80 cm where beam focus is 5.7 cm in front of the target, the deeper penetration of the target will become for a constant number of pulses. The significance of the laser beam focus TOF measurements is that the lens position can be adjusted in situ to compensate for TOF. Typically a good YBCO film is grown using process control to vary the laser cavity voltage and the pressure to maintain a desired TOF of 4.4 µs, but now a third control variable can be added to increase the flexibility of the YBCO deposition. Further, the camera can provide additional spatial information, beyond that of the PMT ES suite to verify the spatial distribution of a particular emission species, such as excited barium.

Shown in Fig. 7 is a Fuzzy logic controller that has been used for TOF regulation, but with an additional controller adjustment for the focus lens position. The Fuzzy controller uses one of the four ES TOF measurements, chosen by the operator along with a desired setpoint the TOF, to continuously update at 0.5 Hz the setpoints of the laser cavity voltage and the chamber pressure based on a prescribed rule base. The rule base uses Fuzzy membership functions to define degrees of membership of five prescribed TOF error membership functions and three prescribed TOF error rate membership functions (Jones, 1997). Based on the degree of membership of these eight membership functions, a Fuzzy associative memory with centroid defuzzification is used to determine the numerical change that needs to take place in the laser cavity voltage. The Fuzzy controller that has been used in the past defines the change in pressure as being a scalar multiple of the calculated change in laser cavity voltage.

The first attempt at using this Fuzzy logic methodology to control all three of the PLD control variables simultaneously has not been attempted yet. Additional components are required to accomplish this task including a Newport MM4005 GPIB based programmable motor controller, and a 850F automated micrometer. A micrometer can be used, instead of using a large linear table and moving the lens from 60 to 80 cm, by adding a second lens to the optical train shown in Fig. 1. The addition of a second lens results in a telescoping arrangement of the laser beam in which slight adjustments to the second lens position will result in very large changes in the distance of focus from the movable lens.

4. CONCLUSIONS

The PLD plume has been monitored using four PMT emission sensors with a four channel oscilloscope being used to simultaneously monitor two different species half way between target and substrate as well as near the substrate. The two PMT ES for excited copper and excited barium emissions (527 and 553 nm) that are near the substrate are mounted on a movable x-y stage providing significant flexibility in determining plume features. The PMT ES waveforms are collected using a computer through a GPIB bus, processed to extract key parameters including TOF, in situ at a 0.5 Hz rate. Using a Fuzzy logic controller, updates to laser voltage and chamber pressure are made, also at 0.5 Hz, to maintain the desired TOF using one of the four ES channels. Based on the experimental data collected from adjustment of the laser beam focus position, Fig. 4, 5, and 6, in situ adjustment of the laser beam focus position should provide improved TOF regulation. Certainly the range of adjustment of the focus position must be limited to a range which will provide acceptable laser beam footprints on the YBCO target.

Fig. 7. Fuzzy controller block diagram showing three simultaneous actuators for controlling the desired emissions: 1) Laser kV, 2) Chamber pressure, and 3) Spot size of laser beam on target.
The use of a NTSC CCD camera provides an inexpensive method to monitor the integrated plume emissions, or a particular emission species, when using a narrow-band filter. Using a polarizer reduces the light reaching the camera by more than 50% to reduce the effects of saturation on the CCD array. Use of a less expensive monochrome acquisition video card is not prohibitive, since a narrow-band filter allows only a fixed range of 10 to 20 nm width to be monitored, relative to the 400 to 700 nm sensitivity of the CCD array. As shown in Fig. 4 and 5, the average intensity measured using a particular column of the CCD images captured is equivalent to the average intensity of PMT ES waveforms collected at some line-of-sight position between the target and substrate. Images collected provide additional spatial information, which may be of interest, such as the location of the broadest part of the plume between the target and substrate. Although the TOF is maintained with process control, CCD images show spatial changes that occur with changing environmental parameters. These spatial images may correlate with factors controlling the final quality of films deposited under identical TOF conditions.

The Fuzzy logic based process controller designed for TOF regulation, by controlling the laser voltage and chamber pressure will be expanded to include adjustment of laser beam focus. Experimental data collected shows that increasing the laser beam focus, which brings the target surface closer to the beam focus, will cause monotonic changes to both the average CCD intensity and PMT ES intensity for excited barium species, as well as continuously decrease the TOF. Correlating the image capture information to TOF and PMT ES averages has significant potential to inexpensively monitor and thereby understand PLD in order to implement multivariable process control.

REFERENCES