

Supplementary Materials

# Optimized Properties in Multifunctional Polyphenylene Sulfide Composites via Graphene Nanosheets/Boron Nitride Nanosheets Dual Segregated Structure under High Pressure

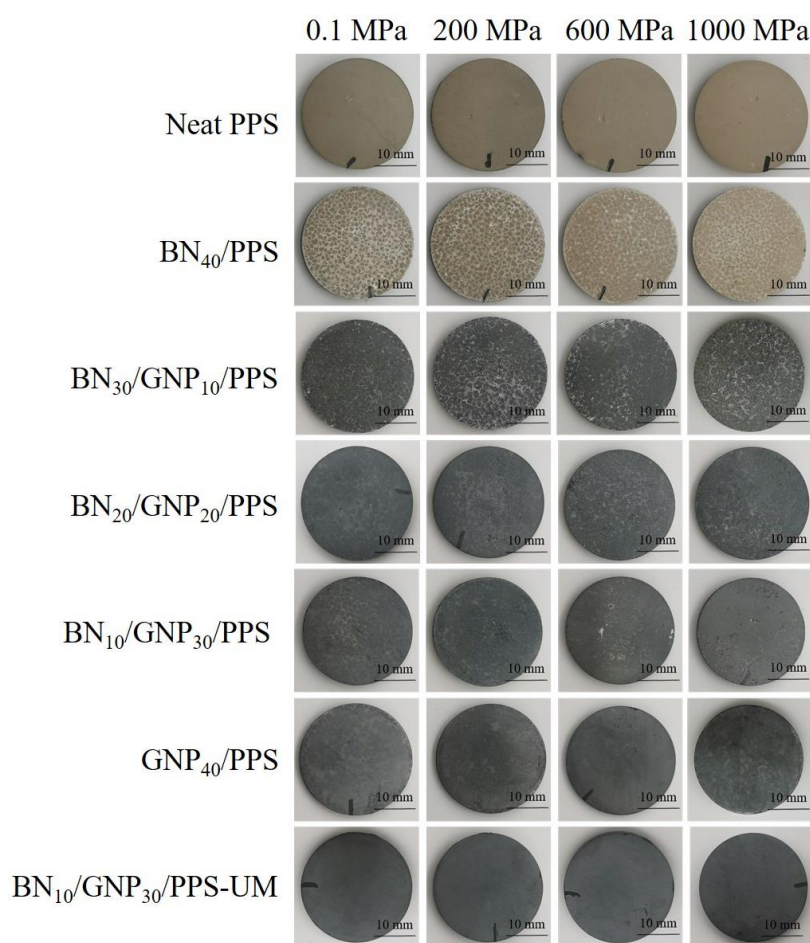
Liangqing Zhang <sup>1</sup>, Shugui Yang <sup>2</sup>, Longgui Peng <sup>1</sup>, Kepeng Zhong <sup>3</sup> and Yanhui Chen <sup>3,\*</sup>

<sup>1</sup> College of Material Science and Engineering, Xi'an University of Science and Technology, Xi'an 710054, China

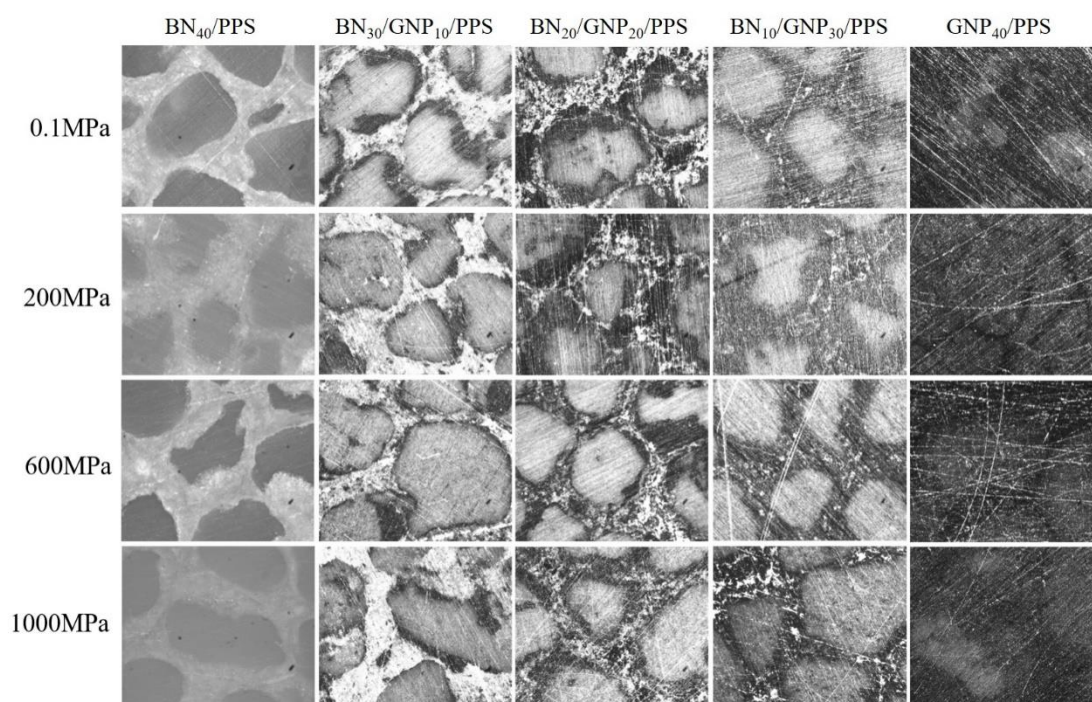
<sup>2</sup> Shaanxi International Research Center for Soft Matter, State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, China

