



**RESEARCH ARTICLE**



**IMPROVEMENT OF THE INTERFACE PROPERTIES OF BaTiO<sub>3</sub> THIN FILM ON (100) SI BY  
INSITU RAPID THERMAL ANNEALING**

**V. R. CHINCHAMALATPURE<sup>1</sup> AND G. N. CHAUDHARI<sup>2\*</sup>**

<sup>1</sup>Hutatma Rashtriya Arts and Science College, Ashti, Distt: Wardha- 442202. India.

<sup>2\*</sup>Nanotechnology Research Laboratory, Shri. Shivaji Science College  
Amravati- 444603, India.

Corresponding Author: cgnroa@yahoo.com

**Abstract**

The interface properties of BaTiO<sub>3</sub> thin films were grown on (100) Si substrate by an insitu rapid thermal annealing technique were investigated. Initially Barium Titanate is prepared by mixing of Barium acetate and Titanium butoxide by using sol-gel method, the thin films deposited by a thermal annealing method at room temperature and 400°C were amorphous and crystalline phase respectively. The SEM of BaTiO<sub>3</sub> shows that the film as deposited is opaque and it revealed a high degree of porosity, after annealing crystallization increases. The X-Ray diffraction shows that the diffraction peaks sharp with increasing temperature which enhance the crystallinity of the films. The FTIR Spectra of BaTiO<sub>3</sub> annealed at 400°C shows the intense band at the edge of detection is assigned to Ti-O mode. The C-V curve shows that the capacitance remains constant within the bias range for as deposited BaTiO<sub>3</sub> film on Si and at 400°C annealing temperature shows large frequency dispersion in the accumulation region.

**Keywords:** In-situ rapid thermal annealing, barium titanate, XRD, FTIR, C-V.

**Introduction**

Barium titanate (BaTiO<sub>3</sub>) is a dielectric materials widely used in the electro-ceramic industry, electro optic devices (1-2). It is a ferroelectric material which has a high dielectric constant and low leakage current density. It is used in the electronics industry (3), as transducers and actuators piezoelectric effect (4) as high-K dielectric capacitors (5) and in memory applications which rely on the hysteresis between two stable states of polarization (6-7). Barium Titanate (BaTiO<sub>3</sub>), a well known dielectric material has been used as an insulating material to fabricate MIS structures. The investigation of micron to nanoscale ferroelectric materials thin films and particles have a deeper understanding of how size affects polarization and ferroelectric order, and the hypothesized importance of size effects in bulk ferroelectric systems (8-9). Contrasting views of the effect of

sample size on ferroelectricity have been debated from experimental and theoretical points of view (10).

Among various methods, BaTiO<sub>3</sub> can be prepared by Hydrothermal (11-12) Precipitation (13-14) and combustion (15) methods and BaTiO<sub>3</sub> film grown using pulsed laser de-Position (16), chemical vapor deposition (CVD) (17), metal organic chemical vapor deposition (18-19), polymeric precursor method (20), molecular beam epitaxy (MBE) (21) and r-f sputtering (22). The e-beam evaporation method shows relatively high deposition rate. So, some efforts have been made to prepare BaTiO<sub>3</sub> films by e-beam evaporation (23-24). Flash evaporation method, which evaporates instantaneously after dropping a small portion of source material onto a heater. Although BaTiO<sub>3</sub>

films can be made by flash evaporation, the deposition rate was too low for preparation of multilayer ceramic chip capacitors (MLCCs) (25).

In present work, BaTiO<sub>3</sub> films were deposited using rapid thermal annealing technique. This technique generates a vapor stream from source. The purpose of this study is to evaluate the variation of composition of sources and films with processing condition and to prepare BaTiO<sub>3</sub> films by this method with high deposition rate.

## Experimental

### Synthesis and deposition of BaTiO<sub>3</sub>

Barium acetate and Titanium butoxide were used as a starting material. BaTiO<sub>3</sub> material is prepared by sol-gel method. Si (100) wafers were well rinsed with warm acetone and ethanol followed by etching in H<sub>2</sub>SO<sub>4</sub> : H<sub>2</sub>O<sub>2</sub> : H<sub>2</sub>O (4: 1: 1) for 1 minute. The precursor was deposited on Si(100) wafers by rapid thermal annealing method.

