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# Chemical mechanical polishing characteristics in  $(Bi, La)Ti<sub>3</sub>O<sub>12</sub>$ damascene process for high-density ferroelectric memories

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#### Abstract

Ferroelectric thin films such as Pb(Zr,Ti)O<sub>3</sub>, SrBi<sub>2</sub>Ta<sub>2</sub>O<sub>9</sub>, and Bi<sub>3.25</sub>La<sub>0.75</sub>Ti<sub>3</sub>O<sub>12</sub> (BLT) thin films have been widely investigated for nonvolatile ferroelectric memories. BLT thin films show advantages such as highly fatigue resistance, low processing temperature, and large remanent polarization. The patterning of these ferroelectric thin films with a vertical sidewall and without plasma damage is strongly required. Chemical mechanical polishing (CMP) process was investigated for the vertical sidewall patterning of BLT thin films. Removal rate and within-wafer nonuniformity (WIWNU%) were examined by change of process parameters. Potential of hydrogen (pH) in slurry was varied for an improvement of the removal rate and WIWNU%. Surface roughness of BLT thin films after CMP process for the improvement of densification was inquired into by atomic force microscopy. The excellent performance such as 188.4 nm/min of removal rate, 2.61% of WIWNU%, 0.95 nm of root mean square roughness, and 6.94 nm of peak-to-valley roughness was obtained. This result will lead the CMP process to pattern the BLT thin films for the vertical sidewall without plasma damage.

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# 1. Introduction

Ferroelectric random access memory (FRAM), which is a non-volatile memory, will be expected to replace the current dynamic random access memory in the near future due to its ideal memory properties such as non-volatility, low power consumption, high speed, and almost unlimited endurance  $[1-3]$  $[1-3]$ . The key component of FRAM is a ferroelectric capacitor consisting of a ferroelectric thin film sandwiched with top and bottom electrodes [\[4\].](#page--1-0) Some ferroelectric thin films such as Pb  $(Zr, Ti)O_3$  (PZT), SrBi<sub>2</sub>Ta<sub>2</sub>O<sub>9</sub> (SBT), and Bi<sub>3.25</sub>La<sub>0.75</sub>Ti<sub>3</sub>O<sub>12</sub> (BLT) thin films have been widely investigated for non-volatile ferroelectric memories [5–[8\].](#page--1-0) BLT thin film shows advantages such as higher fatigue endurance than PZT thin film, lower processing temperature than SBT thin film, and large remanent polarization [\[8,9\].](#page--1-0) Meanwhile, these ferroelectric thin films were patterned by a plasma etching process for high-density ferroelectric memories, however, an angled sidewall was reported in the plasma etching [\[10\]](#page--1-0). The angled sidewall prevents the

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densification of ferroelectric memory and is apt to receive the plasma damage [\[10\]](#page--1-0). Therefore, the patterning with a vertical sidewall of the ferroelectric thin films is necessary by the development of process technology; the chemical mechanical polishing (CMP) characteristics of BLT thin film were investigated for the vertical sidewall by damascene process of CMP in this study [\[11\]](#page--1-0). Surface roughness of BLT thin film was necessary to be improved for the densification of multilevel memory structure. Removal rate, within-wafer non-uniformity (WIWNU%), and surface roughness were examined by the effects of chemical and mechanical parts in CMP process.

## 2. Experiments

All the BLT samples in this paper were spin coated on Pt/Ti/ SiO2/Si substrates. P-type (100) orientation, 6-inch diameter silicon wafers (1–30  $\Omega$ cm) were used as the starting substrates. The substrate was cleaned by rinsing with the solution of  $H<sub>2</sub>SO<sub>4</sub>:H<sub>2</sub>O<sub>2</sub>$  (1:4),  $H<sub>2</sub>O:HF$  (DHF; 10:1), and de-ionized water in order to eliminate the native silicon oxide. Dry thermal oxide of 100 nm was grown by electric furnace in  $O_2$  gas flux at 1100 °C. Titanium of 30 nm for the adhesion and platinum of

