Supplementary Information

Optimizing Dicyandiamide Pretreatment Conditions for Enhanced Structure and Electronic Properties of Polymeric Graphitic Carbon Nitride

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Table S1. Diffraction plane (002) and the corresponding d spacing value of various samples.		
Sample	**2 theta (002)**	**d spacing (nm)**
TP	27.60º	0.32
MP	27.54º	0.32
Stirr	27.34º	0.33
Probe	27.5º	0.32
SE	27.54º	0.32
FD	27.60º	0.32

Table S2. Solid yield (SY) of value of various pre-treated samples in comparison with TP		
Sample name	**Weight obtained (mg)**	**Yield percentage**
TP	567.7	56.77
MP	539.08	53.9
Stirr	568.21	56.82
Probe	567.3	56.73
SE	562.72	56.27
FD	534.08	53.4

Table S3. XPS binding energy positions for each deconvoluted C 1s spectra			
Sample	**C-C**	**CN_3/N-C=N**	**π–π* excitations**
TP	284.8	288.2	294.0
MP	284.8	288.1	293.8
Stirr	284.8	288.2	293.8
Probe	284.8	288.2	293.9
SE	284.8	288.1	293.7
FD	284.8	288.1	293.9

Table S4. Deconvoluted XPS peak assignments, binding energies (eV), FWHM (eV), and fitted peak area (%) of N 1s				
Sample	**Assignments**	**Binding energy (eV)**	**FWHM (eV)**	**Fitted peak area (%)**
TP	C-N=C	398.54	1.59	25
	NH_2	399.6	1.22	19
	NH	400.38	1.43	22
	NC_3	401.16	2.14	34
MP	C-N=C	398.50	1.80	32
	NH_2	399.61	1.09	19
	NH	400.37	1.18	21
	NC_3	401.11	1.55	28
	C-N=C	398.42	1.67	31
	NH_2	399.65	1.13	21

Stirr	NH	400.36	1.16	21
	NC_3	401.11	1.49	27
Probe	C-N=C	398.55	1.77	32
	NH_2	399.60	1.01	19
	NH	400.33	1.03	19
	NC_3	401.10	1.66	30
SE	C-N=C	398.60	1.58	30
	NH_2	399.60	1.28	24
	NH	400.37	1.00	19
	NC_3	401.08	1.43	27
FD	C-N=C	398.59	1.41	26
	NH_2	399.61	1.23	23
	NH	400.40	1.13	21
	NC_3	401.18	1.66	30

Table S5. Elemental composition of the prepared samples obtained from XPS analysis.

Sample	**C 1s (at %)**	**N 1s (at %)**	**O 1s (at %)**	**C/N ratio (at %)**
TP	39.44	57.29	3.27	0.68
MP	42.68	56.11	1.21	0.76
Stirr	39.91	57.62	2.48	0.69
Probe	41.28	56.02	2.69	0.73
SE	40.39	57.85	1.76	0.69
FD	41.5	55.22	3.28	0.75

Table S6. PL emission spectra values of various prepared samples.			
Sample	**λ (nm)**	**FL intensity**	**FWHM**
TP	463	14369800	116.74
MP	460	13850700	113.48
Stirr	467	12409100	117.12
Probe	465	12942300	118.70
SE	463	12866900	116.32
FD	461	14640600	118.49

Table S7. The peak-to-peak separation (ΔE_p) values of various prepared samples.		
Sample	**ΔE_p (mV)**	**ΔE_p mean value (n=3)**
TP	178	173
MP	110	118
Probe	111	110
Stirr	114	110
RC	111	115
FD	110	110

Table S8. The list of crystalline C1-C8 and molecular M1-M4 systems analyzed in this study. For the C1-C8 crystals the corresponding supercell sizes, compositions and C/N ratios are provided together with the supercell dimensions and angles as optimized at PBE-D2 level. For M1-M4 molecular systems the supercell dimensions were kept frozen during atomistic optimizations. The indicated band gaps for C1-C8 and HOMO-LUMO gaps for M1-M4 systems have been determined at HSE06-D2 level.