To evaporate a material from source, the vapor is then transported to the coating surface. The system is largely composed of working chamber and gas flow chamber. To prepare the BaTiO<sub>3</sub> dielectric layer, BaTiO<sub>3</sub> source was evaporated using thermal evaporation technique.

The substrate Si is kept at a distance of 15 cm above the BaTiO<sub>3</sub> source. The experimental samples were grown at deposited temperature and at 400°C. The thickness of thin film BaTiO<sub>3</sub> was 150 nm. The BaTiO<sub>3</sub> was evaporated using thermal annealing and about 200 nm thick film of BaTiO<sub>3</sub> were deposited on Si at the rate of 0.2 nm /s. Thickness was controlled by using a water cooling arrangement. The details of the deposition conditions for the thin film BaTiO<sub>3</sub> are shown in Table (1)

Quantity	Condition
Chamber pressure	4.3 x 10 <sup>-2</sup> mbar
Substrate temperature ( °C )	As deposited, 400°C
Electron beam power (KV)	5
Source- Substrate distance (cm)	15
Evapourant	BaTiO <sub>3</sub>

Table (1): Deposition conditions for the BaTiO<sub>3</sub> film.

## Result and Discussion

### Structural Characterization

Fig.(1) shows that the SEM of film as deposited is opaque and it revealed a high degree of porosity, surface roughness increased in comparison to the optical quality films, after annealing crystallization increases and significant changes in surface morphology takes place.

Fig.(2) Powder X-ray diffraction results show that the BaTiO<sub>3</sub> nanoparticles are amorphous after the first-step thermal decomposition. Thus at higher temperature 400°C, the BaTiO<sub>3</sub> film fully crystallized. The thin films were characterized by X-ray Diffractometer using Cu K radiation = 1.5405 A.U.

The XRD pattern in Fig.(2) shows that after 400°C treatment, all diffraction peaks can be assigned to BaTiO<sub>3</sub> phase without any indication of other crystalline nature. The diffraction peaks are broad, indicating the formation of nanocrystals. The average crystal size can also be calculated as 55-56 nm from the Debye-Scherrer equation by taking account of the peak broadening at (111) diffraction line. The films deposited at room temperature were amorphous. However, the films deposited at 400 °C showed strong (110) and (111) peaks, suggesting that polycrystalline BaTiO<sub>3</sub> films.

In addition, the diffraction pattern in the 2 theta = 30°- 40° region is usually characteristics of the presence of either cubic or tetragonal BaTiO<sub>3</sub> structure. However, XRD result agrees with the recent study of 50 nm, BaTiO<sub>3</sub> nanomaterials using high energy X-ray radiation.

Fig (3) shows the infrared spectra of BaTiO<sub>3</sub> film on Si(100) as a function of temperature at 400 °C. The broad and strong band observed at 490 cm<sup>-1</sup> characteristics of Ti-O bond. Infrared analysis gave some important information about the phase transformations of the films on Si. In the infrared absorption bands, SiO<sub>2</sub> is found at 1090 cm<sup>-1</sup> because the oxide layer on Si surface grows during thermal treatment. It is also detected at this temperature that BaTiO<sub>3</sub> is already formed.

