REFERENCES

- Agarwal, S. and Sharma, G.L. (2002) Humidity sensing properties of (Ba, Sr) TiO₃ thin films grown by hydrothermal–electrochemical method. <u>Sensors and</u> <u>Actuators B: Chemical</u>, 85, 205–211.
- Basse, D.C. (Eds.). (1982) <u>Poly(vinylidene fluoride)</u> <u>Developments in Crystalline</u> <u>Polymers</u>. London: Applied Sciences.
- Biesheuvel, P.M., Verweij, H., (1999) Theory of Cast Formation in Electrophoretic Deposition. <u>Journal of Ameican Ceramic Society</u>, 82, 1451–1455.
- Botelho, G., Lanceros-Mendez, S., Goncalves, A.M., Sencadas, V., Rocha. J.G.
 (2008) Relationship between processing conditions, defects and thermal degradation of poly(vinylidene fluoride) in the β-phase. Journal of Non-Crystalline Solids, 354, 72–78
- Celina, M., Dargaville, T.R., Assink R.A. and Martin J.W. (2005) Selection and Optimization of Piezoelectric Polyvinylidene Fluoride Polymers for Adaptive Optics in Space Environments. <u>High Performace Polymer</u>, 17; 575.
- Das-Gupta, D.K., Abdullah, M. and Shorrocks, N.M. (1988) Dielectric and Pyroelectric Properties of Polymer/Ceramic Composites. <u>Journal of Materials</u> <u>Science Letters</u>, 7, 167-170
- Dong L.J., Xiong C.X., Chen J. and Nan, C.W. (2004) Dielectric Behavior of BaTiO₃/PVDF Nanocomposites *In-situ* Synthesized by the Sol-Gel Method. Journal of Wuhan University of Technology - Mater. Sci. Ed, 19, 9-11.
- Ezhilvalavan, S., and Tseng, T. (2000) Progress in the developments of (Ba, Sr)TiO₃ (BST) thin films for Gigabit era DRAMs. <u>Materials Chemistry and</u> <u>Physics</u>. 65, 227–248.
- Fiedziuszko, S.J., Hunter, I.C., Itoh, T., Kobayashi, Y., Nishikawa, T., Stitzer, S.N., and Wakino, K. (2002) Dielectric Materials, Devises, and Circuits. <u>IEEE</u> <u>Transactions on Microwave Theory and Technique</u>, MTT-50, 706–20.
- George, J.E. (2007) Piezoelectric Sensing Capabilities of Polyvinylidene Fluoride: Application to a Fluid Flow through a Complian Tube. <u>Class 2007</u> <u>Aerospace Engineering Texas A&M University.</u>

- Greaves, R.W., Fowler, E.P., Goodings, A., Lamb, D.R. (1974) The Direct
 Piezoelectric Effect in Extruded Polyethylene. Journal of Materials Science, 9, 1602-1608.
- Harrison, J.S. and Ounaies, Z. (2001) <u>Piezoelectric Polymers</u>. Virginia: National Aeronautics and Space Administration.
- Hayakawa, R. and Wada, Y. (1973) Piezoelectricity and related properties of polymer films. <u>Advances in Poymer Science</u>, 11, 1-55.
- Jeon, J.H. (2004) Effect of SrTiO₃ concentration and sintering temperature on microstructure and dielectric constant of Ba_{1-x}Sr_xTiO₃. Journal of the <u>European Ceramic Society</u>, 24, 1045–1048
- Kohpaiboon, K., and Manuspiya, H. (2007) 0-3 Connectivity of PVDF/BST
 Piezoelectric Composites <u>M.S. thesis</u>, The Petroleum and Petrochemical
 College, Chulalongkorn University, Bangkok, Thailand.
- Kozyrev, A., Ivanov. A., Samoilova. T., Soldatenkov, O., Astafiev, K., Sengupta, L.S. (2000) Nonlinear response and power handling capability of ferroelectric Ba_xSr_{1-x}TiO₃ film capacitors and tunable microwave devices. <u>Journal of Applied Physics</u>, 88, 5334-5342.
- Lilliehorn, T., Blom, T., Simu, U. and Johansson S. (2005) Multilayer piezoelectric copolymer transducers. <u>IEEE Ultrasonics Symposium</u>, 1618-1620.
- Mazur, K. (1989) Polarization Effects in Uncharged Laminate. <u>IEEE Proc. ICSD</u>, <u>Trondheim</u>, 89, 437-441.
- Mazur, K. (1990) Polyester-Urethane Elastomer as a Component of Piezoelectric Laminates. <u>Communications of IRC, Paris</u>, 90, 357-358.
- Mazur, K. (1992) Piezoelectricity of PVDF/PUE, PVDF/PMMA and PVDF/PMMA+BaTiO₃ Laminates. <u>IEEE Transactions on Electrical</u> <u>Insulation</u>, 27(4), 782-786.
- Mort, J. and Pfister, G. (Eds.). (1982) <u>Electronic Properties of Polymers</u>. New York: John Wiley.
- Nalwa, H.S. (Eds.). (1995) <u>Ferroelectric Polymers Chemistry, Physics, and</u> <u>Applications</u>. New York: Marcel Dekker, Inc.
- Robinson, S., Preston, R., Smith, M. and Millar, C. (2000) PVDF Reference Hydrophone Development in the UK—From Fabrication and Lamination to

