

## REFERENCES

1. B. Jaffe, W. R. Cook, and H. Jaffe, *Piezoelectric Ceramics* (Academic Press, London, 1971), pp. 135-148.
2. G. H. Haertling, "Ferroelectric Ceramics: History and Technology," *J. Am. Ceram. Soc.*, **82**(4), 797-818 (1999).
3. N. Setter and R. Waser, "Electroceramic Materials," *Acta. Mater.*, **48**, 151-178 (2000).
4. S. L. Swartz, "Topics in Electronic Ceramics," *IEEE T. Dielect. El. In.*, **25**(5), 935-987 (1990).
5. V. Y. Topolov and A. V. Turik, "A New Monoclinic Phase and Features of Stress Relief in  $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$  Solid Solutions," *J. Phys: Condens. Mat.*, **13**, L771-L775 (2001).
6. B. Noheda, J. A. Gonzalo, L. E. Cross, R. Guo, S.-E. Park, D. E. Cox, and G. Shirane, "Tetragonal-to-Monoclinic Phase Transition in a Ferroelectric Perovskite: The Structure of  $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ ," *Phys. Rev. B.*, **61**(13), 8687-8695 (2000).
7. T. R. Shrout and J. P. Dougherty, "A World Review on Lead Based  $\text{Pb}(\text{B}_1\text{B}_2)\text{O}_3$  Relaxors vs  $\text{BaTiO}_3$  Dielectrics for Multilayer Capacitors," *J. Am. Ceram. Soc.*, **73**, 3-19 (1990).
8. L. E. Cross, "Review on Ferroelectric Materials for Electromechanical Transducer Applications," *Mater. Chem. Phys.*, **43**, 108-115 (1996).
9. "Ferroelectric," ([www.chemsoc.org/.../ezine/2001/dann\\_dec01.htm](http://www.chemsoc.org/.../ezine/2001/dann_dec01.htm), 2001).

10. L. B. Kong, W. Zhu, and O. K. Tan, "Preparation and Characterization of  $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$  Ceramics from High-Energy Ball Milling Powders," *Mater. Lett.*, **42**, 232-239 (2000).
11. N. J. Ramer, S. P. Lewis, E. J. Mele, and A. M. Rappe, "Stress-Induced Phase Transition in  $\text{Pb}(\text{Zr}_{1/2}\text{Ti}_{1/2})\text{O}_3$ ," *J. Phys. Chem. Solids.*, **61**, 317-332 (2000).
12. R. Guo, L. E. Cross, S.-E. Park, B. Noheda, D. E. Cox, and G. Shirane, "Origin of the High Piezoelectric Response in  $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ ," *Phys. Rev. Lett.*, **84**(23), 5423-5426 (2000).
13. I. Grinberg, V. R. Cooper, and A. M. Rappe, "Relationship between Local Structure and Phase Transitions of a Disordered Solid Solution," *Nature*, **419**, 909-911 (2002).
14. M. V. Raymond and D. M. Smyth, "Defect Chemistry and Transport Properties of  $\text{Pb}(\text{Zr}_{1/2}\text{Ti}_{1/2})\text{O}_3$ ," *Integr. Ferroelectr.*, **4**, 145-154 (1994).
15. S. M. Gupta, A. R. Kulkarni, M. Vedpathak, and S. K. Kulkarni, "Surface Study of Lead Magnesium Niobate Ceramic Using X-Ray Photoelectron Spectroscopy," *Mat. Sci. Eng. B-Solid.*, **39**, 34-40 (1996).
16. A. L. Costa, C. Galassi, G. Fabbri, E. Roncari, and C. Capiani, "Pyrochlore Phase and Microstructure Development in Lead Magnesium Niobate Materials," *J. Eur. Ceram. Soc.*, **21**, 1165-1170 (2001).
17. L. B. Kong, J. Ma, W. Zhu, and O. K. Tan, "Rapid Formation of Lead Magnesium Niobate-Based Ferroelectric Ceramics Via a High-Energy Ball Milling Process," *Mater. Res. Bull.*, **37**, 459-465 (2002).

