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APPENDICES

Appendix A Lattice Parameter Calculations

Table A1 The identification of XRD peaks of the $Ba_{0.7}Sr_{0.3}Mg_0TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.509	3.9468	1	0	0	3.9468	
31.947	2.799	1	1	0	3.9584	
39.353	2.2877	1	1	1	3.9468	3.9941
45.753	1.9815	2	0	0	3.9629	
51.496	1.7731	2	0	1	3.9468	4.0398
56.812	1.6192	2	1	1	3.9468	4.0679
Average Lattice Constants =					3.9514	4.0339
					c/a	1.0209

Table A2 The identification of XRD peaks of the $Ba_{0.695}Sr_{0.3}Mg_{0.005}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.47	3.9536	1	0	0	3.9536	
31.907	2.8024	1	1	0	3.9632	
39.315	2.2898	1	1	1	3.9536	3.9911
45.711	1.9832	2	0	0	3.9663	
51.455	1.7745	2	0	1	3.9536	4.0261
56.774	1.6202	2	1	1	3.9536	4.0464
Average Lattice Constants =					3.9573	4.0212
					c/a	1.016

Table A3 The identification of XRD peaks of the $Ba_{0.69}Sr_{0.3}Mg_{0.01}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.48	3.9518	1	0	0	3.9518	
31.92	2.8013	1	1	0	3.9617	
39.329	2.289	1	1	1	3.9518	3.9907
45.73	1.9824	2	0	0	3.9648	
51.479	1.7737	2	0	1	3.9518	4.0248
56.771	1.6203	2	1	1	3.9518	4.0572
Average Lattice Constants =					3.9556	4.0242
					c/a	1.0173

Table A4 The identification of XRD peaks of the $Ba_{0.68}Sr_{0.3}Mg_{0.02}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.433	3.9599	1	0	0	3.9599	
31.877	2.8051	1	1	0	3.9669	
39.282	2.2916	1	1	1	3.9599	3.9881
45.679	1.9845	2	0	0	3.969	
51.409	1.7759	2	0	1	3.9599	4.0169
56.712	1.6218	2	1	1	3.9599	4.0383
Average Lattice Constants =					3.9626	4.0144
					c/a	1.0131

Table A5 The identification of XRD peaks of the $Ba_{0.6}Sr_{0.4}Mg_{0}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.524	3.9442	1	0	0	3.9442	
32.012	2.7935	1	1	0	3.9506	
39.446	2.2825	1	1	1	3.9442	3.9719
45.872	1.9766	2	0	0	3.9532	
51.634	1.7687	2	0	1	3.9442	3.9992
56.965	1.6152	2	1	1	3.9442	4.0197
Average Lattice Constants =					3.9468	3.9969
					c/a	1.0127

Table A6 The identification of XRD peaks of the $Ba_{0.595}Sr_{0.4}Mg_{0.005}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.496	3.9491	1	0	0	3.9491	
31.983	2.796	1	1	0	3.9542	
39.41	2.2845	1	1	1	3.9491	3.9726
45.817	1.9788	2	0	0	3.9577	
51.574	1.7707	2	0	1	3.9491	4.0009
56.916	1.6165	2	1	1	3.9491	4.0135
Average Lattice Constants =					3.9514	3.9957
					c/a	1.0112

Table A7 The identification of XRD peaks of the $Ba_{0.59}Sr_{0.4}Mg_{0.01}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.463	3.9547	1	0	0	3.9547	
31.922	2.8012	1	1	0	3.9615	
39.342	2.2883	1	1	1	3.9547	3.981
45.746	1.9817	2	0	0	3.9634	
51.507	1.7728	2	0	1	3.9547	4.0024
56.836	1.6186	2	1	1	3.9547	4.0156
Average Lattice Constants =					3.9573	3.9997
					c/a	1.0107

Table A8 The identification of XRD peaks of the $Ba_{0.58}Sr_{0.4}Mg_{0.02}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.504	3.9476	1	0	0	3.9476	
31.989	2.7955	1	1	0	3.9534	
39.423	2.2838	1	1	1	3.9476	3.9717
45.832	1.9782	2	0	0	3.9564	
51.603	1.7697	2	0	1	3.9476	3.9963
56.931	1.6161	2	1	1	3.9476	4.0152
Average Lattice Constants =					3.95	3.9944
					c/a	1.0112

