Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Achieving Long-Term Anticorrosion via the Inhibition of Graphene's Electrical Activity

Jiheng Ding[§], Hongran Zhao[§], Dong Ji, Beiyu Xu, Haibin Yu*

Key Laboratory of Marine Materials and Related Technologies, Zhejiang Key Laboratory of Marine Materials and Protective Technologies, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, P. R. China.

*Corresponding author. E-mail: dingjh@nimte.ac.cn (Jiheng Ding), haibinyu@nimte.ac.cn (Haibin Yu)

[§] Jiheng Ding and Hongran Zhao contributed equally to this work.

Table S1. Chemical composition (wt %) of the Q235 used.

Element	**B**	**C**	**O**	**Si**	**Cr**	**Mn**	**Fe**
Weight%	0.56	5.60	0.45	0.26	0.02	0.43	Bal.

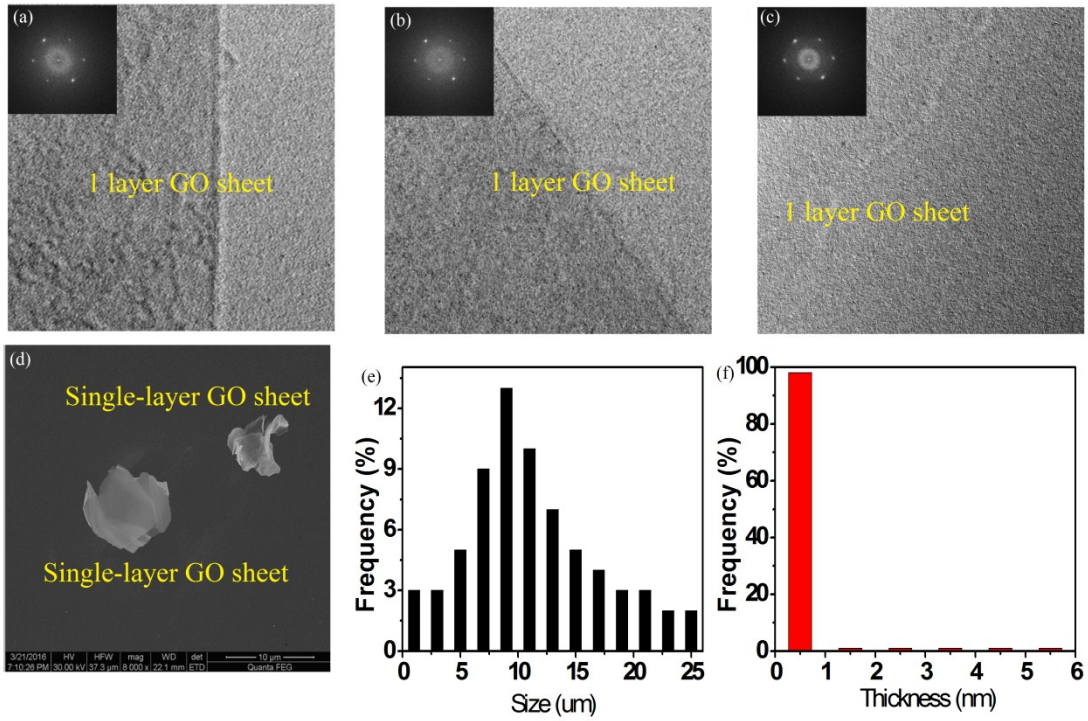

Figure S1. High-resolution TEM images of GO sheets (a-b); SEM images of GO sheets (d); The size (e) and thickness (f) distributions of the GO sheets.

The single-layered feature of the GO sheets was confirmed by high-resolution transmission electron microscopy (HR-TEM) and scanning electron microscopy (SEM) images as shown in Fig.S1. Under the HR-TEM inspection (Fig. S1a-c), the GO sheets exhibit typical single-layer structures and the selected area electron diffraction patterns (corresponding insets) further reveal their single-layer nature. In SEM images (Fig. S1d), the GO sheets exhibit typical wrinkles, demonstrating their fine flexibility. Based on the statistics from the SEM and TEM images, where a statistical analysis of over 100 flakes shows that > 99 % of the sheets are single layer (<1 nm in thickness) with a lateral size of 8±0.5μm (Fig. S1e, f). These results reveal that large-size and few-layered GO was successfully prepared in our work.