System	Supercell Size	Supercell Composition	C/N	Lattice Distances (Å)	Lattice Angles (deg.)	Band Gap (eV)
C1	2x2x1	C96 N160 H96	0.600	a=14.492 b=17.159 c=13.214	α=90.000 β=99.095 γ=90.000	4.31
C2	1x1x2	C192 N296 H120	0.648	a=16.574 b=27.306 c=12.945	α=99.267 β=81.811 γ=94.206	3.33
C3	1x1x4	C96 N144 H48	0.667	a=12.701 b=16.645 c=17.616	α=90.000 β=135.210 γ=90.000	3.34
C4	1x1x2	C144 N208 H48	0.692	a=12.454 b=23.766 c=12.423	α=90.000 β=90.000 γ=90.067	2.85
C5	1x1x3	C144 N204 H36	0.706	a=22.282 b=12.922 c=18.732	α=76.845 β=79.692 γ=90.035	3.00

C6	1x1x2	C72 N100 H12	0.720	a=13.902 b=13.870 c=13.233	α=90.015 β=99.083 γ=60.095	2.98
C7	1x1x2	2 x (C18 N25 H3)[a)] 2x (C24 N32)	0.737	a=13.937 b=13.923 c=13.183	α=89.995 β=89.146 γ=59.972	2.86
C8	1x1x3	C144 N192	0.750	a=11.917 b=20.495 c=13.345	α=90.000 β=90.000 γ=90.000	2.78
M1		C6 N10 H6		a=30.000 b=30.000 c=30.000	α=90.000 β=90.000 γ=90.000	4.58
M2		C12 N19 H6		a=36.000 b=30.000 c=30.000	α=90.000 β=90.000 γ=90.000	3.98
M3		C18 N27 H9		a=40.000 b=40.000 c=40.000	α=90.000 β=90.000 γ=90.000	3.56
M4		C24 N37 H15		a=45.000 b=45.000 c=45.000	α=90.000 β=90.000 γ=90.000	3.71

[a)] Individual compositions of different layers are indicated in this case.

