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APPENDICES

Appendix A PVDF/BC Blend Analysis

The fraction of β phase, F(β), of the samples can be calculated using the followed equation;

$$F(\beta) = \frac{A_\beta}{1.26\,A_\alpha + A_\beta} \qquad (3.1)$$

Where A_α and A_β are absorbance of α and β phase in FTIR spectrum corresponding to wave number of 763 and 840 cm^{-1}, respectively.

Table A1 β-phase contents, F(β) (%) of PVDF and PVDF/BC blends

PVDF/BC	**A_α (763 cm^{-1})**	**A_β (840 cm^{-1})**	**F(β) (%)**
100/0	0.05321	0.16754	71.42
97.5/2.5	0.06774	0.17037	63.11
95/5	0.1165	0.26824	65.02
90/10	0.04606	0.09987	63.25
80/20	0.11269	0.22622	61.44
60/40	0.12514	0.16695	51.43

The crystallity of nanocomposite (X_c) was calculated by:

$$X_c = \frac{\Delta H_m}{\Delta H_o(1-\alpha)} \quad (3.4)$$

Where α is fiber weight content, ΔH_o is the melt enthalpy for 100% crystalline PVDF (102.7 J/g).

Table A2 DSC parameters of PVDF and PVDF/BC blends

Sample	ΔH_m(J/g)	T_g (°C)	T_m (°C)	X_c (%)
PVDF	41.65	-38.96	161.51	40.56
$PVDF_{97.5}BC_{2.5}$	40.09	-40.25	160.88	40.04
$PVDF_{95}BC_5$	37.38	-39.95	161.78	38.31
$PVDF_{90}BC_{10}$	33.10	-40.02	162.01	35.8
$PVDF_{80}BC_{20}$	33.87	-39.18	162.64	41.22
$PVDF_{60}BC_{40}$	29.03	-38.57	159.88	47.55

Table A3 Dielectric constant and dissipation factor of neat PVDF film at different temperature and frequency

Temperature (°C)	Frequency (MHz)					
	Dielectric Constant (ε')			Dissipation Factor (Tanδ)		
	10	100	1000	10	100	1000
-50	3.4270	3.2266	3.1628	.0355	.0161	0.083
-40	3.5271	3.2825	3.1861	.0489	.0238	.0129
-30	3.6967	3.3601	3.2173	.0676	.0356	.0192
-20	3.8750	3.4389	3.2484	.0878	.0484	.0263
-10	4.0963	3.5263	3.2800	.1163	.0627	.0334
0	4.5194	3.6821	3.3321	.1613	.0897	.0467
10	5.0787	3.8762	3.3876	.1956	.1226	.0625
20	5.8449	4.1463	3.4654	.2109	.1608	.0830
30	6.6026	4.4679	3.5525	.1993	.1955	.1056
40	7.2517	4.8617	3.6559	.1677	.2228	.1310
50	7.6544	5.2750	3.7670	.1312	.2355	.1564
60	7.9014	5.7194	3.8939	.1033	.2351	.1832
70	7.9780	6.0244	3.9936	.0864	.2279	.2011
80	8.0268	6.2745	4.0869	.0769	.2186	.2156
90	8.1037	6.4214	4.1496	.0714	.2130	.2242
100	8.0846	6.2943	4.0989	.0790	.2197	.2175

Table A4 Dielectric constant and dissipation factor of neat PVDF and PVDF/BC blend films as function of temperature at frequency of 10 MHz