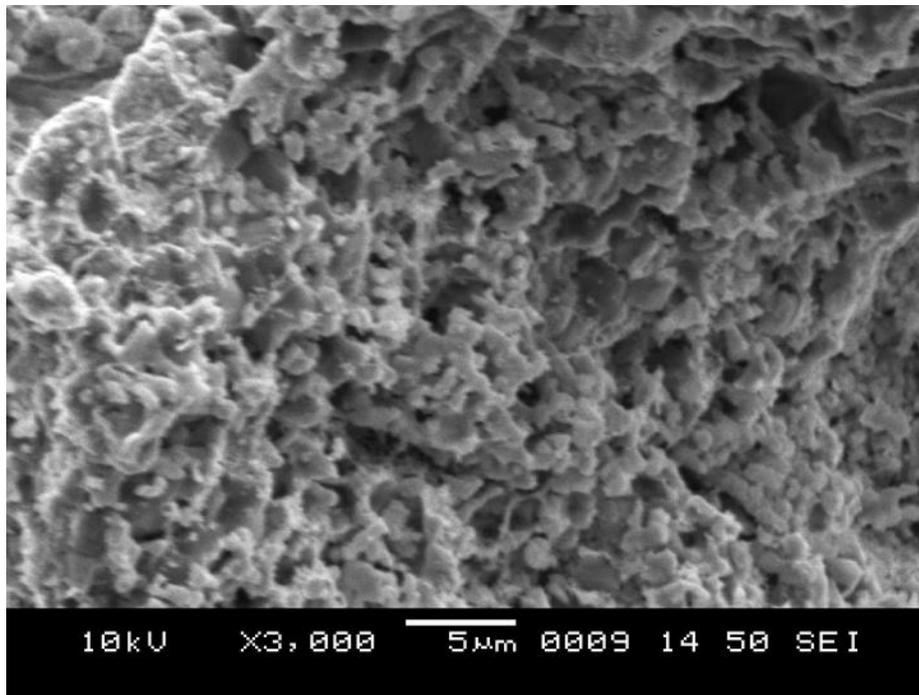
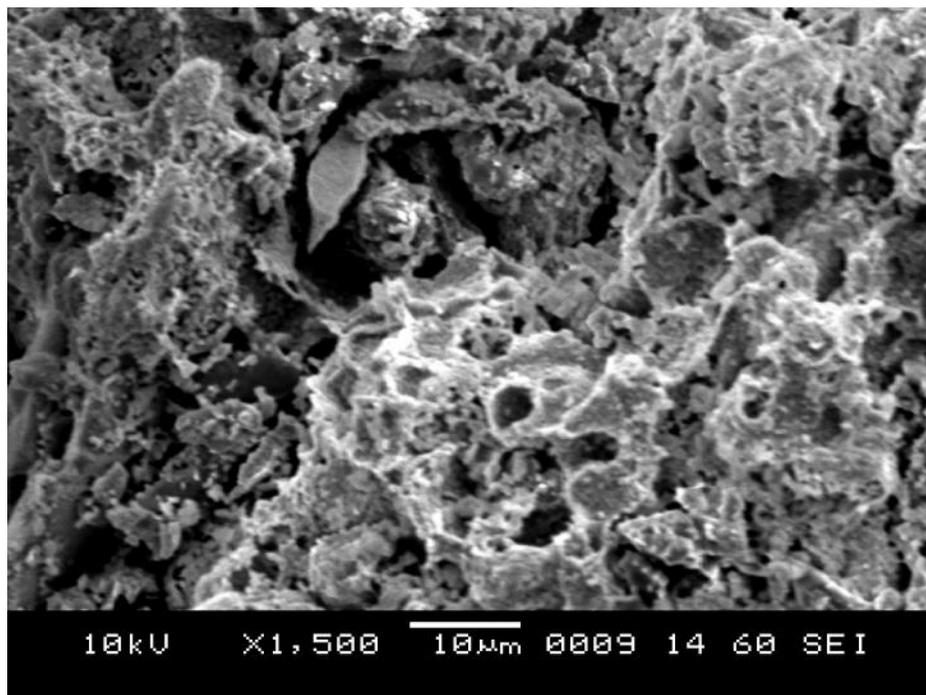


Fig.(1) ( a) shows SEM of BaTiO<sub>3</sub> powder at as deposited.