Table A9 The identification of XRD peaks of the $Ba_{0.5}Sr_{0.5}Mg_0TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.61	3.9293	1	0	0	3.9293	
32.123	2.7841	1	1	0	3.9373	
39.583	2.2749	1	1	1	3.9293	3.9624
45.999	1.9714	2	0	0	3.9429	
51.814	1.763	2	0	1	3.9293	3.9952
57.137	1.6108	2	1	1	3.9293	4.0299
Average Lattice Constants =					3.9329	3.9958
					c/a	1.0160

Table A10 The identification of XRD peaks of the $Ba_{0.495}Sr_{0.5}Mg_{0.005}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.586	3.9335	1	0	0	3.9335	
32.094	2.7866	1	1	0	3.9408	
39.536	2.2775	1	1	1	3.9335	3.9677
45.974	1.9724	2	0	0	3.9449	
51.766	1.7645	2	0	1	3.9335	3.9953
57.129	1.611	2	1	1	3.9335	4.0107
Average Lattice Constants =					3.9366	3.9912
					c/a	1.0139

Table A11 The identification of XRD peaks of the $Ba_{0.49}Sr_{0.5}Mg_{0.01}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.572	3.9359	1	0	0	3.9359	
32.071	2.7886	1	1	0	3.9436	
39.514	2.2787	1	1	1	3.9359	3.969
45.942	1.9738	2	0	0	3.9475	
51.707	1.7664	2	0	1	3.9359	4.0069
57.08	1.6122	2	1	1	3.9359	4.0176
Average Lattice Constants =					3.9391	3.9978
					c/a	1.0149

Table A12 The identification of XRD peaks of the $Ba_{0.48}Sr_{0.5}Mg_{0.02}TiO_3$ powder

2-Theta	d(A)	h	k	l	a-Axis	c-Axis
22.537	3.9419	1	0	0	3.9419	
32.06	2.7894	1	1	0	3.9448	
39.503	2.2793	1	1	1	3.9419	3.9602
45.926	1.9744	2	0	0	3.9488	
51.734	1.7655	2	0	1	3.9419	3.9721
57.059	1.6128	2	1	1	3.9419	3.9945
Average Lattice Constants =					3.9435	3.9756
					c/a	1.008

Appendix B Frequency-dependent Dielectric Properties of Magnesium-Doped Barium Strontium Titanate Powder and PBS-Composite Thin Film

Table B1 Frequency-dependent dielectric constant of magnesium-doped barium strontium titanate powder

$Ba_{1-x-y}Sr_xMg_yTiO_3$		Frequency				
x	y	1E+06	5E+06	1E+07	5E+07	1E+08
0.3	0	602.60	428.23	422.93	421.87	420.27
	0.005	524.12	372.68	369.12	369.47	368.34
	0.01	485.93	341.84	337.99	337.69	336.13
	0.02	426.91	302.31	299.19	299.52	298.66
0.4	0	486.87	345.33	341.28	340.52	338.63
	0.005	470.00	333.91	330.17	330.25	329.07
	0.01	459.37	327.31	324.15	324.23	323.03
	0.02	381.76	270.70	268.34	268.63	267.34
0.5	0	470.76	332.90	328.80	328.29	326.53
	0.005	446.90	316.42	313.00	312.74	311.00
	0.01	416.78	295.69	292.88	293.42	292.43
	0.02	233.97	164.68	163.10	163.60	163.01

Table B2 Frequency-dependent loss tangent of magnesium-doped barium strontium titanate powder

$Ba_{1-x-y}Sr_xMg_yTiO_3$		Frequency				
x	y	1E+06	5E+06	1E+07	5E+07	1E+08
0.3	0	0.1127	0.0165	0.0171	0.0168	0.0146
	0.005	0.1057	0.0115	0.0125	0.0127	0.0106
	0.01	0.1099	0.0148	0.0154	0.0140	0.0111
	0.02	0.1057	0.0120	0.0129	0.0130	0.0110
0.4	0	0.1084	0.0148	0.0155	0.0153	0.0128
	0.005	0.1088	0.0132	0.0143	0.0131	0.0101
	0.01	0.1072	0.0126	0.0143	0.0132	0.0105
	0.02	0.1047	0.0100	0.0120	0.0119	0.0094
0.5	0	0.1097	0.0159	0.0160	0.0145	0.0116
	0.005	0.1093	0.0139	0.0143	0.0138	0.0110
	0.01	0.1051	0.0113	0.0114	0.0114	0.0089
	0.02	0.0998	0.0095	0.0106	0.0108	0.0080