Temp (°C)	PVDF/BC by weight									
	100/0	97.5/ 2.5	95/5	90/10	80/20	100/0	97.5/ 2.5	95/5	90/10	80/20
	Dielectric Constant (ε')					Dissipation Factor (Tanδ)				
-50	3.427	3.314	3.400	3.970	3.078	.0355	.0157	.0333	.0357	.0301
-40	3.527	3.458	3.500	4.101	3.155	.0489	.0320	.0419	.0442	.0424
-30	3.697	3.548	3.247	4.283	3.281	.0676	.0445	.0594	.0626	.0597
-20	3.875	3.786	3.877	4.531	3.440	.0878	.0727	.0901	.0835	.0813
-10	4.096	3.954	4.092	4.893	3.689	.1163	.0910	.1157	.1197	.1160
0	4.520	4.378	4.537	5.330	4.056	.1613	.1303	.1580	.1634	.1528
10	5.079	4.937	5.097	6.023	4.423	.1956	.1716	.1923	.1986	.1761
20	5.845	5.671	5.923	6.917	4.979	.2109	.1881	.2101	.2162	.1892
30	6.603	6.407	6.608	7.813	5.520	.1993	.1877	.2002	.2108	.1760
40	7.252	7.042	7.301	8.694	5.925	.1677	.1580	.1720	.1809	.1512
50	7.654	7.547	7.860	9.375	6.209	.1312	.1279	.1278	.1371	.1236
60	7.901	7.863	8.144	9.746	6.384	.1033	.0987	.0999	.1034	.0996
70	7.978	7.823	8.264	9.895	6.486	.0864	.0865	.0803	.0850	.0819
80	8.027	8.116	8.324	9.984	6.568	.0769	.0773	.0721	.0737	.0723
90	8.104	8.297	8.384	10.09	6.612	.0714	.0572	.0643	.0743	.0714
100	8.085	8.315	8.453	10.05	6.611	.0790	.0633	.0634	.0674	.0797

Table A5 Dielectric constant and dissipation factor of neat PVDF and PVDF/BC blend films as function of frequency at temperature of 20°C

	PVDF/BC	Frequency (MHz)						
		10	20	50	100	200	500	1000
Dielectric Constant (ε')	100/0	5.8449	5.1161	4.5005	4.1463	3.8737	3.6088	3.4654
	97.502.5	5.6707	4.9937	4.4096	4.0634	3.7865	3.5300	3.4044
	95/5	5.8671	5.2420	4.5091	4.1479	3.8941	3.6952	3.6440
	90/10	6.9169	6.0100	5.2874	4.8496	4.5671	4.3514	4.3229
	80/20	4.9794	4.3942	3.9242	3.6447	3.4255	3.2147	3.0962
Dissipation Factor (Tanδ)	100/0	.2109	.2032	.1892	.1608	.1383	.1042	.0830
	97.502.5	.1881	.1906	.1711	.1496	.1263	.0957	.0787
	95/5	.2101	.1995	.1853	.1491	.1172	.0730	.0527
	90/10	.2162	.2046	.1872	.1485	.1132	.0654	.0435
	80/20	.1892	.1797	.1697	.1441	.1239	.0942	.0751

Table A6 The P-E hysteresis loop parameters of PVDF and PVDF/BC blend films

Value	PVDF/BC (wt %)					
	100/0	97.5/2.5	95/5	90/10	80/20	60/40
P_{Max} (μC/cm^2)	0.0671	0.0789	0.0660	0.0778	0.06624	0.05244
P_r (μC/cm^2)	0.0024	0.0048	-0.0002	0.0041	0.00342	0.00119
$-P_r$ (μC/cm^2)	-0.0064	-0.0086	0.0012	-0.0098	-0.00645	-0.0029
V_c (V/m)	73.6429	233.9358	-92.1962	93.5028	84.2390	0
$-V_c$ (V/m)	-117.436	-176.247	5.3362	-146.95	-91.5324	-52.1359
C_{Max}-Eff (nF)	0.0355	0.02048	0.0244	0.0210	0.06294	0.0389
Offset (μC/cm^2)	0.0015	0.00642	-0.0015	0.00306	0.003084	-0.00132

Appendix B $PVDF_{90}BC_{10}$-MWCNT Blend Analysis

Table B1 β-phase contents, F(β) (%), of $PVDF_{90}BC_{10}$ with various MWCNT (phr) loading

MWCNT (phr)	A_{α} (763 cm^{-1})	A_{β} (840 cm^{-1})	F(β) (%)
0	0.04606	0.09987	63.25
1	0.14154	0.30213	60.58
2	0.27173	0.2884	45.71
3	0.15779	0.23521	54.19
4	0.08121	0.09565	48.31
5	0.16201	0.26793	56.76

Table B2 DSC parameters of $PVDF_{90}BC_{10}$ at various MWCNT (phr) loading

MWCNT (phr)	ΔH_m(J/g)	T_g (°C)	T_m (°C)	X_c (%)
0	33.10	-40.02	162.01	35.8
1	32.72	-38.55	160.70	35.4
2	30.86	-38.90	159.18	33.4
3	31.59	-39.13	158.92	34.2
4	34.17	-36.39	162.00	37.0
5	32.86	-37.90	161.95	35.6

