

REFERENCE

- Abdelkefi, H., Khemakhem, H., V'elu, G., Carru, J.C., and Von der M'uhll, R.
(2005) Dielectric properties and ferroelectric phase transitions in Ba_xSr_{1-x} TiO_3 solid solution, Journal of Alloys and Compounds, 399, 1–6.
- Adikarya, S.U., Chana, H.L.W., Choya, C.L., Sundaravelb, B., and Wilsonb, I.H.
(2002) Characterisation of proton irradiated $Ba_{0.65}Sr_{0.35}TiO_3/P(VDF-TrFE)$ ceramic–polymer composites, Journal of Composites Science and Technology, 62, 2161–2167.
- Barbosa, R., Mendes, J A., Sencadas, V., Mano, J. F. and Lanceros-mendez, S.
(2003) Chain Reorientation in β -PVDF Films Upon Transverse Mechanical Deformation Studied by SEM and Dielectric Relaxation. Journal of Ferroelectrics, 294: 73–83,
- Bar-Cohen, Y., Xue, T. and Lih, S.S. (1996) Polymer piezoelectric transducers for ultrasonic, The e-Journal of Nondestructive Testing Vol.1 No.09
- Bauer, D.R. and Mulhaupt, R.G. (1999) Separate contributions to the pyroelectricity in polyvinylidene fluoride from the amorphous and crystalline phases, as well as from their interface, Journal of Applied Physics, 85(6), 3282-3288.
- Birlikseven, C., Altintas, and E., Dursoy, H.Z. (2001) A low temperature pyroelectric study of PVDF thick films, Journal of Materials in Electronics, 12, 601-603.
- Lu, Q., Chen, D., Jiao, X. (2004) Preparation and characterization of $Ba_{1-x}Sr_xTiO_3$ ($x=0.1, 0.2$) fibers by sol–gel process using catechol-complexed titanium isopropoxide, Journal of Alloys and Compounds, 358, pp 76–81.
- Campos, J.S.C., Ribeiro, A.A., Cardoso, C.X. (2007) Preparation of characterization of PVDF/ $CaCO_3$ composites. Journal of Materials Science& Engineering B, 136, pp 123-128
- Cho, S.D., Lee, J.Y., Hyun, J.G., and Paik, K.W. (2004). Study on epoxy/ $BaTiO_3$ composite embedded capacitor films (ECFs) for organic substrate applications. Materials science & engineering B, 110, 233-239.

- Das-Gupta, D.K. (1994) *Ferroelectric Polymers and Ceramic Polymer Composites*, Trans Tech .Publ, Zurich.
- Dang, Z.M., Fan, L.Z., Shen, Y., and Nan, C.W. (2003) Study on dielectric behavior of a three-phase CF/(PVDF + BaTiO₃) Composite, Journal of Chemical Physics Letters, 369, 95–100.
- Dang, Z.M., Fan, L.Z., Shen, Y., and Nan, C.W. (2002) Dielectric behavior of novel three-phase MWNTs/BaTiO₃/PVDF composites, Materials Science and Engineering B, 103, 140-144.
- Garcia, D., Guo, R., and Bhalla, A.S. (2000) Growth and properties of (Ba,Sr)TiO₃ single crystal fibers. Journal of Materials Letters, 42, 136–141.
- Giridharan, N.V., Varatharajan, R. Jayavel, and R. Ramasamy, P. (2000) Fabrication and characterisation of (Ba,Sr)TiO₃ thin films by sol–gel technique through organic precursor route. Journal of Materials Chemistry and Physics, 65, 261–265.
- Gimenes, R., Zaghete, M.A., Bertolini, M., Varela, J.A., Coelho, L.O., and Silva Jr., N.F. (2004) Composites PVDF-TrFE/BT used as bioactive membranes for enhancing bone regeneration, Journal of Der Spiegel, 5385, 539-547.
- Harrison, J.S., Ounaies, Z. (2001) Piezoelectric Polymers. Virginia: National Aeronautics and Space Administration
- Hilczer, B., Lek, J.K., Lomska, M.P., Glinchuk, M. D., Ragulya, A. V. and Pietraszko, A. (2005) Dielectric and pyroelectric response of BaTiO₃-PVDF nanocomposites, Journal of Ferroelectrics, 316, 31–41.
- Hornebecq, V., Huber, C., Maglione, M., Antonietti, M., and Elissalde, C. (2004). Dielectric properties of pure (Ba,Sr)TiO₃ and composites with different grain sizes ranging from the nanometer to the micrometer. Journal of advanced functional materials, 14(9), 899-904.
- Ioachim, A., Ramer, R., Toacsan, M.I., Banciu, M.G., Nedelcu, L., Dutu, C.A., Vasiliu, F., Alexandru, H.V., Berbecaru, C., Stoica, G., Nita, P. (2006) Ferroelectric ceramics based on the BaO-SrO-TiO₂ ternary system for the