18. A. A. Cavalheiro, C. R. Foschini, M. A. Zaghete, C. O. Paiva-Samtos, M. Cilense, J. A. Varela, and E. Longo, "Seeding of PMN Powders Made by the Pechini Method," *Ceram. Int.*, **27**, 509-515 (2001).
19. S. L. Swartz and T. R. Shrout, "Fabrication of Perovskite Lead Magnesium Niobate," *Mater. Res. Bull.*, **17**, 1245-1250 (1982).
20. B. Noheda, D. E. Cox, G. Shirane, J. A. Gonzalo, L. E. Cross, and S.-E. Park, "A Monoclinic Ferroelectric Phase in the Pb(Zr<sub>1-x</sub>Ti<sub>x</sub>)O<sub>3</sub> Solid Solution," *Appl. Phys. Lett.*, **74**(14), 2059-2061 (1999).
21. K. Uchino, "Materials Issues in Design and Performance of Piezoelectric Actuators: an Overview," *Acta. Mater.*, **46**, 3745-3753 (1998).
22. Z. He, J. Ma, R. Zhang, and T. Li, "Fabrication and Characterization of Bilayered Pb(Zr,Ti)O<sub>3</sub>-Based Ceramics," *Mater. Lett.*, **56**, 1084-1088 (2002).
23. D. L. Corker, A. M. Glazer, R. W. Whatmore, A. Stallard, and F. Fauth, "A Neutron Diffraction Investigation into the Rhombohedral Phases of the Perovskite Series PbZr<sub>1-x</sub>Ti<sub>x</sub>O<sub>3</sub>," *J. Phys.: Condens. Mat.*, **10**, 6251-6269 (1998).
24. D. Viehland, J.-F. Li, X. Dai, and Z. Xu, "Structural and Property Studies of High Zr-Content Lead Zirconate Titanate," *J. Phys. Chem. Solids.*, **57**(10), 1545-1554 (1996).
25. X. Dai, Z. Xu, and D. Viehland, "Effect of Oxygen Octahedron Rotations on the Phase Stability, Transformational Characteristics, and Polarization Behavior in the Lead Zirconate Titanate Crystalline Solution Series," *J. Am. Ceram. Soc.*, **78**(10), 2815-2827 (1995).

26. Y. Matsuo and H. Sasaki, "Formation of Lead Zirconate Titanate Solid Solutions," *J. Am. Ceram. Soc.*, **48**(6), 289-291 (1965).
27. D. L. Hankey and J. V. Biggers, "Solid State Reactions in the System PbO-TiO<sub>2</sub>-ZrO<sub>2</sub>," *J. Am. Ceram. Soc. C*, **64**, 172-173 (1981).
28. T. Kimura, A. Takenaka, T. Mifune, Y. Hayashi, and T. Yamaguchi, "Preparation of Needle-Like TiZrO<sub>4</sub> and PZT Powders," *J. Mater. Sci.*, **27**, 1479-1483 (1992).
29. S. S. Chandratreya, R. M. Fulrath, and J. A. Pask, "Reaction Mechanisms in the Formation of PZT Solid Solution," *J. Am. Ceram. Soc.*, 422-425 (1981).
30. A. I. Kingon and J. B. Clark, "Sintering of PZT Ceramics: I, Atmosphere Control," *J. Am. Ceram. Soc.*, **66**(4), 253-256 (1983).
31. B. V. Hiremath, A. I. Kingon, and J. V. Biggers, "Reaction Sequence in the Formation of Lead Zirconate-Lead Titanate Solid Solution: Role of Raw Materials," *J. Am. Ceram. Soc.*, **66**(11), 790-793 (1983).
32. O. Babushkin, T. Lindback, J.-C. Luc, and J.-Y. M. Leblais, "Kinetic Aspects of the Formation of Lead Zirconium Titanate," *J. Eur. Ceram. Soc.*, **16**, 1293-1298 (1996).
33. Z. Brankovic, G. Brankovic, and J. A. Varela, "PZT Ceramics Obtained from Mechanochemically Synthesized Powders," *J. Mater. Sci.*, **14**, 37-41 (2003).
34. B. Noheda, "Structure and High-Piezoelectricity in Lead Oxide Solid Solutions," *Solid State Sci.*, **6**, 27-34 (2002).
35. G. Shirane, K. Suzuki, and A. Takeda, "Crystal Structure of Pb(Zr,Ti)O<sub>3</sub>," *J. Phys. Soc. Jpn*, **7**(1), 333-336 (1952).

36. K. H. Hardtl and H. Rau, "PbO Vapour Pressure in the Pb(Ti<sub>1-x</sub>Zr<sub>x</sub>)O<sub>3</sub> System," *Solid State Commun.*, **7**(1), 41-45 (1969).
37. A. Garg and D. C. Agrawal, "Effect of Net PbO Content on Mechanical and Electromechanical Properties of Lead Zirconate Titanate Ceramics," *Mat. Sci. Eng. B-Solid.*, **56**, 46-50 (1999).
38. W. D. Kingery, H. K. Bowen, and D. R. Uhlmann, *Introduction to Ceramics*, 2 ed. (John Wiley & Sons, London, 1976), pp. 448-513.
39. E. K. Goo, R. K. Mishra, and G. Thomas, "Transmission Electron Microscopy of Pb(Zr<sub>0.52</sub>Ti<sub>0.48</sub>)O<sub>3</sub>," *J. Am. Ceram. Soc.*, **64**(6), 517-519 (1981).
40. G. S. Snow, "Elimination of Porosity in Pb(Zr,Ti)O<sub>3</sub> Ceramics by Liquid-Phase Sintering," *J. Am. Ceram. Soc.*, **57**(6), 272 (1974).
41. K. Kakegawa, J. Mohri, T. Takahashi, H. Yamamura, and S. Shirasaki, "A Compositional Fluctuation and Properties of Pb(Zr,Ti)O<sub>3</sub>," *Solid State Commun.*, **24**, 769-772 (1977).
42. B. M. Jin, J. Kim, and S. C. Kim, "Effects of Grain Size on the Electrical Properties of PbZr<sub>0.52</sub>Ti<sub>0.48</sub>O<sub>3</sub> Ceramics," *Appl. Phys. A-Mater.*, **65**, 53-56 (1997).
43. Z. Surowiak, M. F. Kupriyanov, and D. Czekaj, "Properties of Nanocrystalline Ferroelectric PZT Ceramics," *J. Eur. Ceram. Soc.*, **21**, 1377-1381 (2001).
44. W. Cao and C. A. Randall, "Grain Size and Domain Size Relations in Bulk Ceramic Ferroelectric Materials," *J. Phys. Chem. Solids.*, **57**(10), 1499-1505 (1996).