*Heat of fusion value for 100% crystalline PVDF, ΔH_0= 102.7 J/g

The crystalline relaxation process on dielectric behaviour can be described using Arrhenius law as following equation;

$$\tau = \tau_o \exp\left(\frac{E_a}{kT}\right)$$

Where k is the Boltzmann's constant (8.314×10^{-15} eV/K), T is the maximum relaxation temperature, τ_o is the time constant.

$$\ln \tau = \ln \tau_o + \left(\frac{E_a}{kT}\right)$$

The E_a can be calculated from the slope of ln τ vs. 1/T and τ_o is an intersect with the vertical axis

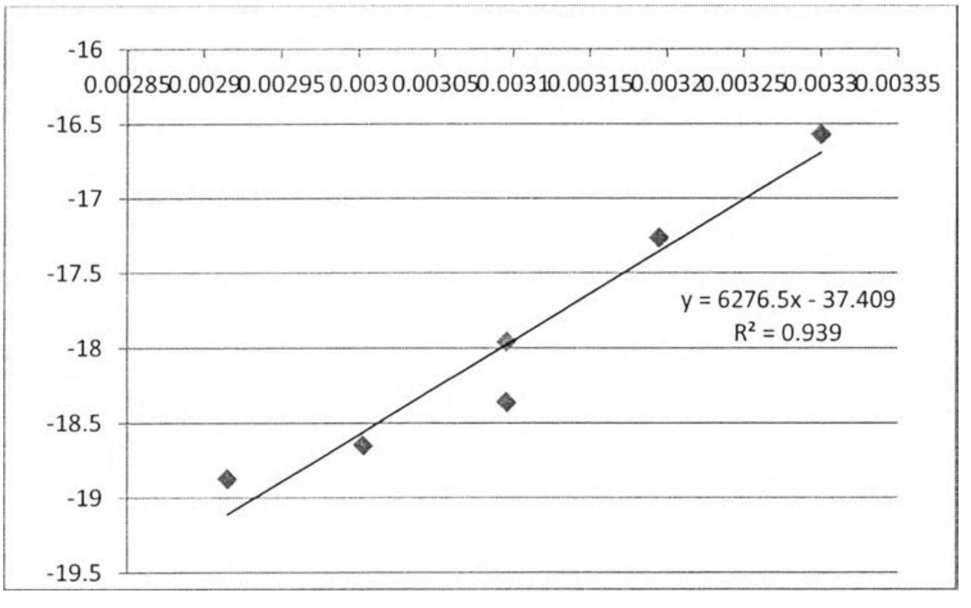

Figure B1 The frequency maximum versus reciprocal of temperature for $PVDF_{90}BC_{10}$ film.

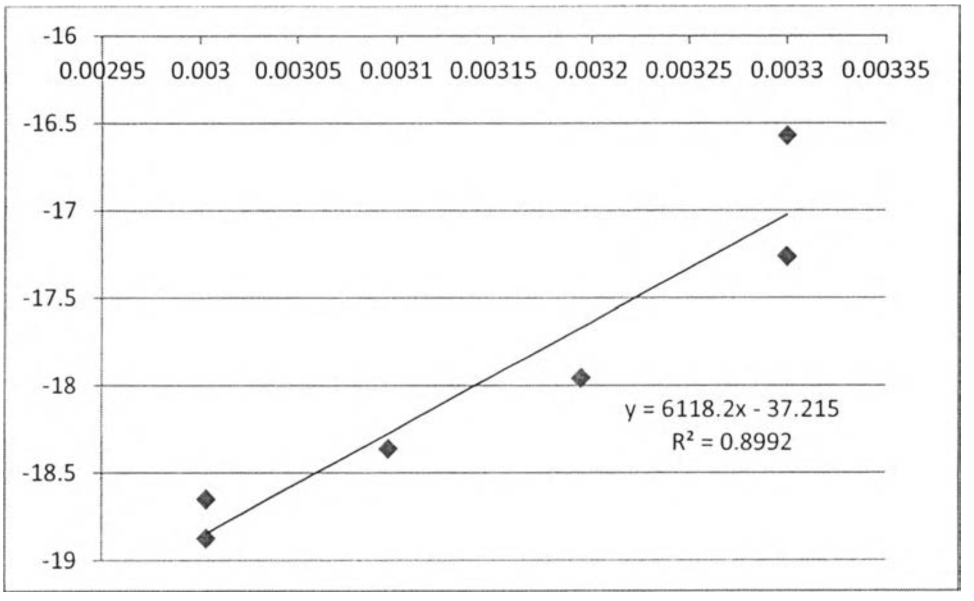

Figure B2 The frequency maximum versus reciprocal of temperature for $PVDF_{90}BC_{10}$-MWCNT 1 phr film.