<sup>3</sup> School of Chemistry and Chemical Engineering, Shaanxi Key Laboratory of Macromolecular Science and Technology, Key Laboratory of Special Functional and Smart Polymer Materials, Ministry of Industry and Information Technology, Northwestern Polytechnical University, Xi'an 710072, China

\* Correspondence: yanhuichen@nwpu.edu.cn; Tel.: +86-029-88431656



**Figure S1.** Digital images of cylindrical tablets of PPS composites hot pressed under different pressures.



**Figure S2.** OM images of PPS composites processed under different pressures.

**Table S1.** Integrated comparison of electrical conductivity and EMI SE among BN/GNP/PPS composites and other previous reported polymer composites with segregated structure.

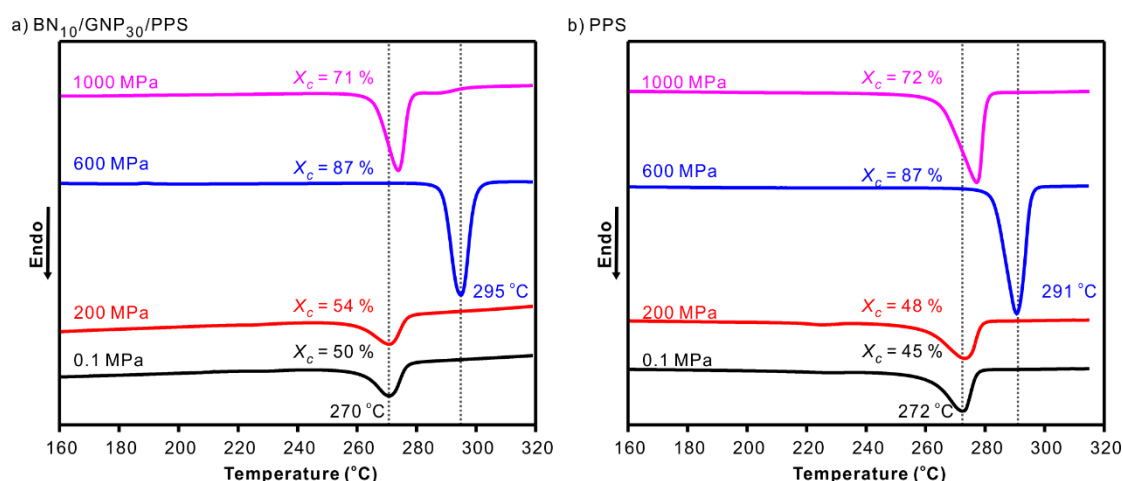
Composites	Filler content	Conductivity (S/m)	Thickness (mm)	EMI SE (dB)	Ref.
CNT/PLA	1.0 wt%	12	2.7	35.5	1
MWCNT/PS	7.0 wt%	61.9	1.8	42	2
Ni/UHWMPE	2.58 vol%	2648	2	55	3
EG/PA6	2.27 vol%	0.55	2	25	4
MWCNTs/epoxy	4 wt%	6.8	2	31.5	5
MWCNT/PS/SiAPP	7 wt%	--	3	11	6
MWCNT/PEBA	1.12 vol%	3.18E-5	1.6	16	7
PP/APP/CNT	7 wt%	37.8	2	32.1	8
CNT/PE	5 wt%	15	2.1	46.4	9
G-CNT/UHMWPE	15 wt%	195.3	2.1	81	10
MWCNT/PMMA	5 wt%	3.19	2	35.9	11
rGO/PMMA	2.6 vol%	91.2	2	63.2	12
Ag/CNT/PLLA/PCL	2.44 vol%	70	2	43.3	13
MWCNT/PLA	0.0054 vol%	6.3	5	45	14
MWCNT/PVDF	7 wt%	6	3	30.89	15
CNT/GO/PU	10 wt%	52	2	52.7	16
CNT/PU	7 wt%	20	2	41.2	17
CNT/PP	5.0 wt%	117	2.2	48.3	18
rGO/PS	3.47 vol%	43.5	2.5	45.1	19
rGO/UHMWPE	0.66 vol%	3.4	2.5	32.4	20
CNT/PVDF	4 wt%	74.5	2	36.8	21
CNT/UHMWPE	2 wt%	49.7	2	33.5	22
CNT/Epoxy	1 wt%	34.5	1.9	22	23