Fig(1) ( b) shows the SEM of BaTiO<sub>3</sub> powder annealed at 400°C

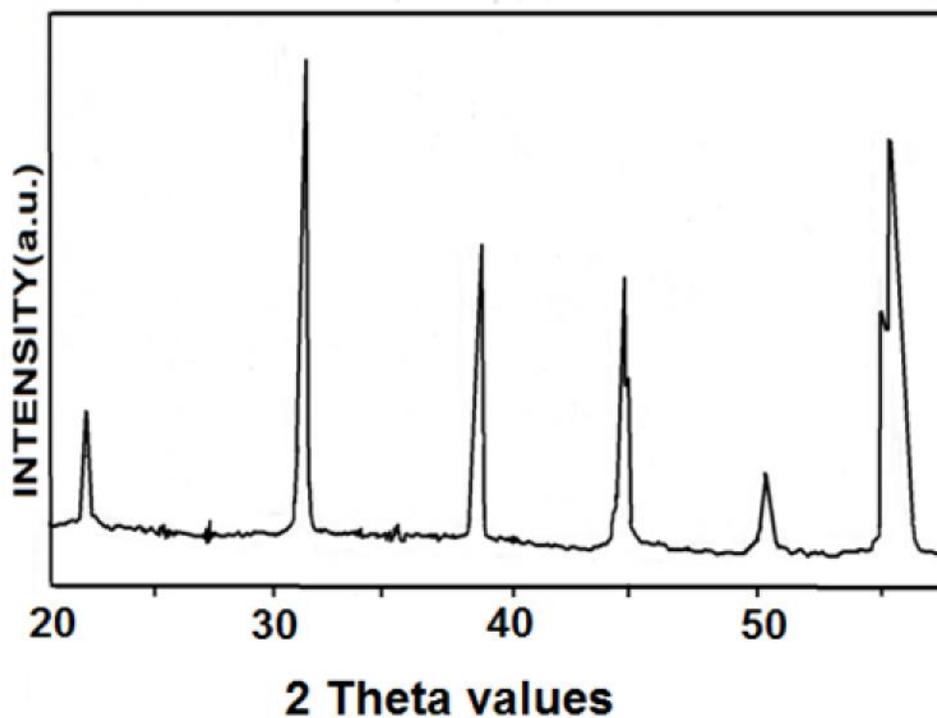


Fig.(2) XRD of BaTiO<sub>3</sub> powder annealed at 400<sup>o</sup> C.

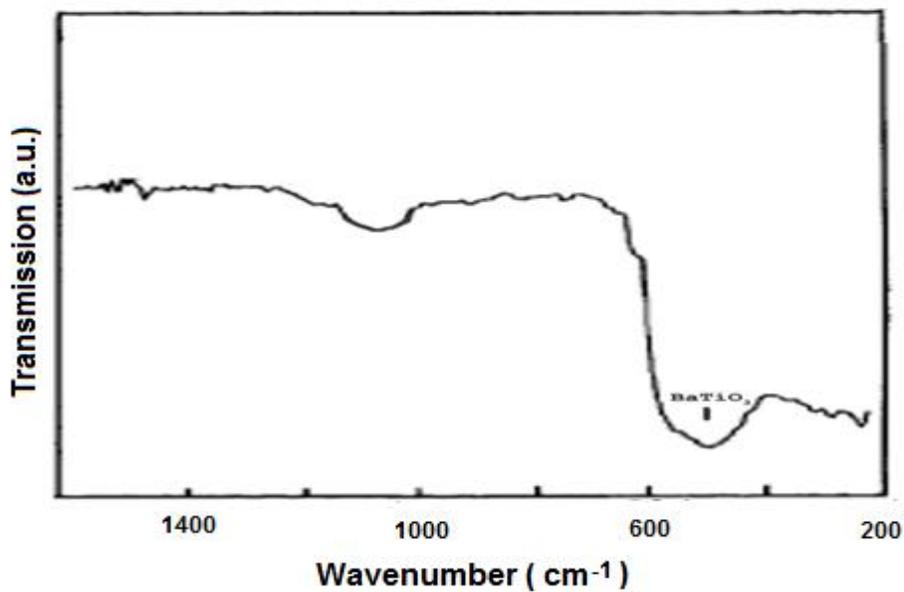


Fig. (3) FTIR of BaTiO<sub>3</sub> powder annealed at 400<sup>o</sup> C.