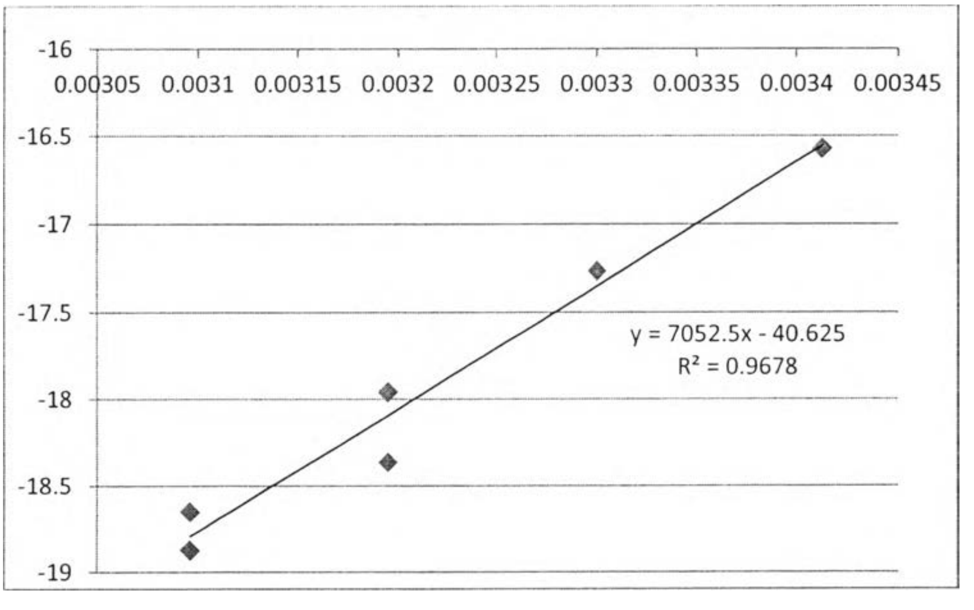

Figure B3 The frequency maximum versus reciprocal of temperature for $PVDF_{90}BC_{10}$-MWCNT 2 phr film.

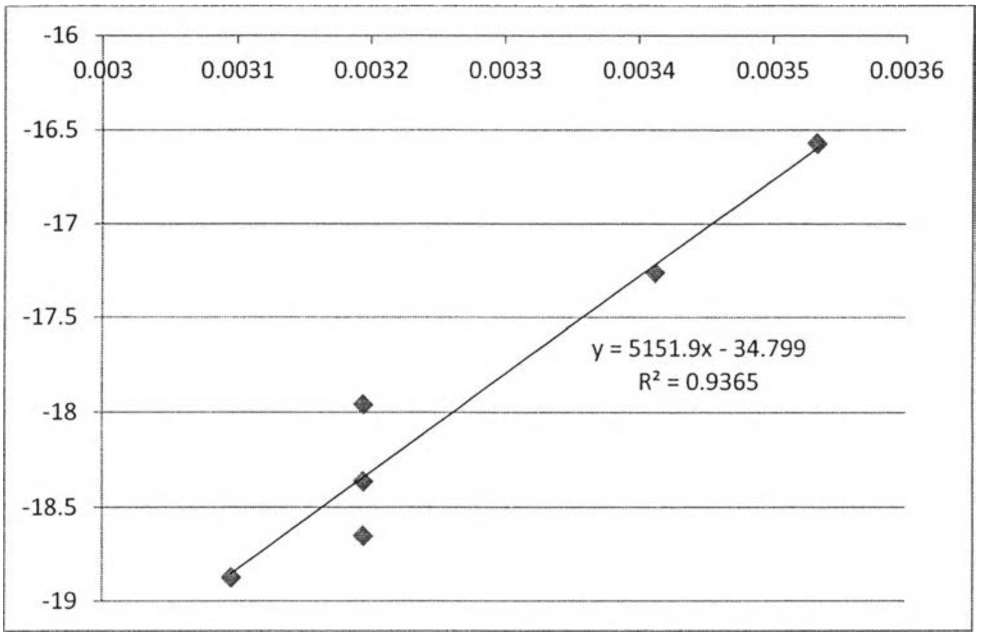

Figure B4 The frequency maximum versus reciprocal of temperature for $PVDF_{90}BC_{10}$-MWCNT 3 phr film.