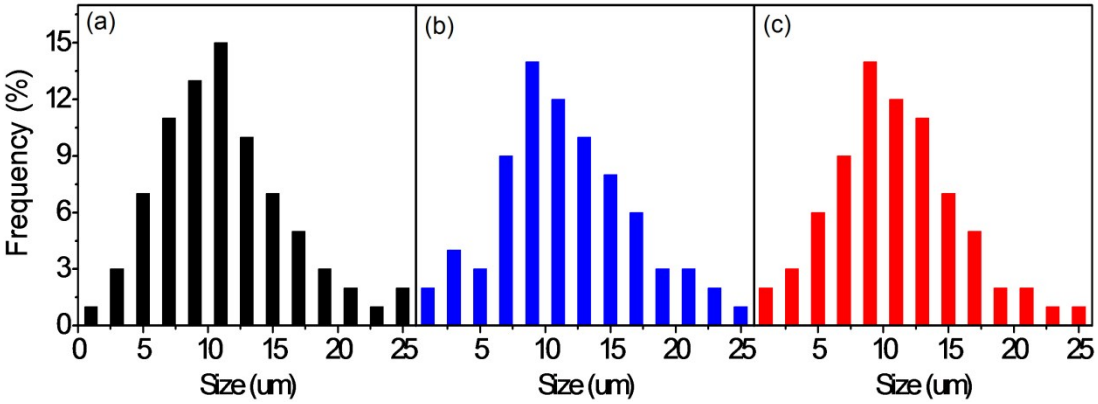

Figure S2. The size distributions of the G (a), BG (b) and NG (c) nanosheets, respectively.

Table S2. Relative percentages of the elemental composition of the doped graphene materials.

Sample	C	O	H	N	B	S
G	97.32	1.94	0.74	0	0	0
BG	94.71	2.14	1.28	0	1.87	0
NG	93.19	2.22	2.13	2.46	0	0

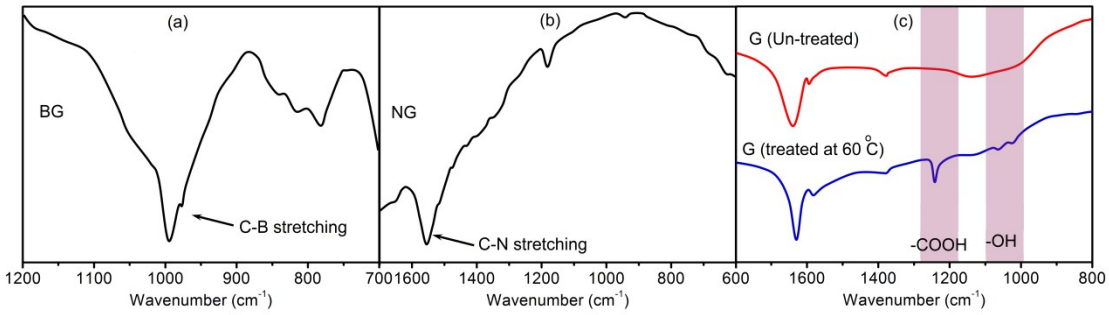

Figure S3. FTIR spectroscopy of BG (a), NG (b), and G sheets treated at 60 ^{o}C.

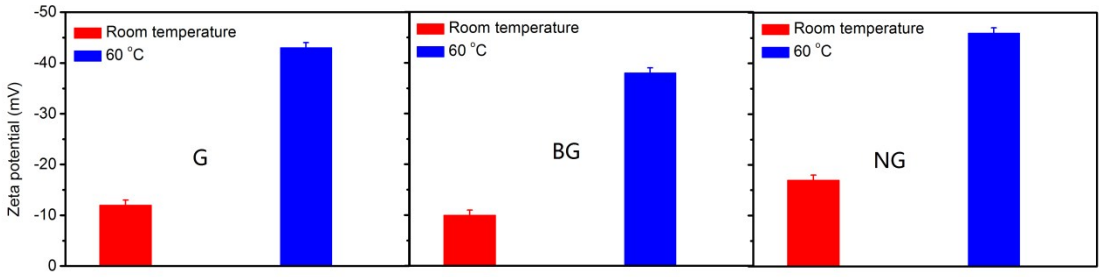

Figure S4. Zeta potential measurement for low and high temperature sonicated graphene-based materials.

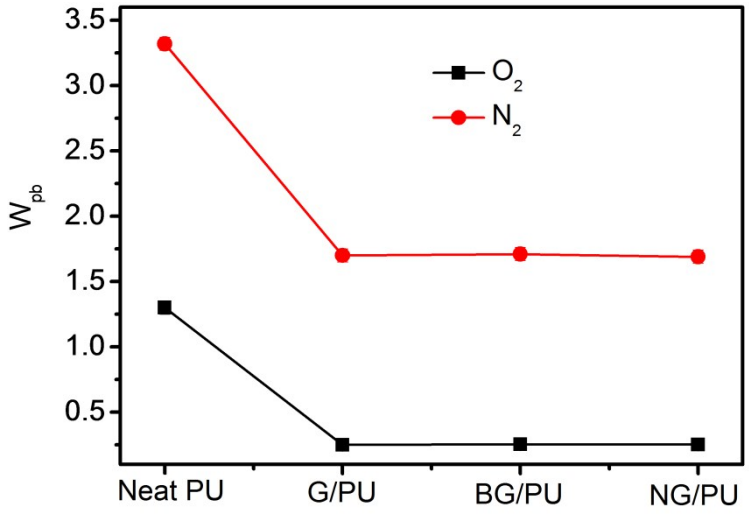

Figure S5. The gas barrier property results for the neat PU and their nanocomposite coating systems.