Table B3 Frequency-dependent dielectric constant of PBS-composite thin film

PBS/BST	**Frequency**				
	1E+07	**5E+07**	**1E+08**	**5E+08**	**1E+09**
100/0	4.56	3.63	3.14	2.24	1.99
90/10	5.26	3.63	3.23	2.70	2.55
80/20	9.08	4.72	4.09	3.30	3.08
70/30	12.13	5.57	4.56	3.63	3.36
60/40	16.70	6.75	5.21	3.42	2.94
50/50	18.70	8.75	7.21	5.42	4.94

Table B4 Frequency-dependent loss tangent of PBS-composite thin film

PBS/BST	**Frequency**				
	1E+07	**5E+07**	**1E+08**	**5E+08**	**1E+09**
100/0	0.3749	0.2612	0.2902	0.2511	0.2045
90/10	0.4004	0.2866	0.2400	0.1340	0.0962
80/20	0.5114	0.4528	0.3472	0.1891	0.1409
70/30	0.5647	0.5757	0.4431	0.2113	0.1587
60/40	0.7203	0.5856	0.4579	0.2283	0.1559
50/50	0.7473	0.6126	0.4849	0.2553	0.1829

Appendix C Data Sheet Of Poly(Butylene Succinate) Grade Blown Film Extrusion (AZ91TN)

Table C1 Data sheet of poly(butylene succinate) grade blown film extrusion (AZ91TN)

Properties	Test method	Unit	AZ91TN
MFR (190℃/21.18 N)	ISO1133	g/10 min	4
Specific gravity	ISO1183	g/cm^3	1.26
Flexural modulus	ISO178	MPa	530
Flexural strength	ISO178	MPa	34
Yield stress	ISO527	MPa	37
Stress at break	ISO527	MPa	37
Strain at break	ISO527	%	300
Izod impact strenght (23℃)	ISO180	kJ/m^2	8.2
Deflection temperature under load (0.45 MPa)	ISO75-2	℃	84
Rockwell hardness (R scale)	ISO2039-2	-	96

Appendix D Breakdown Strength and Effects of Corona-poling of Poly(butylene succinate)/Barium Strontium Titanate Thin-film Composites on Dielectric Constant at Low-Frequency

Table D1 Breakdown strength of poly(butylene succinate)/barium strontium titanate thin-film composites

BST content	Breakdown Strength (kV/mm)
0 wt%	26.30
10 wt%	24.76
20 wt%	24.00
30 wt%	18.10
40 wt%	16.40
50 wt%	15.30

Table D2 Frequency-dependent dielectric constants of poly(butylene succinate)/barium strontium titanate thin-film composites (poled at 12.5 kV/mm, 80℃, and 30 min)

Frequency	State	Frequency				
		1.00E+02	1.00E+03	1.00E+04	1.00E+05	1.00E+06
0 wt%	unpoled	3.11	3.07	3.03	2.95	2.22
	poled	2.99	2.98	2.93	2.83	1.91
10 wt%	unpoled	3.23	3.19	3.12	3.02	2.76
	poled	2.05	2.01	1.98	1.92	1.78
20 wt%	unpoled	4.02	3.98	3.91	3.80	3.54
	poled	3.92	3.88	3.81	3.69	3.43
30 wt%	unpoled	7.88	7.57	7.17	6.80	6.43
	poled	5.91	5.67	5.38	5.10	4.82
40 wt%	unpoled	8.52	8.19	7.92	7.64	7.26
	poled	6.82	6.55	6.33	6.11	5.81
50 wt%	unpoled	17.11	16.79	16.48	16.11	15.61
	poled	14.54	14.27	14.00	13.69	13.27

Appendix E Experimental Data Fitting of Poly(butylene succinate)/Barium Strontium Titanate Thin-film Composites

The prediction of dielectric constant of polymer matrix composite in 0-3 connectivity system as the function of dielectric constant of each phase and filler volume fraction. The equations below were used to calculate the dielectric constant of PBS-BST composite at low filler volume fraction.