MWCNT/PLA	0.8 vol%	30	1.5	25	24
Ag/PLA	5.89 vol%	254	1.5	50	25
CNT/PPS	5.0 wt%	72	2	49.6	26
rGO/PMMA	2.6 vol%	91.2	2.9	63.2	12
GNP/BN/PPS	30 wt%	0.07	2	70	This work

**Table S2.** Integrated comparison of thermal conductivity and EMI SE among BN/GNP/PPS composites and other previous reported polymer composites.

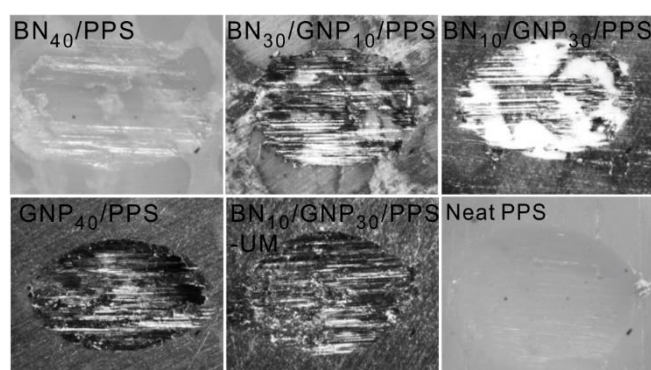
Composites	Filler content	Thermal conductivity (W/m/K)	EMI SE (dB)	Ref.
Cenospheres/ Fe <sub>3</sub> O <sub>4</sub> /MCMB/CNT/PAN	---	4.2	80.5	27
SiC/CNT/MCMB/PAN	---	6.5	67	28
GNP/PS	35 wt%	4.72	33	29
Mxene/PVA	19.5 wt%	4.57	44.4	30
NCCF/MWCNT/PU	---	0.032	24.7	31
MWCNT/POM	40 wt%	1.95	45.7	32
GNP/POM	48 wt%	4.24	44.7	32
Graphene/PDMS	18.1 wt%	3	86	33
Ag/CF/epoxy	7 wt%	2.49	38	34
GNP/rGO/epoxy	20.5 wt%	1.56	51	35
Ag/cellulose	50 wt%	10.55	101	36
Graphene/SiC/PVDF	9.5 wt%	2.13	32.5	37
Graphen/Ni/PVDF	20 wt%	8.96	43.3	38
Ag/TPU	15 vol%	4.45	105.3	39
MXene/BN/PDMS	---	0.65	35.2	40
MXene/PAT/PANI-PpAP	---	0.687	45.18	41
GNS/CINAP/CE	20 wt%	4.13	55	42
Carbon network/epoxy	7.0 vol%	0.58	27.8	43
AgNW/ Fe <sub>3</sub> O <sub>4</sub> /MF	---	0.034	49	44
CCA/rGO/PDMS	3.05 wt%	0.65	51	45
MWCNT/PLA	0.0054 vol%	0.03	50	14
MWCNT-Fe <sub>3</sub> O <sub>4</sub> @Ag	---	0.46	35	46
rGO-WPU/cotton	2 wt%	2.13	48.1	47
CNT@PDA/EVA	70 wt%	17.9	32.4	48
polyacrylate/graphene@polydo pamine	20 wt%	1.68	58	49
CuNWs-TAGA/epoxy	7.2 wt%	0.51	47	50
CNT/acrylic	0.62 vol%	0.008	36.7	51
Ni/PVDF	10 wt%	0.075	26.8	52
MXene/AgNW/CNF	---	15.5	55.9	53
CL@GC/CE	---	0.61	35.57	54
GNP/BN/PPS	40 wt%	6.838	70	This work
Graphene Fe-Ni		4.1	55	42
PC/SAN/G-NI		0.7	29.4	55
Carbon monolith/RGO		0.057	43	56