Use as Secondary Standards. <u>IEEE Transactions on Ultrasonics</u>,

Ferroelectrics, and Frequency Control, 47(6), 1336-1344.

- Scheinbiem, J.I., Newman, B.A. and Su, J. (1994) Piezoelectric laminate films and processes for their manufature. <u>U.S. Patent</u>, 5356500.
- Sitting, E.K. (1971) Definitions Relating to Conversion Losses in Piezoelectric transducers. <u>IEEE Transaction on Sonics and Ultrasonics</u>, SU-18, 231-234.
- Swartz, R.G. and Plummer, J.D. (1980). On the generation of high-frequency acoustic energy with polyvinylidene fluoride. <u>IEEE Transaction on Sonics</u> <u>and Ultrasonics</u>, SU-27(6), 295-303.
- Wada, Y. (1987) Ferro-, Piezo-, and Pyroelectricity. <u>IEEE Transactions on</u> <u>Electrical Insulation</u>, EI-22(3), 255-259.
- Wada, Y. and Hayakawa, R. (1976). Piezoelectricity and Pyroelectricity of Polymers. <u>Japanese Journal of Applied Physics</u>, 15, 2041-2057.
- Zhang Q. and Lewin, P.A. (1993). Wideband and efficient polymer transducers using multiple active piezoelectric films. <u>IEEE Ultrasonics Symposium</u>, 757-760.
- Zheng, J. P. (2000) Dielectric Properties of PVDF Films and Polymer Laminates with PVDF for Energy Storage Applications. <u>IEEE</u>, 1, 423-426.
- Zheng, J.P., Cygan, P.J., and Jow, T.R. (1996) Investigation of Dielectric Properties of Polymer Laminates with Polyvinylidene Fluoride. <u>IEEE Transactions on</u> <u>Dielectrics and Electrical Insulation</u>, 3(1), 144-147.

APPENDICES

Appendix A Barium Strontium Titanate Analysis

Lattice parameter of the calcined $Ba_{0.7}Sr_{0.3}TiO_3$ at $1000^{\circ}C$



Table A1 Lattice parameter of calcined $Ba_{0.7}Sr_{0.3}TiO_3$ at 1000°C

p	eak No.	λ (nm)	(1~3)	h	k	1	2θ(degree)
1		0.15418	3	1	0	0	22.33
2		0.15418	3	1	1	0	31.8
3		0.15418	3	1	1	1	39.28
4		0.15418	3	2	0	0	45.66
5		0.15418	3	2	1	1	56.76
6		0.15418	3	2	2	0	66.68
a=	0.3971				1	1	1
c=	0.39697	1					
c/a	0.99967]					

Lattice parameter of the sintered Ba_{0.7}Sr_{0.3}TiO₃ at 1330°C



Table A2 Lattice parameter of sintered $Ba_{0.7}Sr_{0.3}TiO_3$ at 1330°C for 2 h.

p	eak No.	λ (nm)	(1~3)	h	k	l	2θ(degree)
1		0.15418	3	1	0	0	22.44
2		0.15418	3	1	1	0	31.9
3		0.15418	1	1	1	1	39.3
4		0.15418	1	2	0	0	45.72
5		0.15418	1	2	1	1	56.76
6		0.15418	1	2	2	0	66.52
a=	0.3969			I	1	1	
c=	0.39749						
c/a	1.001325						

Appendix B Polyvinylidene Fluoride Analysis

The degree of crystallinity was measured as the ratio between $\Delta H_{\rm m}$ and ΔH_0 , as following equation

% Crystallinity =
$$\frac{\Delta H_m}{\Delta H_0} x 100$$

where $\Delta H_{\rm m}$ is the melting enthalpy of the material under study and ΔH_0 is the melting enthalpy of totally crystalline material ($\Delta H_0 = 104.50$ J/g for PVDF).