Series mixing rule :

$$\frac{1}{\varepsilon'_{eff}} = \frac{V_f}{\varepsilon'_{filler}} + \frac{(1-V_f)}{\varepsilon'_{matrix}}$$

Modified-Lichtnecker's mixing rule :

$$\log \varepsilon'_{eff} = \log \varepsilon'_{matrix} + (1-n)V_f \log(\frac{\varepsilon'_{filler}}{\varepsilon'_{matrix}})$$

Maxwell-Wagner equation :

$$\varepsilon'_{eff} = \varepsilon'_{matrix}\left[\frac{2\varepsilon'_{matrix}+\varepsilon'_{filler}+2V_f(\varepsilon'_{filler}-\varepsilon'_{matrix})}{2\varepsilon'_{matrix}+\varepsilon'_{filler}-V_f(\varepsilon'_{filler}-\varepsilon'_{matrix})}\right]$$

Where ε'_{eff} , ε'_{filler} , and ε'_{matrix} are the dielectric constant of the composite, filler, and polymer matrix, respectively, V_f is the volume fraction of the filler. In Modified-Lichtnecker's equation, n is fitting parameter, which was reported to be 0.3 in well-dispersed system. In this work, the experimental data was mostly fitted to the Modified-Lichtnecker's equation where n = 0.28 and at the filler volume fraction between 0.2 and 0.4, which can be implied that the fillers phase were essentially dispersed in matrix phase.

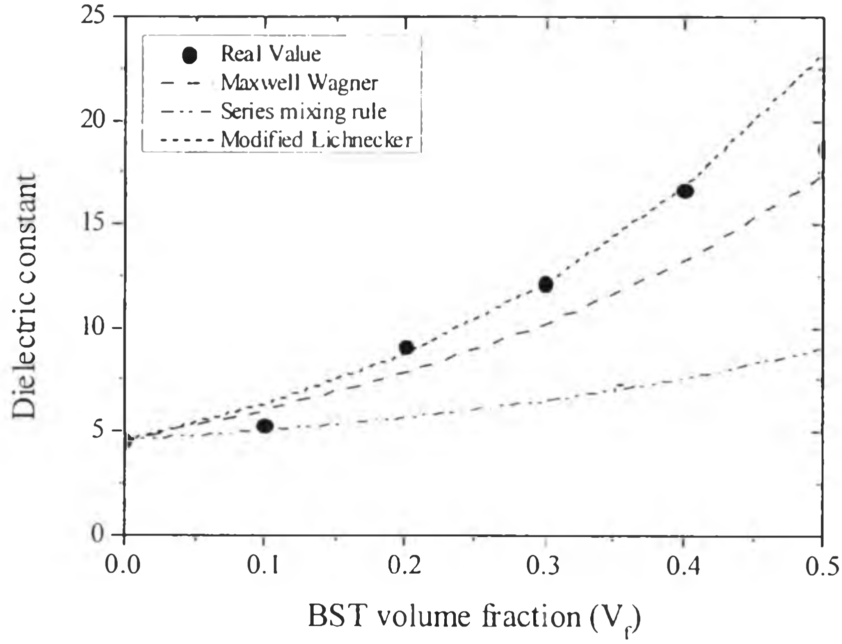

Figure E1 Dielectric constant of PBS/BST composites as a function of volume fraction of BST filler and experimental data fitting.

Appendix F Poly(butylene succinate)/modified - Barium Strontium Titanate Composite Thin-film Characterizations

Preparation of modified - Barium Strontium Titanate

The PBS-nanocomposite with the PBS:BST ratio of 90:10 were taken into further investigation on the effect of surface treatment in BST powder using Triethoxyvinylsilane coupling agent, Ethylene glycol and Propylene glycol as surface treatment agents. Firstly, barium strontium titanate powder were stirred in ethanol for 10 min at 60℃. Secondly, each surface treatment agent was slowly dropped for 10%(v/v) and taken to sonicated bath for 30 min. Then, the mixture were taken to centrifugal machine at 5000 rpm for 10 min to separate the surface treated powder and ethanol, dried in the heating oven for 24 hr. Finally, the surface treated BST powder were mixed with PBS pellets by using solution mixing method following by compression molding.

