

## 2009 annual reports

Hiroshi Funakubo (Department of Innovative and Engineered Materials)

### 1. Highlights in 2009

#### 1) Development of Lead Free Piezoelectric Material

Novel lead-free systems have been developed using epitaxial films the novel mechanism applicable to wide range of materials is also proposed. In addition, the synthesized method below 300°C is under investigated.

#### 2) Characterization of Polar-axis oriented PZT Thick Films

Electrical and piezoelectric properties of polar-axis oriented-PZT agreed well with the prediction from the theory.

#### 3) Growth of Tunable Capacitor

General design to obtain the large tunability, i.e. the change of the capacitance with electrical field, was developed.

### 2. Articles

Original articles

- 1) “Good Conformability of Indium-tin-oxide Tin Films Prepared by Spray Chemical Vapor Deposition”, Takeshi Kondo, Yutaka Sawada, Hiroshi. Funakubo, Kensuke Akiyama, Takanori Kiguchi, Meihan Wang, and Takayuki Uchida, *Electrochem. Solid-State Lett.*, 12(5) D42-D44 (2009).
- 2) “Fabrication of Conductive Oxide Polycrystalline BaPbO<sub>3</sub> Films by Chemical Solution Deposition and Their Electrical resistivity”, Hiroshi Naganuma, Kayoko Yamada, Hiromi Shima, Kensuke Akiyama, Takashi Iijima, Hiroshi Funakubo and Soichiro Okamura, *J. Electroceram.*, 22, 78-81 (2009).
- 3) “Impact of 90° Domain Wall Motion in Pb(Zr<sub>0.43</sub>Ti<sub>0.57</sub>)O<sub>3</sub> Film on the Ferroelectricity Induced by an Applied Electric Field”, Hitoshi Morioka, Keisuke Saito, Hiroshi Nakaki, Rikyu Ikariyama, Toshiyuki Kurosawa, and Hiroshi Funakubo, *Appl. Phys. Exp.*, 2, 041401-1 -3 (2009).
- 4) “Influence of Pb and La Contents on the Lattice Configuration of La-Substituted Pb(Zr,Ti)O<sub>3</sub> Films Fabricated by CSD Method”, Hiromi Shima, Ken Nishida, Hiroshi Funakubo, Takashi Iijima, Takashi Katoda, Hiroshi Naganuma and Souichiro Okamura, *IEEE Trans. Urtla, Ferro. Freq. Control*, 56(4), 687-692 (2009).