Table B1% Crystallinity of PVDF

Material	T _m Onset (°C)	Tm Peak (°C)	heat of fusion ΔH _m , J/g	Crystallinity %
solution-casted PVDF	162.3	172.3	65.5	62.6
stretched PVDF X2	162.2	170.1	65.9	63.1
stretched PVDF X4	161.9	170.3	64.4	61.6
stretched PVDF X6	165.3	169.1	66.3	63.4

F(β) of PVDF system containing α and β phase the relative fraction of the β phase, $F(\beta)$, was calculated by the following equation:

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

Where X_{α} and X_{β} are the degree of crystallinity of α and β phases, A_{α} and A_{β} are the absorbances of α and β phases at 763 and 840 cm⁻¹, respectively.

Table B2 $F(\beta)$ of PVDF

Material	$\begin{array}{c} A_{\alpha} \text{ (normalized} \\ 763 \text{ cm}^{-1} \text{)} \end{array}$	A_{eta} (normalized 840 cm ⁻¹)	% β phase content $F(\beta)$,
solution-casted PVDF	0.01695	0.42586	95.22
stretched PVDF X2	0.02108	0.33825	92.72
stretched PVDF X4	0.027384	0.43282	92.62
stretched PVDF X6	0.030339	0.446089	92.11

Table B3 Physical properties of PVDF (SOLEF 1008).

SOLEF® 1008 PVDF Homopolymer

SOLEF

Low viscosity - Injection			
Physical properties	Standards	Units	
Density	ISO 1183	g/cm³	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		MPa/psi	53 - 57/7685 - 8265
Tensile stress at break	- 23 °C, 50 mm/min	MPa/psi	35 - 50/5076 - 7250
Elongation at yield		%	5 - 10
Elongation at break		%	5 - 10
Modulus	23 °C, 1 mm/min	MPa/psi	2600/377000
Flexion	ASTM D 790		8
Maximum load	23 °C	MPa/psi	78/11310
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	The state of the second	78
Abrasion resistance	TABER CS 10/1 kg	mg/1000 nev	5 - 10
Friction coefficient static	ASTM D 1894		0.2 - 0.4
dynamic	and the state		0.2 - 0.3
Thermal properties			
Crystallinity by DSC	ASTM D 3418		
Molting point		°C/°F	174/345
Heat of fusion (80 °C to end of metting)		J/g	67
Crystallizing point		°C/°F	140/284
Crystallizing heat		J/g	59
VICAT point (4 mm thick)	ISO 306	Chapter 1	Marine Co.
load 1 kg		°C/*F	171/340
Deflection temperature (4 mm thick)	ASTM D 648		
load 0.46 MPa	after annoaling	°C/*F	148/298
load 1.82 MPa		°C/*F	115/239
Glass transition (Tg)	DMTA	°C/°F	- 30/ -22
Brittleness temperature	ASTM D 746 A	°C/°F	0 - 10/32 - 50
(on 3 mm proceed sheet)			

page

Solvay Solexis



SOLEF

SOLEF® 1008

PVDF Homopolymer

hermal properties (continuation)	Standards	Units	
Molding shrinkage		%	2 - 3
Thermal stability	TGA beginning- and al 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

Voltage < 1V, after 2 min - 500 V at 23 °C	Intensity = 10 mA, after 2 min al 23 °C	an E	DIN 53483	ohm.cm	≥ 1.10 ¹⁴
Voltage < 1V, after 2 min - 500 V at 23 °C ↓ DIN 53483 ohm/square ≥ 1.10 ¹⁴	Volume resistivity	5	ASTM D 257		
	Voltage < 1V, after 2 min - 500 V at 23 °C	L	DIN 53483	ohm/squana	≥ 1.10 ¹⁴

UL-94 Flammabili	ty test	UL-94	Class	V-0	
Limiting Oxygen I	ndex (sheet 3 mm thick)	ASTM D 2863	%	44	

The data and numerical results contained in this document are provided for the sake of general information and are given in good faith. They reflect the state of our knowledge all the time of publication. Because the possibilities and application conditions of our product are many and varied, and lie beyond our control, we can in no event be held responsible all the necessary information on planned applications have not been formally brough to our attention. The information presented here cannot be considered as a suggestion to use our product without taking into account existing patents, or legal provisions or regulations, whether national or local. The purchaser is obliged to verify whether the possession, use or marketing of our products is subject within his territory to paticular rules, especially with respect to public health, hygiene and worker and/or consumer safely. The purchaser alone assumes the duties of information and advice for the utimate uses. Solvey Soleris can in no event be held responsible for a possible failure on the part of the purchaser to respect these regulations, proveiuna duties.