Morphological Investigation

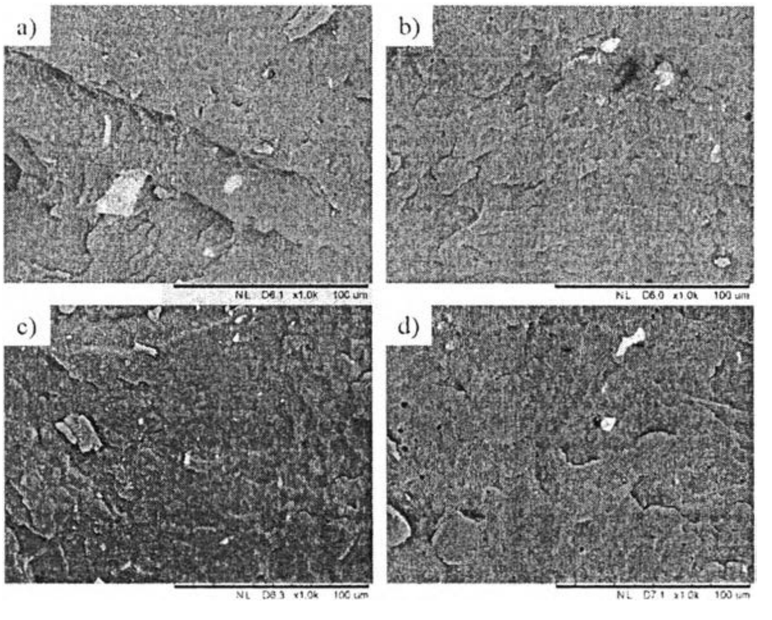

Figure F1 SEM image of surface treated BST/PBS composite a) non-treated b) Triethoxyvinyl silane-treated c) Ethylene glycol-treated d) Propylene glycol-treated.

Mechanical Properties

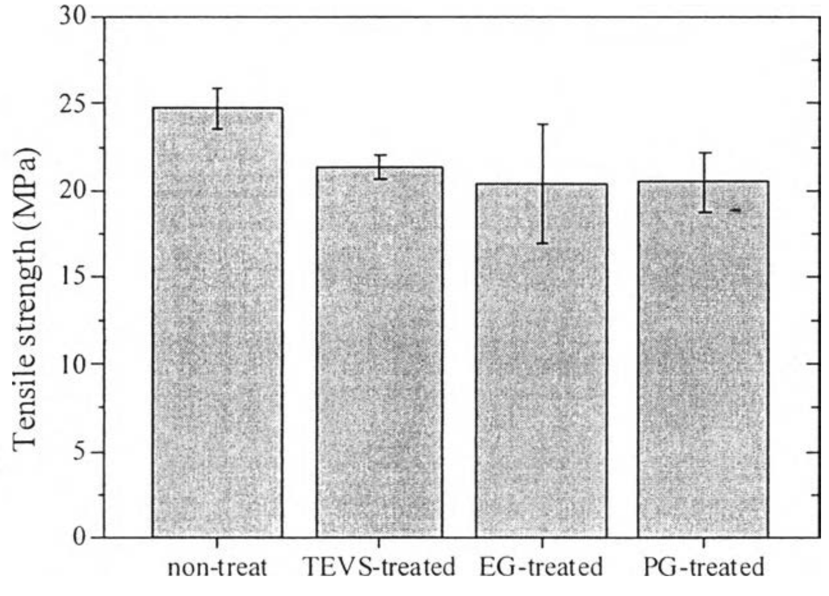

Figure F2 Tensile strength of surface treated BST/PBS composite.