- microwave applications, Journal of the European Ceramic Society, 27, pp 1177-1180
- Jiang, Y.Y., Wu, Y., Zeng, Z., Yang, H., and Li, Y.W. (2005)
Characterization and ferroelectric properties of electric poled PVDF films,
12th International Symposium on Electrets, 132- 135.
- Judovits, L. (2006). Thermal analysis of poly(vinylidene fluoride) film,
Journal of Thermochemica Acta, 442, 92–94.
- Kim, B. S., Lee, J.Y. and Porter, R.S.(1998) The Crystalline phase Transformation of
Poly(Vinylidene Fluoride)/Poly(vinyl Fluoride) Blend Films, Polymer
Engineering And Science, 38(9), pp 1359-1365.
- Lang, S.B. and Das-Gupta, D. K. (2000) Pyroelectricity, Fundamentals and
Application, Ferroelectrics Review, 2, 217-354.
- Lu, Q., Chen, D., and Jiao, X. (2003) Preparation and characterization of Ba
Sr TiO ($x=0.1, 0.2$) fibers by sol–gel process using catechol-complexed
titanium isopropoxide, Journal of Alloys and Compounds, 358, 76–81.
- Mohammadi, B., Yousefi, A.A., Bellah, S. M. (2007) Effect of tensile strain rate and
elongation on crystalline structure and piezoelectric properties of PVDF
thin films, Polymer Testing, 26, pp 42-50.
- Mao, C., Dong, X., Zeng, T. Chen, H. Cao, F. (2006) Nonhydrolytic sol-gel synthesis
and dielectric properties of ultrafine-grained and homogenized
 $Ba_{0.7}Sr_{0.3}TiO_3$, Ceramic International, xxx, pp xxx
- Naarayan, S.S. Rao, L., and Sivakumak, S.M. (2005) Electromechanical behavior of
form I uniaxially and biaxial stretched PVDF, Journal of Ferroelectrics,
325, 155–164.
- Nayak, M., Lee, S.Y., and Tseng, T.Y. (2002) Electrical and dielectric
properties of $(Ba_{0.5}Sr_{0.5})TiO_3$ thin films prepared by a hydroxide–alkoxide
precursor-based sol–gel method, Materials Chemistry and Physics, 77,
34–42.
- Olszowy, M. (1997) Piezoelectricity and Dielectric Properties of
PVDF/ $BaTiO_3$ Composites, Journal of Der Spiegel, 3181, 69-72.

- Rollik, D., Bauer, S. and Gerhard-Multhaupt, R. (1999) Separate contributions to the pyroelectricity in poly(vinylidene fluoride) from the amorphous and crystalline phases, as well as from their interface, Journal of Applied Physics, 85(6), pp 3282-3288
- Salimi, A. and Yousefi, A. A. (2004) Conformational changes and phase transformation mechanisms in PVDF solution-cast films, Journal of Polymer Science: Part B: Polymer Physics, 42, 3487–3495.
- Safari, A. (1994) Development of piezoelectric composites for transducers. Journal of Physics III France, 4, 1129-1149.
- Tahan, D., Safari, A., and Klein, L. C. (1994) Sol-preparation of barium strontium titanate thin films, Journal of IEEE Applications of Ferroelectrics, 427-430.
- Wua, D., Li, A., Ling, H., Yin, X., Ge, C., Wang, M., Ming, N. (2000) Preparation (Ba_{0.5}Sr_{0.5})TiO₃ thin films by sol–gel method with rapid thermal annealing. Journal of Applied Surface Science, 165, 309–314.
- Zhou, L., Vilarinho, P. M., and Baptista, J. L. (1999) Dependence of the Structural and Dielectric Properties of Ba_{1-x}Sr_xTiO₃ Ceramic Solid Solutions Raw Material Processing. Journal of the European Ceramic Society, 19, 2015-2020.
- Su B., Holmes, J.E., Cheng, B.L. and Button, T.W. (2002) Processing Effects on the Microstructure and Dielectric Properties of barium Strontium Titanate (BST) Ceramics. Journal of Electroceramics, 9, 111–116