January 2003

page 2/2 © 2000-2003. SOLVAY SOLEXIS S.A. Rue de Ransbeek, 310 - B 1120 Brussels (Belgium) www.solvaysolexis.com - Tel., 32 2 / 264 21 11 - Fax. 32 2 / 264 35 53





Appendix C Lamination Film Analysis

The effective dielectric constant of the multilayer structure is a series connection model of PVDF and PVDF/BST composite layer dielectrics. the effective dielectric constant, ε_{eff} , can theoretically be expressed as:

$$\frac{1}{\varepsilon_{eff}} = \left[\frac{1}{\varepsilon_1} + \frac{1}{\varepsilon_2} + \frac{1}{\varepsilon_3} + \dots\right] / (t_1 + t_2 + t_3 + \dots)$$
 where *t* is thickness of each layer

Table C1 Dielectric constant of PVDF x4 and PVDF/BST40% composite

Matarial	Thickness	Dielectric constant (ε_r)						
	(µm)	1 kHz	10 kHz	100 kHz	1 MHz			
PVDF x4	40	4.77	4.60	4.37	3.89			
composite	280	23.03	22.53	21.62	18.41			

 Table C2
 Dielectric constant of Laminate films

	Dielectric constant								
Material	11	cHz	10 kHz		100 kHz		1 MHz		
	εr	Eeff	Er	Eeff	E _r	Eeff	Er	Eeff	
P-C	19.01	15.58	18.63	15.15	17.8	14.48	15.03	12.55	
P-C-P	17.22	12.445	16.84	12.07	16.18	11.52	13.64	10.06	
C-P-C	18.48	18.35	18.09	17.88	17.45	17.12	14.9	14.74	

RemarkP-C is PVDF-Composite bilayer filmP-C-P is PVDF-Composite-PVDF trilayer filmC-P-C is PVDF-Composite-PVDF trilayer film ε_r is experimental dielectric constant

CIRRICULUM VITAE

Name:Mr. Teerapol SodsongDate of Birth:January 28, 1985

Nationality: Thai

University Education:

2003-2007 Bachelor Degree of Science in Chemistry, Faculty of Science, Chulalongkorn University, Bangkok, Thailand.

Proceeding:

- Suwansumpan, D.; Sodsong, T.; Manuspiya H.; Laoratanakul P.; and Amar S. Bhalla. (2008, August 19-20) Porous PVDF Film: Effect of bubble shapes to Piezoelectric Properties. <u>Proceedings of the Thai-Japan Joint Symposium on</u> <u>Advances in Materials Science and Environmental Technology 2008</u>, Bangkok, Thailand.
- Sodsong, T.; and Manuspiya, H. (2009, March 22-27) Improvement in dielectric properties by using lamination technique of stretched polyvinylidene fluoride and polyvinylidene fluoride/barium strontium titanate composite film. <u>Proceedings of the NanoThailand Symposium 2008</u>, Bangkok, Thailand.
- Sodsong, T.; and Manuspiya, H. (2009, April 22) Piezoelectric Polymer for Mechanical Sensor in Smart Card Application. <u>Proceedings of the 15th PPC</u> <u>Symposium on Petroleum, Petrochems, and Polymers, Bangkok, Thailand.</u>

Presentations:

- Suwansumpan, D.; Sodsong, T.; Manuspiya H.; Laoratanakul P.; and Amar S. Bhalla. (2008, August 19-20) Porous PVDF Film: Effect of bubble shapes to Piezoelectric Properties. Paper presented at <u>the Thai-Japan Joint Symposium on</u> <u>Advances in Materials Science and Environmental Technology 2008</u>, Bangkok, Thailand.
- Sodsong, T.; and Manuspiya, H. (2009, March 22-27) Improvement in dielectric properties by using lamination technique of stretched polyvinylidene fluoride and polyvinylidene fluoride/barium strontium titanate composite film. Paper presented at the NanoThailand Symposium 2008, Bangkok, Thailand.



 Sodsong, T.; and Manuspiya, H. (2009, April 22) Piezoelectric Polymer for Mechanical Sensor in Smart Card Application. Paper presented at <u>the 15th PPC</u> <u>Symposium on Petroleum, Petrochems, and Polymers</u>, Bangkok, Thailand.