MXD6/EG/CNTs	2.1	50	57
WCM/N-G/ AgNWs	0.14	60	58
Graphene/SBR matrix	2.9	45	59
Titanium foam	0.8	45	60
PVDF/GNP-Ni	0.66	57.3	61



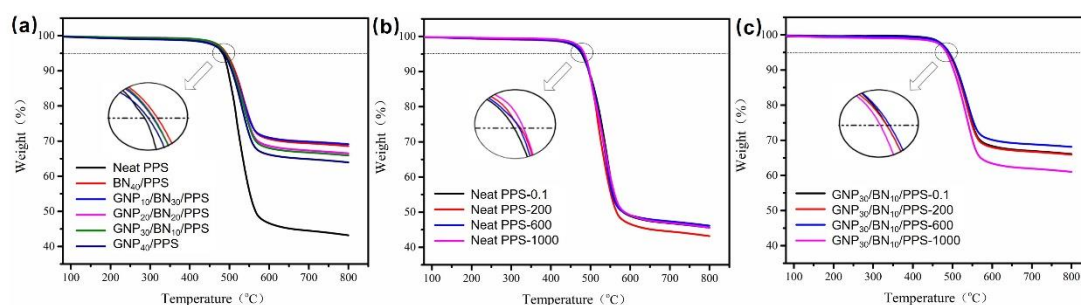
**Figure S3.** DSC heating curves of BN<sub>10</sub>/GNP<sub>30</sub>/PPS with segregated structures (a) and neat PPS (b) processed under different pressures.

The DSC heating curves of BN<sub>10</sub>/GNP<sub>30</sub>/PPS with segregated structures and neat PPS are presented in Figure S3. Obviously, both PPS composites and neat PPS display a much larger melting peak when they were crystallized under 600 MPa and 1000 MPa. The crystallinity of both PPS composites and neat PPS increases from ~50% of processed under 0.1 MPa and 200 MPa to ~80% of processed under 600 MPa and 1000 MPa, owing to the high pressure (600 MPa and 1000 MPa) accelerates the crystallization of PPS. Additional intriguing phenomenon observed here is the PPS samples crystallized at 600 MPa show an increased melting temperature (~295 °C) comparing with that processed under other pressures (~270 °C). This is due to the formation of extended-chain crystals in PPS matrix, which would be discussed in the forthcoming paper.



**Figure S4.** OM observations of worn surfaces of PPS composites and neat PPS after the test.

Figure S4 show OM images of the wear surfaces. In general, groove marks, along with surface lumps and detached areas on the wear surface are clearly visible, suggesting that the dominant wear mechanism are abrasion by surface asperities on the pin, and adhesion in the contact area.



**Figure S5.** TGA curves of Neat PPS and GNP/BN/PPS composites at 200 MPa (a); TGA curves of Neat PPS (b) and GNP<sub>30</sub>/BN<sub>10</sub>/PPS composites (c) under different molding pressures.

**Table S3.** The compressive strength of neat PPS and PPS composites processed under different pressures (MPa).

Samples	0.1 MPa	200 MPa	600 MPa	1000 MPa
Neat PPS	163.5	165.8	163.5	164.7
BN <sub>40</sub> /PPS	107.4	110.9	93.4	95.7
GNP <sub>S10</sub> /BN <sub>30</sub> /PPS	105.1	100.4	93.4	95.8
GNP <sub>S20</sub> /BN <sub>20</sub> /PPS	116.8	116.8	110.9	105.1
GNP <sub>S30</sub> /BN <sub>10</sub> /PPS	140.1	133.1	129.6	128.5
GNP <sub>40</sub> /PPS	146.0	141.3	137.8	134.3
GNP <sub>30</sub> /BN <sub>10</sub> /PPS-UM	156.5	156.5	153.0	154.2

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