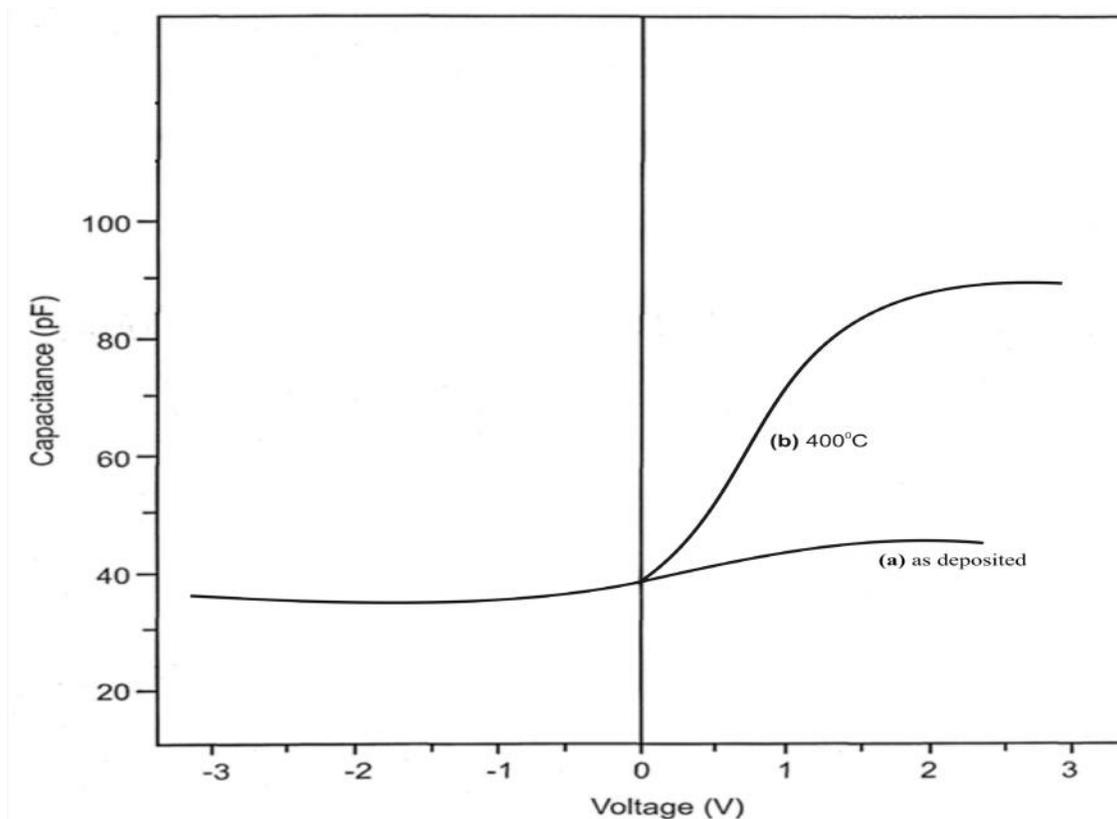


Fig.(4) shows C-V Characterization of BaTiO<sub>3</sub> thin film deposited on Si(100) at as deposited and rapid in-situ annealing at 400°C.

### Capacitance - Voltage (C-V) Characteristics

The electrical properties of BaTiO<sub>3</sub>/Si(100) were studied by using capacitance-voltage characteristics. Fig 4(a) and 4(b) shows 1MHz Capacitance-Voltage C-V characteristics for the sample at as deposited and 400°C of BaTiO<sub>3</sub>/Si(100). Fig. 4(a) shows that the capacitance remains constant within that bias range, which is indicative of a firmly pinned surface, this suggest either surface state are very fast or deposited layer is leaky.

Fig. 4(b) shows BaTiO<sub>3</sub> thin film deposited on Si(100) at 400°C showed large frequency dispersion in the accumulation region. The capacitance was changed from the inversion value to a value above flat band capacitance. The Capacitance-Voltage C-V characteristics of BaTiO<sub>3</sub> / Si(100) structure improved at 400°C shown in Fig 4(b).

### Conclusions

This method present in terms of added degrees of control in the manipulation of thin film properties for the preparation of thin films which exhibit highly uniform nanostructured texture and grain sizes. The BaTiO<sub>3</sub> / Si (100) structures were studied by structural and electrical characteristics. The XRD of BaTiO<sub>3</sub>/Si (100) indicates the enhance crystallinity of the films with annealing temperature at 400°C. The infrared spectra of BaTiO<sub>3</sub> film on Si as a function of temperature at 400°C, the broad and strong band observed at 490 cm<sup>-1</sup> characteristics of Ti-O bond. The C-V measurement of BaTiO<sub>3</sub> thin film deposited on (100)Si annealed at 400°C shows large frequency dispersion in the accumulation region. The result shows that the deposited BaTiO<sub>3</sub> insulator is good enough to use as gate insulator of MIS FET.