APPENDICES

Appendix A Barium Strontium Titanate Analysis

Table A1 Lattice parameter of calcine Ba_{0.7}Sr_{0.3}TiO₃ at 800°C before sintering

sol-gel calcine						
peak No.	X (nm)	(1~3)	h	k	l	2θ(degree)
1	0.154184	3	1	0	0	22.380
2	0.154184	3	1	1	0	31.800
3	0.154056	1	1	1	1	39.280
4	0.154056	1	2	0	0	45.660
5	0.154056	1	2	1	1	56.760
6	0.154056	1	2	2	0	66.680
a=	3.959					

Table A2 Lattice parameter of calcine Ba_{0.7}Sr_{0.3}TiO₃ at 800°C after sintering

sinter tetragonal						
peak No.	λ (nm)	(1~3)	h	k	l	2θ(degree)
1	0.154184	3	1	0	0	22.680
2	0.154184	3	1	1	0	32.100
3	0.154184	3	1	1	1	39.540
4	0.154184	3	2	0	0	45.960
5	0.154184	3	2	1	1	56.960
6	0.154184	3	2	2	0	66.740
a=	3.9794					
c=	3.98214					

Table A3 Lattice parameter of calcine Ba_{0.7}Sr_{0.3}TiO₃ at 100°C before sintering

BST sol-gel at 1000 is cubic						
peak No.	X (nm)	(1~3)	h	k	l	2θ(degree)
1	0.154184	3	1	0	0	22.200
2	0.154184	3	1	1	0	31.800
3	0.154056	1	1	1	1	39.200
4	0.154056	1	2	0	0	45.600
5	0.154056	1	2	1	1	56.800
6	0.154056	1	2	2	0	66.600
a	3.95954					

Table A4 Lattice parameter of calcine $\text{Ba}_{0.7}\text{Sr}_{0.3}\text{TiO}_3$ at 1000°C after sintering

BST sinter at 1000 is tetragonal

peak No.	X (nm)	(1~3)	h	k	l	2 θ (degree)
1	0.154184	3	1	0	0	22.640
2	0.154184	3	1	1	0	32.080
3	0.154184	3	1	1	1	39.900
4	0.154184	3	2	0	0	45.840
5	0.154184	3	2	1	1	56.860
6	0.154184	3	2	2	0	66.680
a	3.9903					
c	3.95356					

Appendix B Polyvinylidene Fluoride Analysis

% Crystallinity of PVDF is calculated by following Eq. 1:

$$\% \text{crystallinity} = (\Delta H_f / \Delta H_o^f) \times 100 \quad (1)$$

where ΔH_o^f is heat of fusion of 100 % crystallized PVDF ΔH_f is the measure heat fusion of each them

Table B1 Melting temperature and crystallinity temperature of PVDF at different ratio

Sample	T _m (°C)		T _c (°C)	
	Onset	Peak	Onset	Peak
Pure PVDF	167.915	172.666	156.861	151.3
ratio2 PVDF	165	168.866	141.26	136.8
ratio4	165.803	170.533	142.087	134.966

Table B2 % Crystallinity of PVDF

Sample	ΔH_f (J/g)	% Crystallinity	True % Crystallinity	% PVDF	ΔH_o^f (J/g)
Pure PVDF	31.648	35.00885	35.00885	100	90.4
ratio2 PVDF	39.817	44.04535	44.04535	100	90.4
ratio4	40.478	44.77655	44.77655	100	90.4

F(β) of PVDF is calculated by following Eq. 2:

$$F(\beta) = \frac{X_\beta}{X_\alpha + X_\beta} = \frac{A_\beta}{(K_\beta + K_\alpha)A_\alpha + A_\beta} = \frac{A_\beta}{1.26 A_\alpha + A_\beta} \quad (2)$$