45. J. T. Reszat, A. E. Glazounov, and M. J. Hoffmann, "Analysis of Intrinsic Lattice Deformation in PZT-Ceramics of Different Compositions," *J. Eur. Ceram. Soc.*, **21**, 1349-1352 (2001).
46. B. K. Kim, "Probing of Nanoscaled Nonstoichiometric 1:1 Ordering in  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ -Based Relaxor Ferroelectrics by Raman Spectroscopy," *Mat. Sci. Eng. B-Solid.*, **94**, 102-105 (2002).
47. A. Mergen and W. E. Lee, "Fabrication, Characterisation and Formation Mechanism of  $\text{Pb}_{1.83}\text{Mg}_{0.29}\text{Nb}_{1.71}\text{O}_{6.39}$  Pyrochlore," *J. Eur. Ceram. Soc.*, **17**, 1033-1047 (1997).
48. B. P. Burton and E. Cockayne, "Why  $\text{Pb}(\text{B},\text{B}')\text{O}_3$  Perovskites Disorder at Lower Temperatures Than  $\text{Ba}(\text{B},\text{B}')\text{O}_3$  Perovskites," *Phys. Rev. B*, **60**(18), 12542-12545 (1999).
49. V. S. Vikhnin, R. Blinc, R. Pirc, S. E. Kapphan, I. L. Kuslova, and P. A. Markovin, "A Model of Polar Clusters in Ferroelectric Relaxors of PMN-Type: Polaronic and Charge Transfer Effects," *Ferroelectrics*, **268**, 257-262 (2002).
50. N. W. Thomas, S. A. Ivanov, S. Ananta, R. Tellgren, and H. Rundlof, "New Evidence for Rhombohedral Symmetry in the Relaxor Ferroelectric  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ ," *J. Eur. Ceram. Soc.*, **19**, 2667-2675 (1999).
51. J. Chen, M. P. Harmer, and D. M. Smyth, "Compositional Control of Ferroelectric Fatigue in Perovskite Ferroelectric Ceramics and Thin Films," *J. Appl. Phys.*, **76**, 5394 (1994).
52. L. A. Bursill, H. Qian, J.-L. Peng, and X. D. Fan, "Observation and Analysis of Nanodomain Textures in the Dielectric Relaxor Lead Magnesium Niobate," *Physica. B*, **216**, 1-23 (1995).

53. R. Blinc, J. Dolinsek, A. Gregorovic, B. Zalar, C. Filipic, Z. Kutnjak, A. Levstik, and R. Pirc, "Local Polarization Distribution and Edwards-Anderson Order Parameter of Relaxor Ferroelectrics," *Phys. Rev. Lett.*, **83**(2), 424-427 (1999).
54. A. Levstik, Z. Kutnjak, C. Filipic, and R. Pirc, "Glassy Freezing in Relaxor Ferroelectric Lead Magnesium Niobate," *Phys. Rev. B*, **57**(18), 11204-11211 (1998).
55. M. Inada, "Analysis of the Formation Process of the Piezoelectric PCM ceramics," *Jpn. Natl. Tech. Rept.*, **27**, 95-102 (1977).
56. N. Wakiya, K. Shinozaki, and N. Mizutani, "Estimation of Phase Stability in  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$  and  $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$  Using the Bond Valence Approach," *J. Am. Ceram. Soc.*, **80**, 3217 (1998).
57. Lucas and W. T. Petuskey, "Phase Equilibria in the Lead-Magnesium-Niobium- Oxygen System at 1000°C," *J. Am. Ceram. Soc.*, **84**(9), 2150-2152 (2001).
58. M. Lejeune and J. P. Boilot, "Low Firing Dielectric Based on Lead Magnesium Niobate," *Mater. Res. Bull.*, **20**(5), 493-499 (1985).
59. E. R. Camargo, M. Kakihana, E. Longo, and E. R. Leite, "Pyrochlore-Free  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$  Prepared by a Combination of the Partial Oxalate and the Polymerized Complex Methods," *J. Alloy. Compd.*, **314**, 140-146 (2001).
60. E. R. Camargo, E. Longo, and E. R. Leite, "Synthesis of Ultra-Fine Columbite Powder  $\text{MgNb}_2\text{O}_6$  by the Polymerized Complex Method," *J. Sol-Gel. Sci. Techn.*, **17**, 111-121 (2000).