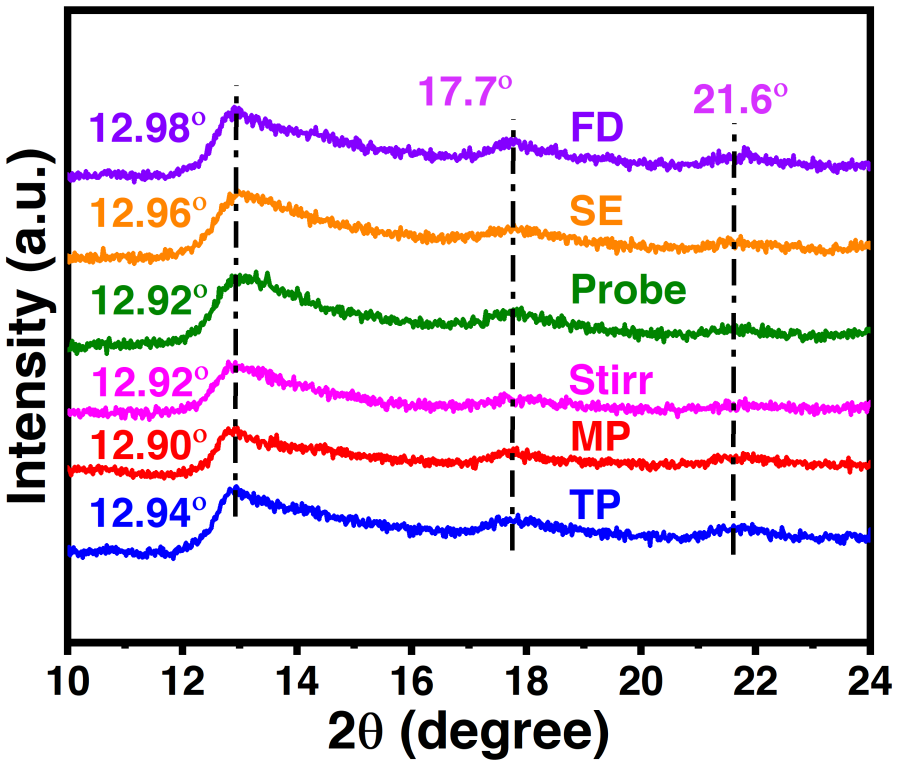

Figure S1. Enlarged XRD patterns between 10 º to 24º

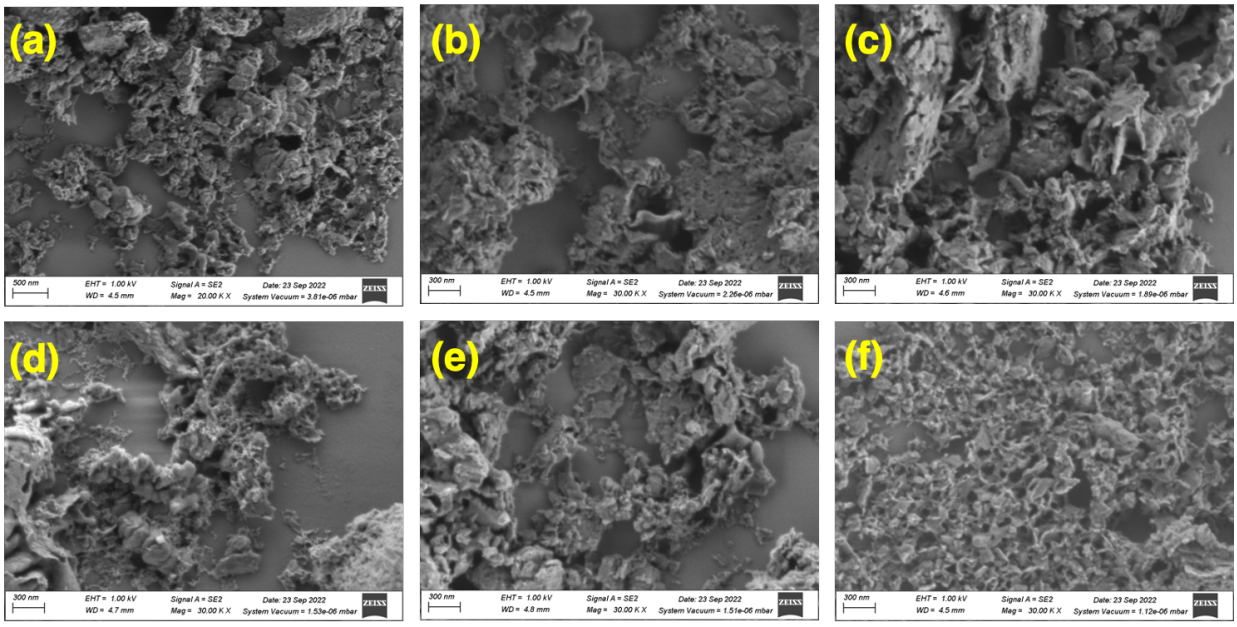

Figure S2. SEM images of the PgCN samples prepared using various methodologies (a) TP, (b) MP, (c), Stirr, (d) Probe, (e) SE, and (f) FD.

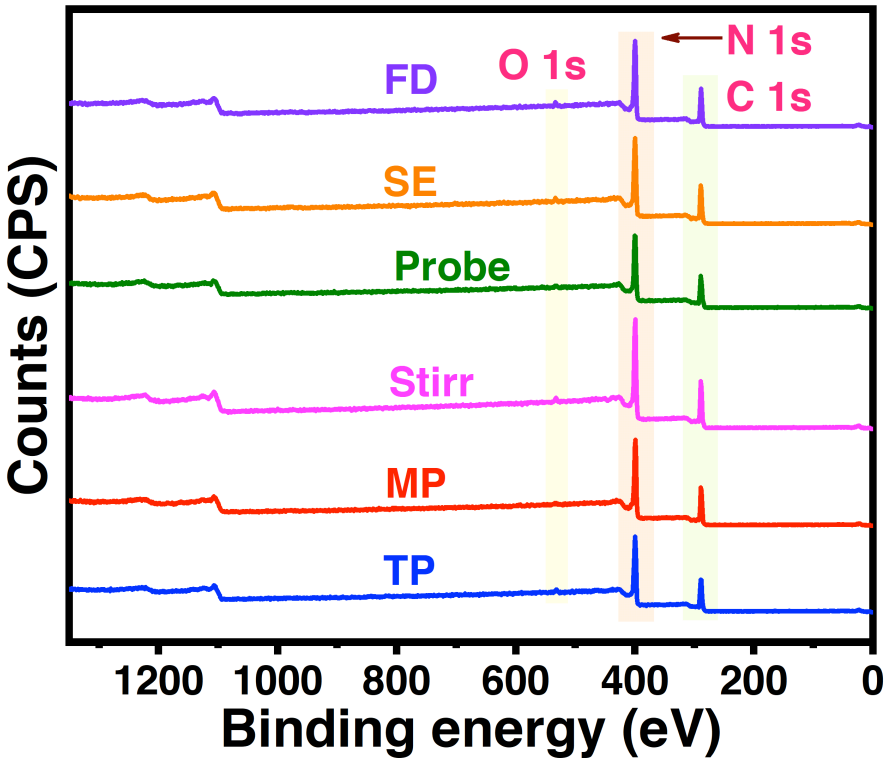

Figure S3. XPS survey spectra of various samples.