120 nm were deposited by DC sputtering at room temperature under the conditions of Ar gas flux,  $1.0 \times 10^6$  Torr vacuum, 300 W of DC power with a 4-inch Ti target (99.999% purity) and a 4-inch Pt target (99.99% purity) respectively. Precursor solution for  $Bi_3,25La_0,75Ti_3O_12$  was prepared by the sol–gel method using bismuth (III) acetate [Bi(CH<sub>3</sub>CO<sub>2</sub>)<sub>3</sub>], lanthanum– acetate hydrate  $[(CH_3CO_2)La·H_2O]$ , titanium (IV) iso-propoxide  $\{Ti[OCH(CH_3)_2]_4\}$ , solvents of an acetic acid  $[CH_3CO_2H]$ , and 2-methoxyethanol  $\left[CH_3OCH_2CH_2OH\right]$  [\[8\]](#page--1-0). The film was deposited by the spin-coating technique at 3000 rpm for 30 s. After the spin-coating procedure the film was dried at 400 °C for 1 min to remove the organic contaminations. Then, the prebaked films were annealed at 700 °C for 1 h in oxygen ambient for crystallization. The final thickness of BLT thin film was about 124 nm. All test wafers were polished by CMP with a G&P POLI-450 CMP polisher. The parameters of CMP process are summarized as follows: head speed, slurry flow rate, and polishing time were 50 rpm, 90 ml/min, and 30 s, respectively. The ranges of table speed and down force were changed from 20 rpm and 9.8 kPa to 60 rpm and 29.4 kPa. The conditioning pressure was fixed by 2 kg/cm<sup>2</sup> for 60 s. IC-1400 of Rohm and Haas Electronic Materials Company was used for polishing pad. The silica  $(SiO<sub>2</sub>)$  abrasive slurry of pH 11.3 was used for BLT-CMP. The pH was adjusted from 10.3 to 12.3 using the ACS standard buffer solutions supplied from Fisher Scientific. To prevent aging effect, the slurries before polishing were dispersed by ultrasonic wave homogenizer of Sonic Tech. The post-CMP cleaning was carried out by a sequence of 3 min in SC-1 chemicals (NH<sub>4</sub>OH:H<sub>2</sub>O<sub>2</sub>:H<sub>2</sub>O=1:2:7), 2 min in diluted HF of 1:10, and 4 min in ultrasonic cleaning. The thickness of 9 points from the center to the edge was measured clockwise on each wafer using ellipsometer (J. A. Woollam Co., M-2000 V). The surface morphology of BLT thin film after CMP process was measured with atomic force microscopy (AFM; PSIA, XE-100).

## 3. Results and discussion

The removal rate of BLT thin films are shown as a function of pressure (down force) and velocity (table speed) in Fig. 1. The highest removal rate of BLT thin films was 116.50 nm/min



Fig. 2. WIWNU% of BLT thin films polished with changes of pressure and velocity.

at 29.4 kPa of pressure and 60 rpm of velocity. According to Preston's equation, the removal rate should show a linear proportion to both pressure and velocity, however, the experimental result did not show it [\[12\]](#page--1-0). Hernandez et al. proposed the power law as a generalization version of Preston's equation for a better description of the removal rate as follow [\[13\]:](#page--1-0)

$$
RR = kp^a v^b \tag{1}
$$

where RR is the removal rate,  $p$  is the pressure,  $\nu$  is the velocity, and k is a proportionality constant independent of pressure and velocity [\[13\]](#page--1-0). The exponents  $a$  and  $b$  are used to gauge the contribution of pressure and velocity to the removal rate [\[13\]](#page--1-0). Because the Preston coefficient  $k$  depends on properties of pad, slurry, and materials and all other process parameters were fixed, fitting all removal rate data of BLT thin films in Fig. 1 with the power law yielded values of  $a=0.36\pm0.18$  and  $b=$  $0.28 \pm 0.13$  for the exponents of the pressure and velocity, respectively [\[13,14\].](#page--1-0) This result indicated that the removal rate of BLT thin films was more dependent on pressure than on velocity. Fig. 2 shows the WIWNU% of the polished BLT thin films as a function of pressure and velocity. The WIWNU% was generally decreased (improved) as pressure and velocity increased. The very good WIWNU% below 5.0% was obtained



Fig. 1. Removal rate of BLT thin films polished with changes of pressure and velocity.



Fig. 3. RMS surface roughness  $(R_{\rm rms})$  of the polished BLT thin films with changes of pressure and velocity.

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