61. M. Lejeune and J. P. Boilot, "Influence of Ceramic Processing on Dielectric Properties of Perovskite Type Compound:  $\text{Pb}(\text{Mg}_{1/3}, \text{Nb}_{2/3})\text{O}_3$ ," *Ceram. Int.*, **9**(4), 119-122 (1983).
62. C.-H. Lu and H.-S. Yang, "Development of Fast Formation Process for Synthesizing Submicron Lead Magnesium Niobate Perovskite Powder," *Mater. Sci. Eng. B-Solid*, **84**, 159-162 (2001).
63. D. Saha, A. Sen, and H. S. Maiti, "Low Temperature Liquid Phase Sintering of Lead Magnesium Niobate," *Ceram. Int.*, **25**, 145-151 (1999).
64. S. L. Swartz, T. R. Shrout, W. A. Schulze, and L. E. Cross, "Dielectric Properties of Lead Magnesium Niobate Ceramics," *J. Am. Ceram. Soc.*, **67**(5), 311-315 (1984).
65. W.-F. A. Su, "Effects of Additives on Perovskite Formation in Sol-Gel Derived Lead Magnesium Niobate," *Mater. Chem. Phys.*, **62**, 18-22 (2000).
66. D. M. Wan, J. Wang, S. C. Ng, and L. M. Gan, "Formation and Characterization of Lead Magnesium Niobate Synthesized from the Molten Salt of Potassium Chlorate," *J. Alloy. Compd.*, **274**, 110-117 (1998).
67. P. Bonneaua, P. Garniera, E. Husson, and A. Morell, "Structure Study of PMN Ceramics by X-Ray Diffraction Between 297 and 1023 K," *Mater. Res. Bull.*, **24**(2), 201-206 (1989).
68. P. Bonneau, P. Garnier, G. Calvarin, E. Husson, J. R. Gavarri, A. W. Hewat, and A. Morell, "X-Ray and Neutron Diffraction Studies of the Diffuse Phase Transition in  $\text{PbMg}_{1/3}\text{Nb}_{2/3}\text{O}_3$  Ceramics," *J. Solid State Chem.*, **91**(2), 350-361 (1991).

69. E. Husson, M. Chubb, and A. Morell, "Superstructure in  $\text{PbMg}_{1/3}\text{Nb}_{2/3}\text{O}_3$  Ceramics Revealed by High Resolution Electron Microscopy," *Mater. Res. Bull.*, **23**(3), 357-361 (1988).
70. N. D. Mathan, E. Husson, P. Gaucher, and A. Morell, "Structural Study of a Poled  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$  Ceramic at Low Temperature," *Mater. Res. Bull.*, **26**(11), 1167-1172 (1990).
71. Q. M. Zhang, H. You, M. L. Mulvihill, and S. J. Jang, "An X-Ray Diffraction Study of Superlattice Ordering in Lead Magnesium Niobate," *Solid. State. Commun.*, **97**(8), 693-698 (1996).
72. K. H. Yoon, B. D. Lee, J. Park, and J. H. Park, "Dielectric and Piezoelectric Properties of  $(x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-(1-x)\text{Pb}(\text{Zr}_{1/2}\text{Ti}_{1/2})\text{O}_3$  Thin Films Prepared by the Sol-Gel Method," *J. Appl. Phys.*, **90**(4), 1968-1972 (2001).
73. V. Koval, C. Alemany, J. Briancin, and H. Brunckova, "Dielectric Properties and Phase Transition Behavior of  $x\text{PMN}-(1-x)\text{PZT}$  Ceramics Systems," *J. Electroceram.*, **10**, 19-29 (2003).
74. N. S. Almodovar, R. Font, J. Portelles, O. Raymond, and J. M. Siqueiros, "Phase Formation and Characterization of  $[\text{Fe},\text{Mg}]\text{NbO}_4$  as a New Precursor for the PMN-PFN System," *J. Mater. Sci.*, **37**, 5089-5093 (2002).
75. S. G. Jun and N. K. Kim, "Dielectric Properties of PFW-PMN Relaxor System Prepared by B-site Precursor Method," *J. Mater. Sci.*, **35**, 2093-2097 (2000).
76. H. Ouchi, M. Nishida, and S. Hayakawa, "Piezoelectric Properties of  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-\text{PbTiO}_3-\text{PbZrO}_3$  Ceramics Modified with Certain Additives," *J. Am. Ceram. Soc.*, **49**(11), 577-582 (1966).