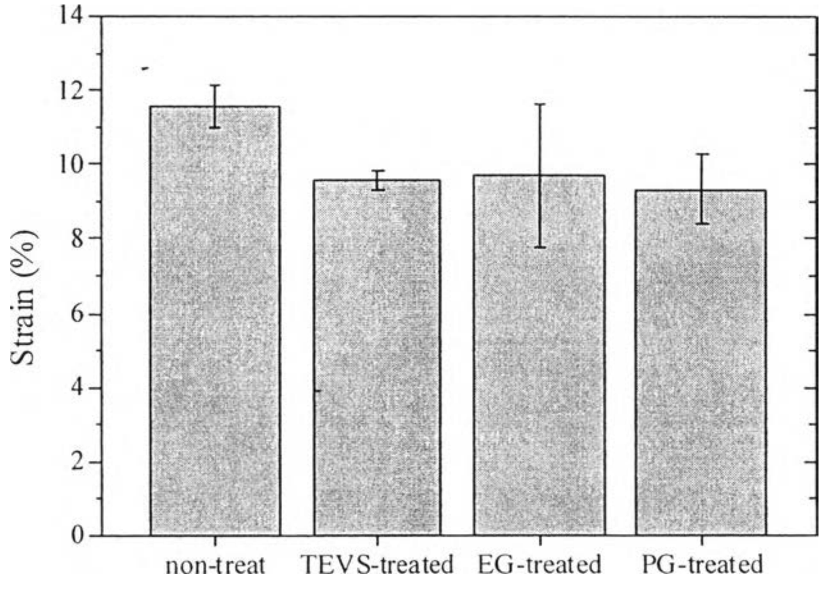

Figure F3 Strain at break of surface treated BST/PBS composite.

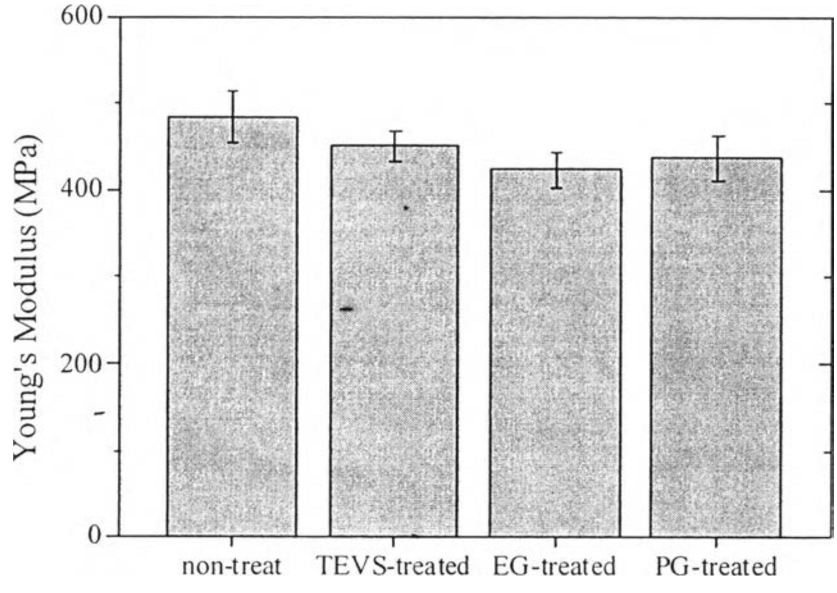

Figure F4 Tensile modulus of surface treated BST/PBS composite.