- 5) “Composition Control and Thickness Dependence of {100}-oriented Epitaxial BiCoO<sub>3</sub>-BiFeO<sub>3</sub> Films Grown by Metalorganic Chemical Vapor Deposition”, Shintaro Yasui, Mitsumasa Nakajima, Hiroshi Naganuma, Soichiro Okamura, Ken Nishida, Takashi Yamamoto, Takashi Iijima, Masaki Azuma, Hitoshi Morioka, Keisuke Saito, Mutsuo Ishikawa, Tomoaki Yamada, and Hiroshi Funakubo, *J. Appl. Phys.*, 105, 061620-1-5 (2009).
- 6) “Effect of Bottom Electrode on Dielectric Property of Sputtered-(Ba,Sr)TiO<sub>3</sub> Films”, Shinichi Ito, Tomoaki Yamada, Kenji Takahashi, Shoji Okamoto, Takafumi Kamo, Hiroshi Funakubo, Ivoyl Koutsaroff, Marina Zelner, and Andrew Cervin-Lawry, *J. Appl.Phys.*, 105, 061606-1-4 (2009).
- 7) “Crystal Structure and Electrical Property Comparisons of Epitaxial Pb(Zr, Ti)O<sub>3</sub> Thick Films Grown on (100)CaF<sub>2</sub> and (100)SrTiO<sub>3</sub> Substrates”, Takashi Fujisawa, Hiroshi Nakaki, Rikyu Ikariyama, Tomoaki Yamada, Mutsuo Ishikawa Hiroshi Funakubo, and Hitoshi Morioka, *J. Appl. Phys.*, 105, 061614-1-5 (2009).
- 8) “Growth of Epitaxial Pb(Zr,Ti)O<sub>3</sub> Thick Films on (100)CaF<sub>2</sub> Substrates with Perfect Polar-axis-orientation and Their Electrical and Mechanical Property Characterization” , Takashi Fujisawa, Hiroshi Nakaki, Rikyu Ikariyama, Mitsumasa Nakajima, Tomoaki Yamada, Mutsuo Ishikawa, Hitoshi Morioka, Takashi Iijima and Hiroshi Funakubo, *Mater. Res. Soc. Symp. Proc.*, 1129, 1129-V11-02 (2009).
- 9) “Low Temperature Preparation of (111)-oriented Pb(Zr, Ti)O<sub>3</sub> Films Using Lattice – matched (111)SrRuO<sub>3</sub>/Pt Bottom Electrode by Metal Organic Chemical Vapor Deposition”, Hiroki Kuwabara, Akihiro Sumi, Shoji Okamoto, Hiromasa Hoko, Jeffrey S. Cross, and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 48(4), 04C067-1-4 (2009).
- 10) “Combinatorial Preparation Process of Pb(Zr<sub>1-x</sub>Ti<sub>x</sub>)O<sub>3</sub> Thin Films by Chemical Solution Deposition Methods”, Gang He, Takashi Iijima and Hiroshi Funakubo, *J. Ceram. Soc. Jpn.*, 117(5), 698-702 (2009).
- 11) “The Effect of Precursor Ligands on the Deposition Characteristics of Ru Films by MOCVD”, Kazuhisa Kawano, Hiroaki Kosuge, Noriaki Oshima, and Hiroshi Funakubo, *Electrochem. Solid-State Lett.*, 12(10), D80-D83 (2009).
- 12) “Effect of Film Thickness on Ferroelectric Domain Structure and Properties of Pb(Zr<sub>0.35</sub>Ti<sub>0.65</sub>)O<sub>3</sub>/SrRuO<sub>3</sub>/SrTiO<sub>3</sub> Heterostructures”, Hitoshi Morioka, Keisuke Saito, Shintaro Yokoyama, Takahiro Oikawa, Toshiyuki Kurosawa, and Hiroshi Funakubo, *J. Mater. Sci.*, 44, 5318 – 5324 (2009).
- 13) “Orientation Controlled Deposition of Pb(Zr,Ti)O<sub>3</sub> Films Using Micron-size Patterned SrRuO<sub>3</sub> Buffer Layer”, Ken Nishida, Takashi Yamamoto, Minoru Osada, Osami Sakata, Shigeru Kimura, Keisuke Saito, Masamichi Nishide, Takashi Katoda, Shintaro Yokoyama and Hiroshi Funakubo, *J. Mater. Sci.*, 44, 5339 – 5344 (2009).