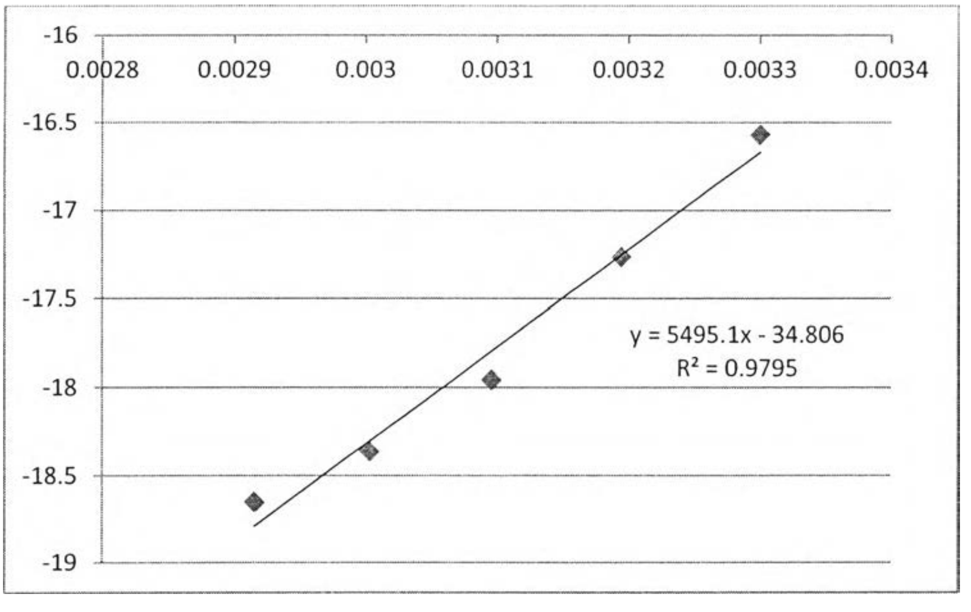

Figure B5 The frequency maximum versus reciprocal of temperature for $PVDF_{90}BC_{10}$-MWCNT 4 phr film.

Table B3 The P-E hysteresis loop parameter of $PVDF_{90}BC_{10}$ and $PVDF_{90}BC_{10}$-MWCNT blend films

Value	**MWCNT (phr)**					
	0	**1**	**2**	**3**	**4**	**5**
P_{Max} (μC/cm^2)	0.0671	0.0789	0.0660	0.0778	0.06624	0.05244
P_r (μC/cm^2)	0.0024	0.0048	-0.0002	0.0041	0.00342	0.00119
-P_r (μC/cm^2)	-0.0064	-0.0086	0.0012	-0.0098	-0.00645	-0.0029
E_c (V/m)	73.6429	233.9358	-92.1962	93.5028	84.2390	0
-E_c (V/m)	-117.436	-176.247	5.3362	-146.95	-91.5324	-52.1359
C_{Max}-Eff (nF)	0.0355	0.02048	0.0244	0.0210	0.06294	0.0389
Offset (μC/cm^2)	0.0015	0.00642	-0.0015	0.00306	0.003084	-0.00132

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Presentations:

1. O-Rak, K.; Ummartyotin, S.; Sain, M.; and Manuspiya, H. (2012, December 9-14) Direct Piezoelectricity of Multi-walled Carbon Nanotubes Filled Bacterial Cellulose Based Nanocomposites. Paper presented at The 8th Asian Meeting on Ferroelectrics (AMF-8), Pattaya, Thailand.
2. O-Rak, K.; Ummartyotin, S.; Sain, M.; and Manuspiya, H. (2013, April 24) Dielectric Behavior of Carboxyl Multi-walled Carbon Nanotube Filled Poly(vinylidene fluoride)/Bacterial Cellulose Blends. Paper presented at The 4th Research Symposium on Petrochemical and Materials Technology and The 19th PPC Symposium on Petroleum, Petrochemicals, and Polymer, Bangkok, Thailand.
3. O-Rak, K.; Ummartyotin, S.; Sain, M.; and Manuspiya, H. (2013, May 21-23) Preparation and Characterization of Transparent Nanocomposite Films of Bacterial Cellulose/Acrylic Resin. Paper presented at The 3rd International Symposium - Frontiers in Polymer Science 2013, Sitges, Spain.