### Acknowledgment

We are thankful to University Grant Commission, Delhi for their financial assistance.

## References

1. Zhao , Z, Buscaglia V, Viviani M, T, Mitoseria, L, Testino, A Phys. Rev. B: Condens Matter, **70**, 24107, (2004).
2. Caruntu, G., Rarig Jr. R, Dumitru, I and O. Conner, C. J, Journal of Mater Chem.**16**, 752-758,(2005).
3. D. D. Fong, G. B. Stephenson, S. K. Streiffer, J. A. Eastman, O. Auciello, P. H. Fuoss, and C. Thompson, Science **304**, 1650 ,(2004).
4. N. A. Hill, Journal of Phys. Chem. B **104**, 6694, (2000).
5. D. Kotecki et al., IBM J. Res. Dev. **43**, 367 ,(1999).
6. B. H. Park, B. S. Kang, S. D. Bu, T. W. Noh, and W. Jo, Nature London ,**401**, 682 , (1999).
7. J. F. Scott, Ferroelectric Memories ,Springer Verlag, Berlin, (2000).
8. D.-J. Kim, J.-P. Maria, A. I. Kingon, and S. K. Streiffer, J. Applied. Physics. **93**, 5568, (2003).
9. G. Burns and F. H. Dacol, Phys. Rev. B **28**, 2527 , (1983).
10. B. E. Vugmeister and H. Rabitz, Phys. Rev. B **57**, 7581 , (1998).
11. Clark, I. J., Takeuchi, T., Ohtori, N. and Sinclair, D. C. J. Mater. Chem., **9**, 83–91. (1999)
12. Testino, A., Buscaglia, V., Buscaglia, M. T., Viviani, M. and Nanni, P.,Chem. Mater.**17**, 5346–5356. (2005).
13. Choi, G. J., Lee, S. K., Woo, K. J., Koo, K. K. and Cho, Y. S., Chem. Mater.**10**, 4104–4113. (1998).
14. Testino, A., Buscaglia, M. T., Viviani, M., Buscaglia, V. and Nanni, P., Journal of American Ceram. Soc., **87**, 79–83, (2004).
15. Anuradha, T. V., Ranganathan, S., Mimani, T. and Patil, K. C., Scr. Mater, **44**, 2237–2241, (2001).
16. T. T. W. Kim, Y. S. Yoon, S. S. Yom and C. O. Kim, Applied Surface Science, **90**, 75-80, ( 1995).
17. X. D. Wu, A. Inam, M. S. Hegde, B. Wilkens, C. C. Chang, S. Miura, and S. Matsubara, Applied Physics Letters, **54**, 754-756, (1989).
18. D. K. Fork, F. A. Ponce, J. C. Tramontana and T. H. Geballe, Applied Physics Letters, **58**, 2294-2296, ( 1991).
19. V. Agrawal and M. Liu, Journal of Materials Science, **32** , 619- 625,( 1997).
20. S. M. ZanettiL, E. R. Eite, E. Longo and J. A. Varela, Journal of Materials Research, **13**, 2932-2935,( 1998).
21. S. O. Brien, L. Brus, and C. B. Murray, J. American Chem. Soc. **123**, 12085, (2001).
22. K. Fujimoto, Y. Kobayashi, K. Yamamoto and Y. Bando, ibid , **56**, 527, (1990).
23. C. Liu, B. S. Zou, A. J. Rondinone, and Z. J. Zhang, J. American Chem. Soc.**123**, 4344, (2001).
24. R. A. Zarate, A. L. Cabrera, U. G. Volkmann and V. Fuenzalida, Journal of. Phys. Chem. Solids **59**,1639-1645, (1998).
25. S. Gu, T. J. Lu, D. D. Hass, and H. N. G. Wadley, Acta Mater. **49**, 2539-2547, (2001).