- 14) "In situ Observation of the Fatigue-Free Piezoelectric Microcantilever by Two-Dimensional X-ray Diffraction", Hitoshi Morioka, Keisuke Saito, Takeshi Kobayashi, Shintaro Yasui, Toshiyuki Kurosawa, and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 48, 09KA03-1-5 (2009).
- 15) "Electric-Field-Induced Transverse Displacement in Pt/Pb(Zr,Ti)O<sub>3</sub> Film/Pt/Si Structure", Takashi Yamamoto, Mitsutaka Yamamoto, Ken Nishida, Hiroshi Funakubo, Takashi Iijima, Toru Aiso, and Yasuko Ichikawa, *Jpn. J. Appl. Phys.*, 48, 09KA04-1-4(2009).
- 16) "Preparation of (001)-Oriented CaBi<sub>4</sub>Ti<sub>4</sub>O<sub>15</sub> and SrBi<sub>4</sub>Ti<sub>4</sub>O<sub>15</sub> Films Using LaNiO<sub>3</sub> Nucleation Layer on Pt-passivated Si Wafer", Yuki Mizutani, Hiroshi Uchida, Hiroshi Funakubo, and Seiichiro Koda, *Jpn. J. Appl. Phys.*, 48, 09KA10-1-4 (2009).
- 17) "Growth of Epitaxial KNbO<sub>3</sub> Thick Films by Hydrothermal Method and Their Characterization", Mutsuo Ishikawa, Keisuke Yazawa, Takashi Fujisawa, Shintaro Yasui, Tomoaki Yamada, Tomohito Hasegawa, Takeshi Morita, Minoru Kurosawa, and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 48, 09KA14-1-4(2009).
- 18) "Characteristics of Undoped and Mn-Doped BiFeO<sub>3</sub> Films Formed on Pt and SrRuO<sub>3</sub>/Pt Electrodes by Radio-Frequency Sputtering", Jeong Hwan Kim, Hiroshi Funakubo, Yoshihiro Sugiyama, and Hiroshi Ishiwara, *Jpn. J. Appl. Phys.*, 48, 09KB02-1-4(2009).
- 19) "Piezoelectric Properties of {100}-Oriented Epitaxial BiCoO<sub>3</sub>-BiFeO<sub>3</sub> Films Measured Using Synchrotron X-ray Diffraction", Shintaro Yasui, Osami Sakata, Mitsumasa Nakajima, Satoru Utsugi, Keisuke Yazawa, Tomoaki Yamada, and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 48, 09KD06-1-4 (2009).
- 20) "Effects of Substrate Clamping on Electrical Properties of Polycrystalline Piezoelectric Films", Tetsu Miyoshi, Mitsumasa Nakajima, and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 48, 09KD09-1-6 (2009).
- 21) "Electronic and Structural Properties of BiZn<sub>0.5</sub>Ti<sub>0.5</sub>O<sub>3</sub>", Kaoru Miura, Makoto Kubota, Masaki Azuma, and Hiroshi Funakubo, *Jpn. J. Appl. Phys.*, 48, 09KF05-1-4 (2009).
- 22) "Raman Spectroscopy Evaluation of Oxygen Vacancy Migration by Electrical Field in Multilayer Ceramic Capacitors", Ken Nishida, Hiroshi Kishi, Minoru Osada, Hiroshi Funakubo, Masamichi Nishide, Hironari Takeuchi, Takashi Katoda, and Takashi Yamamoto, *Jpn. J. Appl. Phys.*, 48, 09KF11-1-4 (2009).
- 23) "Composition dependency of epitaxial Pb(Zr,Ti)O<sub>3</sub> films with different film thickness", Shintaro Yokoyama, Hiroshi Funakubo, Hitoshi Morioka, Keisuke Saito, Tomoaki Yamada and Mutsuo Ishikawa, *Ferroelectrics*, 389(1), 10 - 17 (2009).
- 24) "Ligand Structure Effect on A Divalent Ruthenium Precursor for MOCVD", Kazuhisa Kawano, Hiroaki Kosuge, Noriaki Oshima, Tadashi Arie, Yutaka Sawada, and Hiroshi Funakubo, Kazuhisa Kawano, Hiroaki Kosuge, Noriaki Oshima, Tadashi Arie, Yutaka Sawada, and Hiroshi Funakubo, *Mater. Res. Soc. Symp. Proc.*, 1155, 1155-C09-11 (2009).

- 25) “Electronic and Structure Properties of BaTiO<sub>3</sub>: Proposal about the Role of Ti 3s and 3p States for Ferroelectricity”, Kaoru Miura, Tatsuo Furuta, Hiroshi Funakubo, Solid State Commun., 150, 205-208 (2009).
- 26) “Investigation of Oxygen vacancies in Micro-patterned PZT Thin Films using Raman spectroscopy”, Ken Nishida, Minoru Osada, Shintaro Yokoyama, Takafumi Kamo, Takashi Fujisawa, Keisuke Saito, Hiroshi Funakubo, Takashi Katoda, and Takashi Yamamoto, Key Eng. Mater., 421-422, 135-138 (2010).
- 27) “Effect of Incubation Time on Deposition Behavior of Ruthenium Films by MOCVD Using (2,4-Dimethylpentadienyl) (ethylcyclopentadienyl) Ruthenium”, Masaki Hirano, Kazuhisa Kawano, and Hiroshi Funakubo, Key Eng. Mater., 421-422, 87-90 (2010).
- 28) “Polarized Raman Study for Epitaxial PZT Thick Film with the Mixture Orientation of (100)/(001)”, Mitsumasa Nakajima, Takashi Fujisawa, Ken Nishida, Takashi Yamamoto, Minoru Osada, Hiroshi Naganuma, Soichiro Okamura, and Hiroshi Funakubo, Key Eng. Mater., 421-422, 99-102 (2010).