77. J.-C. Shaw, K.-S. Liu, and I.-N. Lin, "Dielectric Behavior at Morphotropic Phase Boundary for PMN-PZT Ceramics," *Scripta. Mater.*, **29**, 981-986 (1993).
78. K. Kakegawa, T. Wakabayashi, and Y. Sasaki, "Synthesis of a Solid Solution of  $\text{PbTiO}_3\text{-PbZrO}_3\text{-Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$  System Through Use of 8-Quinolinol and Its Properties," *J. Mater. Sci.*, **27**, 1291-1296 (1992).
79. M. Villegas, J. R. Jurado, C. Moure, and P. Duran, "Perovskite Phase Formation in the  $\text{PbMg}_{1/3}\text{Nb}_{2/3}\text{O}_3\text{-PbZrO}_3\text{-PbTiO}_3$  System by the Columbite Route," *J. Mater. Sci.*, **30**, 1391-1396 (1995).
80. P. E. J. Flewitt and R. K. Wild, *Microstructural Characterisation of Materials and Alloys* (Institute of Metals, London, 1985).
81. P. Bordet, A. Mchale, A. Santoro, and R. S. Roth, "Powder Neutron Diffraction Study of  $\text{ZrTiO}_4\text{-Zr}_5\text{Ti}_7\text{O}_{24}$  and  $\text{FeNb}_2\text{O}_6$ ," *J. Solid State Chem.*, **64**, 30-46 (1986).
82. A. Yamamoto, T. Yamada, H. Ikawa, O. Fukunaga, K. Tanaka, and F. Marumo, "Modulated Structure of Zirconium Titanate," *Acta. Crystallogr. C*, **47**, 1588-1591 (1991).
83. R. Ruh, G. W. Hollenberg, E. G. Chavles, and V. A. Pale, "Phase Relations and Thermal Expansion in the System  $\text{HfO}_2\text{-TiO}_2$ ," *J. Am. Ceram. Soc.*, **59**, 495-499 (1976).
84. A. Bianco, M. Paci, and R. Freer, "Zirconium Titanate: from Polymeric Precursors to Bulk Ceramics," *J. Eur. Ceram. Soc.*, **18**, 1235-1243 (1998).
85. F. Khairulla and P. P. Phule, "Chemical Synthesis and Structural Evolution of Zirconium Titanate," *Mat. Sci. Eng. B-Solid.*, **12**, 327-336 (1992).

86. *Powder diffraction File no.34-415* (International Centre for Diffraction Data, Newton Square, PA, 2000).
87. R. E. Newnham, "Crystal Structure of ZrTiO<sub>4</sub>," *J. Am. Ceram. Soc.*, **50**, 216 (1967).
88. *Powder diffraction File no.37-1484* (International Centre for Diffraction Data, Newton Square, PA, 2000).
89. *Powder diffraction File no.17-923* (International Centre for Diffraction Data, Newton Square, PA, 2000).
90. *Powder diffraction File no.21-1272* (International Centre for Diffraction Data, Newton Square, PA, 2000).
91. *Powder diffraction File no.21-1276* (International Centre for Diffraction Data, Newton Square, PA, 2000).
92. P. J. Sanchez-Soto, M. A. Ariles, G. Colon, M. Macias, and J. A. Navio, "Thermal Evolution of TiO<sub>2</sub>-ZrO<sub>2</sub> Composites Prepared by Chemical Coating Processing," *Mater. Lett.*, **20**, 339 (1994).
93. S.-L. Yang and J. M. Wu, "ZrO<sub>2</sub>-TiO<sub>2</sub> Ceramics Humidity Sensors," *J. Mater. Sci.*, **26**, 631-636 (1991).
94. C. L. Wang, H. Y. Lee, and F. Azough, "The Microstructure and Microwave Dielectric Properties of Zirconium Titanate Ceramics in the Solid Solution System ZrTiO<sub>4</sub>-Zr<sub>5</sub>Ti<sub>7</sub>O<sub>24</sub>," *J. Mater. Sci.*, **32**, 1693-1701 (1997).
95. F. Azough, R. Freer, and J. Petzelt, "A Raman Spectral Characterization of Ceramics in the System ZrO<sub>2</sub>-TiO<sub>2</sub>," *J. Mater. Sci.*, **28**, 2273-2276 (1993).