Frequency-dependent Dielectric Properties

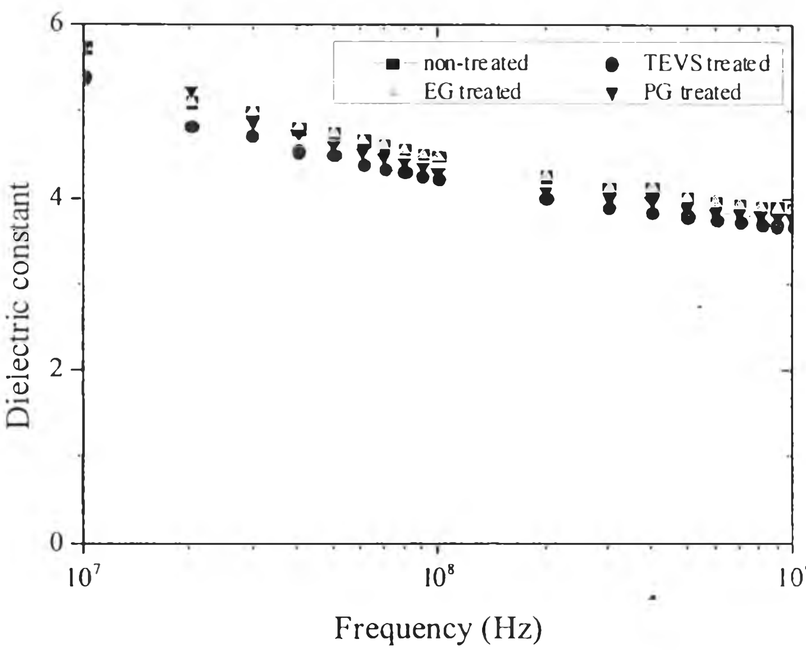

Figure F5 Dielectric constant of surface treated BST/PBS composite.

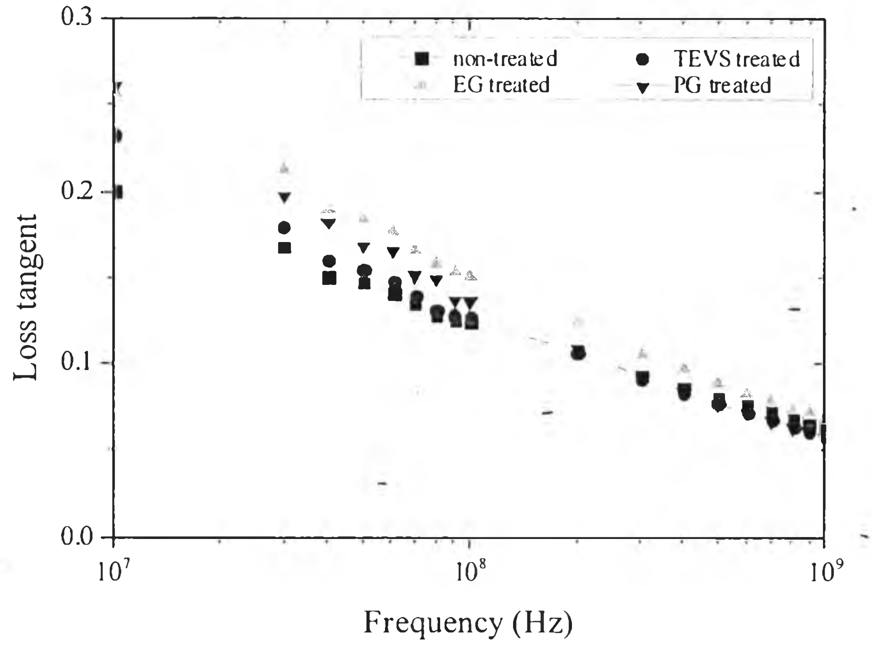

Figure F6 Loss tangent of surface treated BST/PBS composite.

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Presentations:

1. Kittichin Plungpongpan, Kulkeerati Koyanukkul, Attaphon Kaewvilai, Nollapan Nootsuwan, Prartana Keawsuwan, Apirat Laobuthee (2012, December 8) Preparation of PVP/MHEC Blended Hydrogels via Gamma Irradiation and their Calcium ion Uptaking and Releasing Ability. Poster presented at 10th Eco-Energy and Materials Science and Engineering Symposium, Ubolratchathanee, Thailand.
2. Kittichin Plungpongpan, Nollaphan Nootsuwan, Nattamon Koonsaeng, Apirat Laobuthee, Hathaikarn Manuspiya (2013, December 18-21) Magnesium Dopant Effect on Dielectric Permittivity of Mg-doped Barium Strontium Titanate ($Ba_{0.7-x}Sr_{0.3}Mg_xTiO_3$) Derived via Sol-Gel Process. Poster presented at 11th Eco-Energy and Materials Science and Engineering Symposium, Ubolratchathanee, Thailand.
3. Kittichin Plungpongpan, Nollaphan Nootsuwan, Nattamon Koonsaeng, Apirat Laobuthee, Hathaikarn Manuspiya (2014, April 22) Preparation and Characterization of Poly(Butylene Succinate) Thin-film Composite Incorporated with Barium Strontium Titanate Powder. Paper presented at the 5th Research Symposium on Petrochemical and Materials Technology and the 20th PPC Symposium on Petroleum, Petrochemicals and Polymers, Bangkok, Thailand.