Supporting Information

Structural revolution of PVDF crystallized on MWCNT film on a gradient temperature and its dielectric properties

Shuhui Li[a], Hai Wang[a], Xiaobin Liang [b*], Meijie Qu[a], Mingshuai Fan[a], Rui Zhang[a], Ken Nakajima [b], Yuezhen Bin[a*]

[a] Department of Polymer Science and Engineering, School of Chemical Engineering, Dalian University of Technology, Dalian 116024, PR China

[b] Department of Chemical Science and Engineering, School of Materials and Chemical Technology, Tokyo Institute of Technology, Tokyo 152-8552, Japan

[a]*E-mail: binyz@dlut.edu.cn

[b]*E-mail: liang.x.ac@m.titech.ac.jp

Table S1. The sample names under different preparation conditions

Nucleus / T_c (℃)	MWCNT	No nucleus
117	PCNT-117	PN-117
111	PCNT-111	PN-111
104	PCNT-104	PN-104
…	…	…
51	PCNT-51	PN-51
45	PCNT-45	PN-45

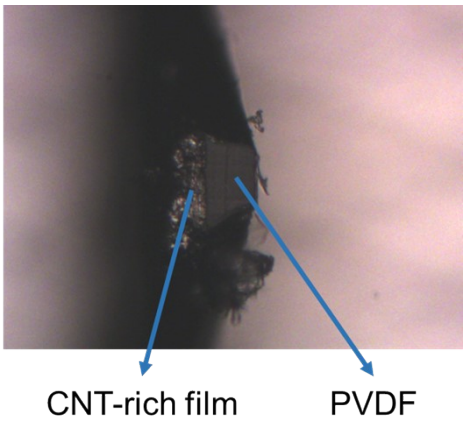

Figure S1. The samples' observation surface after slicing for AFM.

Table S2. The detail crystal characteristics of PN.

Samples	X_c/%	q/nm^{-1}	L/nm	L_c/nm	L_a/nm
PN-117	43.4	0.535	11.7	5.1	6.6
PN-111	44.4	0.553	11.4	5.0	6.3
PN-104	43.4	0.559	11.2	4.9	6.3
PN-98	43.2	0.588	10.7	4.6	6.1
PN-93	42.2	0.599	10.5	4.4	6.1
PN-87	41.1	0.626	10.0	4.1	5.9
PN-81	35.6	0.663	9.5	3.4	6.1
PN-71	28.7	0.79	8.0	2.3	5.7
PN-61	27.1	0.801	7.8	2.1	5.7

X_c is calculated by equation (2):

$$X_c = \sum A_{cryst} / (\sum A_{cryst} + \sum A_{amorp}) \quad (2)$$

where A_{cryst} and A_{amorp} are the peaks area of crystal and amorphous regions.

X_c can also be calculated from DSC by equation (3):

$$X_C = \frac{\Delta H}{\Delta H_0}$$

where X_c is crystallinity; ΔH is the enthalpy of melting of PVDF samples, expressed in

J/g; ΔH_0 is the standard enthalpy of melting of 100% crystallinity PVDF, expressed in 104.6 J/g.

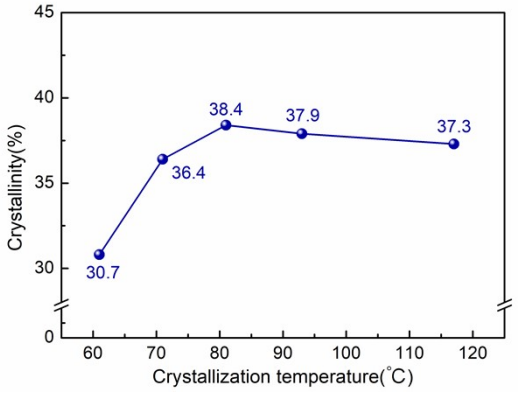

Figure S2. Crystallinity of PN crystallized at different T_c calculated from DSC.

The crystal structure was composed of lamellar regions and amorphous regions which was shown in Figure 5(e). The q value was calculated by equation (3):

$$q = 4\pi sin\theta/\lambda \tag{3}$$

where θ is the Bragg angle; λ is the wavelength of X-ray, λ=0.154 nm.

The L of PVDF is corresponding to q value, which is calculated by equation (4):

$$L = 2\pi/q \tag{4}$$

The L_c is calculated by equation (5):

$$L_c = L \cdot X_c \tag{5}$$

The L_a is caculated by equation (6):

$$L_a = L - L_c \tag{6}$$

where L is long period, L_c is lamellar thickness, L_a is amorphous thickness, q is scattering vector, X_c is crystallinity.

Table S3. The calculated peak area ratio of γ (021) plane.

T_c (℃)	117	111	104	98	93	87	81	71	61
PN	1.7%	1.0%	1.6%	2.7%	3.6%	4.6%	8.5%	14.2%	15.4%
PCNT	7.0%	7.4%	8.8%	10.6%	9.0%	9.4%	8..9%	14.2%	17.1%

Table S4. The detail crystal characteristics of PCNT.

Samples	X_c/%	q/nm^{-1}	L/nm	L_c/nm	L_a/nm
PCNT-117	44.8	0.544	11.5	5.2	6.4
PCNT-111	45.2	0.580	10.8	4.9	5.9
PCNT-104	43.7	0.604	10.4	4.6	5.9
PCNT-98	41.5	0.613	10.2	4.3	6.0
PCNT-93	40.6	0.646	9.7	3.9	5.8
PCNT-87	38.5	0.665	9.5	3.6	5.8
PCNT-81	38.4	0.660	9.5	3.7	5.9
PCNT-71	31.7	0.778	8.1	2.6	5.5
PCNT-61	28.3	0.711	8.8	2.5	6.3

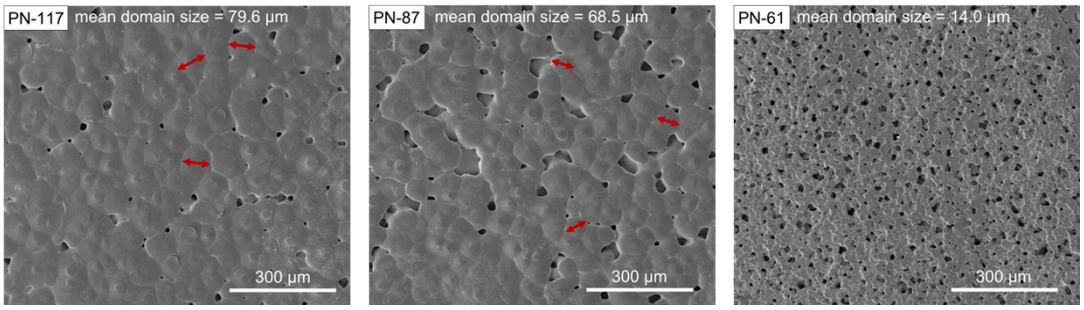

Figure S3. SEM of PVDF films which was crystallized at 117 ℃, 87 ℃ and 61 ℃.