96. E. R. Leite, M. Cerqueira, L. A. Perazoli, R. S. Nasar, E. Longo, and J. A. Varela, "Mechanism of Phase Formation in  $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$  Synthesized by a Partial Oxalate Method," *J. Am. Ceram. Soc.*, **79**, 1563-1568 (1996).
97. *Powder diffraction File no.50-346* (International Centre for Diffraction Data, Newton Square, PA, 2000).
98. *Powder diffraction File no.73-2022* (International Centre for Diffraction Data, Newton Square, PA, 2000).
99. J. C. Fernandes, D. A. Hall, M. R. Cockburn, and G. N. Greaves, "Phase Coexistence in PZT Ceramic Powders," *Nucl. Instrum. Meth. B*, **97**, 137-141 (1995).
100. N. Chakrabarti and H. S. Maiti, "Chemical Synthesis of PZT Powder by Auto-Combustion of Citrate-Nitrate Gel," *Mater. Lett.*, **30**, 169-173 (1997).
101. H. C. Wang and W. A. Schulze, "The Role of Excess Magnesium Oxide or Lead Oxide in Determining the Microstructure and Properties of Lead Magnesium Niobate," *J. Am. Ceram. Soc.*, **73**(4), 825-832 (1990).
102. H. Chazono and H. Kishi, "Sintering Characteristic in the  $\text{BaTiO}_3\text{-Nb}_2\text{O}_5\text{-Co}_3\text{O}_4$  Ternary System: II, Stability of so-called "Core-Shell" Structure," *J. Am. Ceram. Soc.*, **83**, 101-106 (2000).
103. S. Fushimi and T. Ikeda, "Phase Equilibrium of the System  $\text{PbO}\text{-TiO}_2\text{-ZrO}_2$ ," *J. Am. Ceram. Soc.*, **50**(3), 129-132 (1967).
104. C. H. Wang, S. J. Chang, and P. C. Chang, "Effect of Sintering Conditions on Characteristics of  $\text{PbTiO}_3\text{-PbZrO}_3\text{-Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$ ," *Mat. Sci. Eng. B-Solid.*, **111**, 124-130 (2004).

105. Y.-D. Hou, M.-K. Zhu, H. Wang, B. Wang, C.-S. Tian, and H. Yan, "Effect of Atmospheric Powder on Microstructure and Piezoelectric Properties of PMZN-PZT Quaternary Ceramics," *J. Eur. Ceram. Soc.*, **24**, 3731-3737 (2004).
106. A. Megriche and M. Troccaz, "Effect of Excess PbO Addition on the Properties of Ferroelectric Doped PZT Ceramics," *Mater. Res. Bull.*, **33**(4), 569-574 (1998).
107. E. R. Nielsen, E. Ringgaard, and M. Kosec, "Liquid-Phase Sintering of  $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$  Using  $\text{PbO}-\text{WO}_3$  Additive," *J. Eur. Ceram. Soc.*, **22**, 1847-1855 (2002).
108. A. Boutarfaia, "Investigations of Co-Existence Region in Lead Zirconate-Titanate Solid Solutions: X-Ray Diffraction Studies," *Ceram. Int.*, **26**, 583-587 (2000).
109. M. A. Akbas, M. A. McCoy, and W. E. Lee, "Microstructural Evolution During Pressureless Sintering of Lead Lanthanum Zirconate Titanate Ceramics with Excess Lead (II) Oxide," *J. Am. Ceram. Soc.*, **78**(9), 2417-2424 (1995).
110. S. Ananta, R. Tipakontitkul, and T. Tunkasiri, "Synthesis, Formation and Characterization of Zirconium Titanate (ZT) Powders," *Mater. Lett.*, **57**, 2637-2642 (2003).
111. R. Tipakontitkul and S. Ananta, "A Modified Two-Stage Mixed Oxide Synthetic Route to Lead Zirconate Titanate Powders," *Mater. Lett.*, **58**, 449-454 (2004).
112. L. B. Kong and J. Ma, "PZT Ceramics Formed Directly from Oxide Via Reaction Sintering," *Mater. Lett.*, **51**, 95-100 (2001).

113. X. X. Wang, K. Murakami, O. Sugiyama, and S. Kaneko, "Piezoelectric Properties, Densification Behavior and Microstructure Evolution of Low Temperature Sintered PZT Ceramics with Sintering Aids," *J. Eur. Ceram. Soc.*, **21**, 1367-1370 (2001).
114. K. Carl and K. H. Hardtl, "Direct Determination of the Coexistence Region in the Solid Solutions  $Pb(Zr_xTi_{1-x})O_3$ ," *Phys. Status Solidi A*, **8**, 87 (1971).
115. *Powder diffraction File no.45-946* (International Centre for Diffraction Data, Newton Square, PA, 2000).
116. *Powder diffraction File no.27-1312* (International Centre for Diffraction Data, Newton Square, PA, 2000).
117. *Powder diffraction File no.33-875* (International Centre for Diffraction Data, Newton Square, PA, 2000).
118. S. Ananta, "Phase and Morphology Evolution of Magnesium Niobate Powders Synthesized by Solid-State Reaction," *Mater. Lett.*, **58**, 2781-2786 (2004).
119. L. B. Kong, J. Ma, H. Huang, and R. F. Zhang, "Crystallization of Magnesium Niobate from Mechanochemically Derived Amorphous Phase," *J. Alloy. Compd.*, **340**, L1-L4 (2002).
120. N.-K. Kim, "Synthesis Chemistry of  $MgNb_2O_6$  and  $Pb(Mg_{1/3}Nb_{2/3})O_3$ ," *Mater. Lett.*, **32**, 127-130 (1997).
121. S. M. Gupta, D. S. Harendranath, and A. R. Kulkarni, "Synthesis and Characterisation of Lead Magnesium Niobate Having Exceptionally High Dielectric Constant," *Ceram. Int.*, **23**, 191-196 (1997).