### 3. Review

- 1) “Growth of BiFeO<sub>3</sub> Films by MOCVD and Their Property, Hiroshi Funakubo, Shintaro Yasui, Tomoaki Yamada, and Mutsuo Ishikawa, Materials Integration, 22(2) 25-31 (2009).
- 2) “Growth of Pb(Zr,Ti)O<sub>3</sub> Films for Giant Piezoelectric Response, Hiroshi Funakubo, Takashi Fujisawa, Tomoaki Yamada, and Mutsuo Ishikawa, J. Soc. Automotive Eng. Jpn. , 63(4), 94-95 (2009).
- 3) “Growth of Polar-axis-oriented Tetragonal Pb(Zr,Ti)O<sub>3</sub> Thick Films and Their Properties” , Hiroshi Funakubo, Takashi Fujisawa, Satoru Utsugi, Takaki Yamada and Mutsuo Ishikawa, Materials Integration, 22(9), 17-24 (2009).

### 4. Invited presentation in International/domestic conference

International conference

- 1) Hiroshi Funakubo, Takashi Fujisawa, Hiroshi Nakaki, Satoru Utsugi, Rikyu Ikariyama, Hitoshi Morioka, Mutsuo Ishikawa and Tomoaki Yamada , “Growth of Polar-axis-oriented Epitaxial Pb(Zr, Ti)O<sub>3</sub> Thick Films Grown on (100)CaF<sub>2</sub> Substrates” , 8th Pacific Rim Conference on Ceramic and Glass Technology, May 31-June 5, 2009, Hyatt Regency Vancouver, Vancouver, British Columbia, Canada, S27-014-2009. [Invited]
- 2) Hiroshi Funakubo, Takashi Fujisawa, Hiroshi Nakaki, Rikyu Ikariyama, Satoru Utsugi, Hitoshi Morioka, Mutsuo Ishikawa, Takashi Iijima and Tomoaki Yamada, “ Characterization of Polar axis-oriented Epitaxial Pb(Zr, Ti)O<sub>3</sub> Thick Films Grown on (100)CaF<sub>2</sub> Substrates” , 14th US-Japan Seminar on Dielectric and Piezoelectric Materials, Oct 11, 2009, Welches,

Oregon, USA. A1 p.1-3. [Plenary Talk]

- 3) Hiroshi Funakubo, Shitaro Yasui, Takashi Fujisawa, Takafumi Kamo, Mitsumasa Nakajima, Satoru Utsugi, Tomoaki Yamada, and Osami Sakata; , “*In-situ* Observation of Piezoresponse and Electrical Property by Time-Resolved Synchrotron X-Ray Diffraction:” , Materials Science & Technology 2009 Conference & Exhibition, (MS&T 09), Oct. 27th, 2009, Pittsburg, USA, p.85.[Invited]
- 4) Hiroshi Funakubo, Takashi Fujisawa, Hiroshi Nakaki, Satoru Utsugi, Hitoshi Morioka, Takashi Iijima, Osami Sakata, Mutsuo Ishikawa and Tomoaki Yamada, “Characterization of Polar-axis-oriented Epitaxial Pb(Zr, Ti)O<sub>3</sub> Thick Films” , 2009 MRS Fall Meeting, Hynes Convention Center and Sheraton Boston Hotel Boston, MA, U.S.A., Dec. 3, 2009, F10.2.[Invited]

Domestic conference

Total 1

## 5. Others

Patent

Total 10 (Japan, 2009)

Award

Total 2 including

- 1) Hitoshi Morioka、 Best Presentation Awards of Doctor Thesis.
- 2) Mitsumasa Nakajima、 Best Presentation Awards of Master Thesis.