Table B3 F(β) of PVDF

pure PVDF	A β (763)	1.66		
solution PVDF	A β (763)	2.5		
ratio2	A β (763)	2.45		
ratio4	A β (763)	2.4		
pure PVDF	A α (840)	1.68	A α (840)*1.26	2.1168
solution PVDF	A α (840)	1.3	A α (840)*1.26	1.638
ratio2	A α (840)	1.58	A α (840)*1.26	1.9908
ratio4	A α (840)	1.3	A α (840)*1.26	1.638
pure PVDF	A α (840)*1.26+A β (763)	3.7768		
solution PVDF	A α (840)*1.26+A β (763)	4.138		
ratio2	A α (840)*1.26+A β (763)	4.4408		
ratio4	A α (840)*1.26+A β (763)	4.038		
pure PVDF	F(β)	0.439526	%	43.95255
solution PVDF	F(β)	0.604157	%	60.41566
ratio2	F(β)	0.551702	%	55.17024
ratio4	F(β)	0.594354	%	59.43536

Table B4 Physical properties of PVDF (SOLEF 1008).

SOLEF® 1008

PVDF Homopolymer

Low viscosity - Injection

Physical properties	Standards	Units	
Density	ISO 1183	g/cm ³	1.78
Water absorption (24 h at 23°C)	ISO 62 (method 1)	%	< 0.04
Melt Flow Index	ASTM D 1238		
	230°C, 10 kg	g/10 min	-
	230°C, 5 kg	g/10 min	24
	230°C, 2.16 kg	g/10 min	8
Mechanical properties			
Tensile	ASTM D 638		
Tensile stress at yield	23 °C, 50 mm/min	MPa/psi	53 - 57/7685 - 826
Tensile stress at break		MPa/psi	35 - 50/5075 - 725
Elongation at yield	23 °C, 1 mm/min	%	5 - 10
Elongation at break		%	5 - 10
Modulus	23 °C, 1 mm/min	MPa/psi	2600/377000
Flexion	ASTM D 790		
Maximum load	23 °C	MPa/psi	79/11310
Modulus	2 mm/min	MPa/psi	2200/319000
IZOD Impact (notched V 10 mm - at 23 °C - 4 mm thick)	ASTM D 256	J/m	55
Shore D Hardness (2 mm thick)	ASTM D 2240	-	78
Abrasion resistance	TABER CS 10/1 kg	mg/1000 rev	5 - 10
Friction coefficient: static	ASTM D 1894	-	0.2 - 0.4
dynamic			0.2 - 0.3
Thermal properties			
Crystallinity by DSC	ASTM D 3418		
Melting point		°C/°F	174/345
Heat of fusion (80 °C to end of melting)		J/g	67
Crystallizing point		°C/°F	140/284
Crystallizing heat		J/g	59
VICAT point (4 mm thick) load 1 kg	ISO 306	°C/°F	171/340
Deflection temperature (4 mm thick) load 0.46 MPa	ASTM D 648 after annealing	°C/°F	148/298
load 1.82 MPa		°C/°F	115/239
Glass transition (T _g)	DMTA	°C/°F	-30/ -22
Brittleness temperature (on 2 mm pressed sheet)	ASTM D 746 A	°C/°F	0 - 10/32 - 50
Thermal properties (continuation)			
Molding shrinkage		%	2 - 3
Thermal stability	TGA beginning and at 1% weight loss in air	°C/°F	375 - 400/707 - 752
Linear thermal expansion coefficient	ASTM D 696	10 ⁻⁶ K ⁻¹	120 - 140
Thermal conductance at 23°C	ASTM C 177	W/m.K	0.2
Specific heat	23 °C & 100 °C	J/g.K	1.2 - 1.6
Electrical properties			
Surface resistivity	ASTM D 257		
Voltage < 1V, after 2 min - 500 V at 23 °C	DIN 53483	ohm/square	≥ 1.10 ¹⁴
Volume resistivity	ASTM D 257		
Intensity = 10 mA, after 2 min at 23 °C	DIN 53483	ohm.cm	≥ 1.10 ¹⁴
Fire resistance			
UL-94 Flammability test	UL-94	Class	V-0
Limiting Oxygen Index (sheet 3 mm thick)	ASTM D 2863	%	44

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1. Kittikun Kohpaiboon, Hathaikarn Manuspiya, Pitak Laoratanakul (2006, December 10-14) 0-3 Connectivity of PVDF/BST Piezoelectric Composites. Poster presented at 5th Asian Meeting on Electroceramics, Bangkok, Thailand.