122. B.-H. Kim, O. Sakurai, N. Wakiya, and N. Mizutani, "Effect of Atmosphere on Stability of  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$  (PMN) Ceramics," *Mater. Res. Bull.*, **32**(4), 451-459 (1997).
123. J. Wang, X. Junmin, W. Dongmei, and N. Weibeng, "Mechanochemical Fabrication of Single Phase PMN of Perovskite Structure," *Solid State Ionics*, **124**, 271-279 (1999).
124. Y. Yu, C. Feng, C. Li, Y. Yang, W. Yao, and H. Yan, "Formation of Columbite-type Precursors in the Mixture of  $\text{MgO}$ - $\text{Fe}_2\text{O}_3$ - $\text{Nb}_2\text{O}_5$  and the Effects on Fabrication of Perovskites," *Mater. Lett.*, **51**, 490-499 (2001).
125. X. Junmin, J. Wang, N. Weibeng, and W. Dongmei, "Activation-Induced Pyrochlore-to-Perovskite Conversion for a Lead Magnesium Niobate Precursor," *J. Am. Ceram. Soc.*, **82**(8), 2282-2284 (1999).
126. S. Ananta and N. W. Thomas, "Relationships between Sintering Conditions, Microstructure and Dielectric Properties of Lead Magnesium Niobate," *J. Eur. Ceram. Soc.*, **19**, 629 (1999).
127. *Powder diffraction File no.38-1477* (International Centre for Diffraction Data, Newton Square, PA, 2000).
128. *Powder diffraction File no.27-1199* (International Centre for Diffraction Data, Newton Square, PA, 2000).
129. L. B. Kong, J. Ma, W. Zhu, and O. K. Tan, "Preparation of PMN Powders and Ceramics Via a High-Energy Ball Milling Process," *J. Mater. Sci. Lett.*, **20**, 1241-1243 (2001).

130. K. Sreedhar and A. Mitra, "Formation of Lead Magnesium Niobate Perovskite from  $MgNb_2O_6$  and  $Pb_3Nb_2O_8$  Precursors," *Mater. Res. Bull.*, **32**(12), 1643-1649 (1997).
131. J. P. Guha, "Reaction Chemistry and Subsolidus Phase Equilibria in Lead-Based Relaxor Systems: Part I, Formation and Stability of the Perovskite and Pyrochlore Compounds in the System  $PbO$ - $MgO$ - $Nb_2O_5$ ," *J. Mater. Sci.*, **34**, 4985-4994 (1999).
132. *Powder diffraction File no.81-861* (International Centre for Diffraction Data, Newton Square, PA, 2000).
133. *Powder diffraction File no.33-769* (International Centre for Diffraction Data, Newton Square, PA, 2000).
134. L. Wu and Y.-C. Liou, "The Effect of Heating Rate on the Properties of PMN Relaxor Ceramics," *Ceram. Int.*, **21**, 335-338 (1995).
135. T. J. Yang and U. Mohideen, "Nanoscale Measurement of Ferroelectric Domain Wall Strain and Energy by Near-Field Scanning Optical Microscopy," *Phys. Letts. A*, **250**, 205-210 (1998).
136. P. K. Davies and M. A. Akbas, "Chemical Order in PMN-Related Relaxors: Structure, Stability Modification, and Impact on Properties," *J. Phys. Chem Solids.*, **61**, 159-166 (2000).
137. A. J. Moulson and J. M. Herbert, *Electrocermics*, 2 ed. (John Wiley and Sons, Chichester, 2003).
138. G. H. Hertling, "Hot Pressed Lead Zirconate-Lead Titanate Ceramics Containing Bismuth," *Am. Ceram. Soc. Bull.*, **43**(12), 875-879 (1964).

139. Y. C. Liou and J. H. Chen, "PMN Ceramics Produced by a Simplified Columbite Route," *Ceram. Int.*, **30**, 17-22 (2004).
140. R. Yimnirun, S. Ananta, E. Meechoowas, and S. Wongsaenmai, "Effects of Uniaxial Stress on Dielectric Properties of Lead Magnesium Niobate-Lead Zirconate Titanate Ceramics," *J. Phys. D: Appl. Phys.*, **36**, 1615-1619 (2003).
141. C. Lei, K. Chen, and X. Zhang, "Structure and Dielectric Relaxation Behavior Near the MPB for  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $\text{Pb}(\text{Ni}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $\text{PbTiO}_3$ ," *Mat. Sci. Eng. B-Solid.*, **111**, 107-112 (2004).
142. A. Boutarfaia and S. E. Bouaoud, "Tetragonal and Rhombohedral Phase Co-existence in the System:  $\text{PbZrO}_3$ - $\text{PbTiO}_3$ - $\text{Pb}(\text{Fe}_{1/5},\text{Ni}_{1/5},\text{Sb}_{3/5})\text{O}_3$ ," *Ceram. Int.*, **22**, 281-286 (1996).
143. *Powder diffraction File no.50-376* (International Centre for Diffraction Data, Newton Square, PA, 2000).
144. C. H. Wang, "The Microstructure and Characteristics of 0.875PZT-0.125PMN Ceramics with Addition of Pb-Based Flux," *J. Eur. Ceram. Soc.*, **22**, 2033-2038 (2002).
145. K. P. Chen, C. Li, X. Zhang, and Y. Huang, "Microstructure and Electrical Properties of  $0.9\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $0.3\text{PbTiO}_3$  Ceramics by Spark Plasma Sintering," *Mat. Letts.*, **57**, 20-23 (2002).
146. C.-H. Lu and D.-P. Chang, "Simultaneous Synthesis and Sintering of Lead Magnesium Niobate Ferroelectric Ceramics," *Mat. Sci. Eng. B-Solid.*, **64**, 195-198 (1999).

147. J. H. Chen and Y.-C. Liou, "0.9Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-0.1PbTiO<sub>3</sub> Relaxor Ferroelectric Ceramics Produced by a Simplified Columbite Route and a Reaction-Sintering Process," *Ceram. Int.*, **30**, 157-162 (2004).
148. T. Y. Ling, X. Junmin, and J. Wang, "Stabilization of Perovskite Phase and Dielectric Properties of 0.95PZN-0.05BT Derived from Mechanical Activation," *J. Alloy. Compd.*, **297**, 92-98 (2000).
149. D. L. Corker, R. W. Whatmore, E. Ringgaard, and W. W. Wolny, "Liquid-Phase Sintering of PZT Ceramics," *J. Eur. Ceram. Soc.*, **20**, 2039-2045 (2000).
150. A. Watanabe, H. Haneda, Y. Moriyoshi, S. Shirasaki, S. Kuramoto, and H. Yamamura, "Preparation of Lead Magnesium Niobate by a Coprecipitation Method," *J. Mater. Sci.*, **27**, 1245-1249 (1992).
151. S. M. Gupta, "Dielectric and Microstructure Studies of Lead Magnesium Niobate Prepared by Partial Oxalate Route," *J. Eur. Ceram. Soc.*, **16**, 473-480 (1996).
152. L. B. Kong, J. Ma, and H. Huang, "Low Temperature Formation of Yttrium Aluminum Garnet from Oxides Via a High-Energy Ball Milling Process," *Mater. Lett.*, **56**, 344-348 (2002).
153. V. Koval, C. Alemany, J. Briancin, H. Brunckova, and K. Saksl, "Effect of PMN Modification on Structure and Electrical Response of  $x$ PMN-(1- $x$ )PZT Ceramic Systems," *J. Eur. Ceram. Soc.*, **23**, 1157-1166 (2003).
154. K. H. Yoon and H. R. Lee, "Effect of Ba<sup>2+</sup> Substitution on Dielectric and Electric-Field-Induced Strain Properties of PMN-PZ-PT Ceramics," *J. Am. Ceram. Soc.*, **83**(11), 2693-2698 (2000).

155. O. Noblanc, P. Gaucher, and G. Calvarin, "Structural and Dielectric Studies of  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $\text{PbTiO}_3$  Ferroelectric Solid Solutions Around the Morphotropic Boundary," *J. Appl. Phys.*, **79**(8), 4291-4297 (1996).
156. D. M. Wan, J. M. Xue, and J. Wang, "Nanocrystalline 0.54PZN-0.36PMN-0.1PT of Perovskite Structure by Mechanical Activation," *Mat. Sci. Eng. A-Struct.*, **286**, 96-100 (2000).
157. Z.-G. Ye and M. Dong, "Morphotropic Domain Structures and Phase Transitions in Relaxor-Based Piezo-Ferroelectric  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $x\text{PbTiO}_3$  Single Crystals," *J. Appl. Phys.*, 2312-1319 (2000).
158. G. Singh, V. S. Tiwari, and V. K. Wadhawan, "Crossover from Relaxor to Normal Ferroelectric Behaviour in  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $x\text{PbZrO}_3$  Ceramic Near  $x = 0.5$ ," *Solid State Commun.*, **118**, 407-411 (2001).
159. H.-T. Lin, D. C. V. Aken, and W. Huebner, "Modeling the Dielectric Response and Relaxation Spectra of Relaxor Ferroelectrics," *J. Am. Ceram. Soc.*, **82**(10), 2698-2704 (1999).