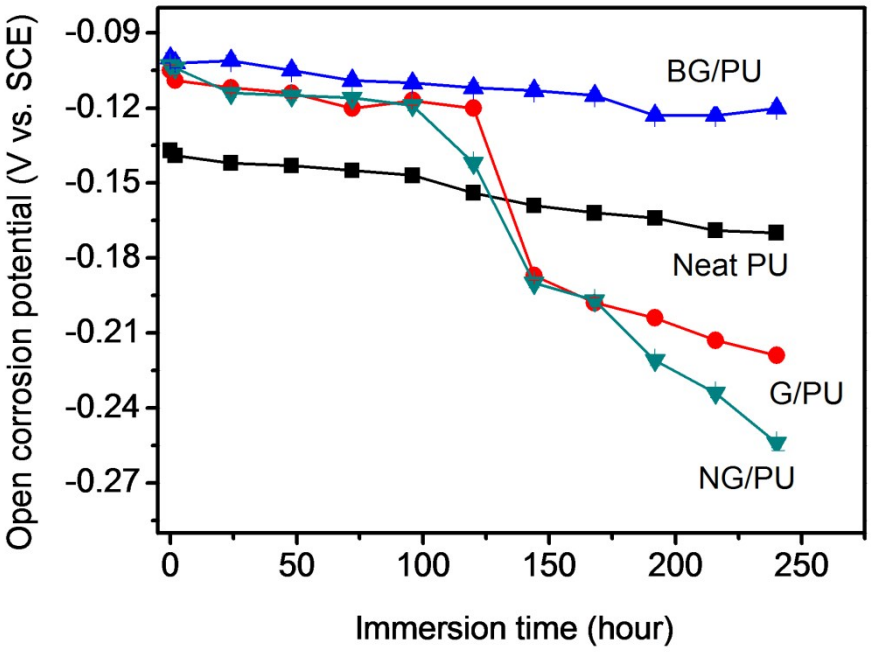

Figure S6. Open corrosion potential of neat PU, G/PU, BG/PU, and NG/PU nanocomposite coatings during 240 h immersion in 3.5 wt.% $NaCl_{(aq)}$.

Table S3. The impedance modulus at the lowest frequency ($Z_{f=0.01\ Hz}$) for different coatings with different immersion time.

Sample	$Z_{f=0.01\ Hz}$ ($\Omega\ cm^2$)										
	2h	24h	48h	72h	96h	120h	144h	168h	192h	216h	240h
neat PU	6.05×10^9	4.46×10^9	3.93×10^9	2.16×10^9	1.24×10^9	7.33×10^8	5.01×10^8	4.08×10^8	6.38×10^7	3.89×10^7	9.25×10^6
G/PU	8.92×10^9	8.15×10^9	4.76×10^9	4.21×10^9	4.25×10^9	2.91×10^9	1.15×10^9	5.88×10^6	4.05×10^6	5.25×10^6	5.51×10^6
BG/PU	1.29×10^{10}	9.96×10^9	9.58×10^9	9.27×10^9	8.98×10^9	8.80×10^9	8.87×10^9	7.20×10^9	6.69×10^9	6.92×10^9	6.67×10^9
NG/PU	7.2×10^9	418×10^9	3.5×10^9	1.85×10^9	8.97×10^8	7.74×10^8	6.97×10^8	3.85×10^6	3.64×10^6	2.53×10^6	2.15×10^6

Table S4.The fitting R_c results of the collected EIS results of different coatings as a function of immersion time.

Sample	R_c (Ω cm^2)										
	2h	24h	48h	72h	96h	120h	144h	168h	192h	216h	240h
neat PU	2.29×10^8	1.01×10^8	7.19×10^7	7.07×10^7	5.89×10^7	3.25×10^7	1.01×10^7	6.88×10^6	3.32×10^6	9.33×10^5	5.12×10^5
G/PU	3.35×10^9	1.73×10^9	6.54×10^8	5.27×10^8	3.17×10^8	4.22×10^7	4.76×10^5	3.13×10^5	3.17×10^5	2.15×10^5	2.55×10^5
BG/PU	6.82×10^9	4.55×10^9	2.67×10^9	1.86×10^9	2.96×10^9	1.03×10^9	9.21×10^8	7.34×10^8	5.65×10^8	4.76×10^8	2.54×10^8
NG/PU	3.62×10^9	1.02×10^9	5.43×10^8	4.23×10^8	2.83×10^8	1.54×10^7	4.43×10^5	2.76×10^5	2.96×10^5	2.07×10^5	2.13×10^5

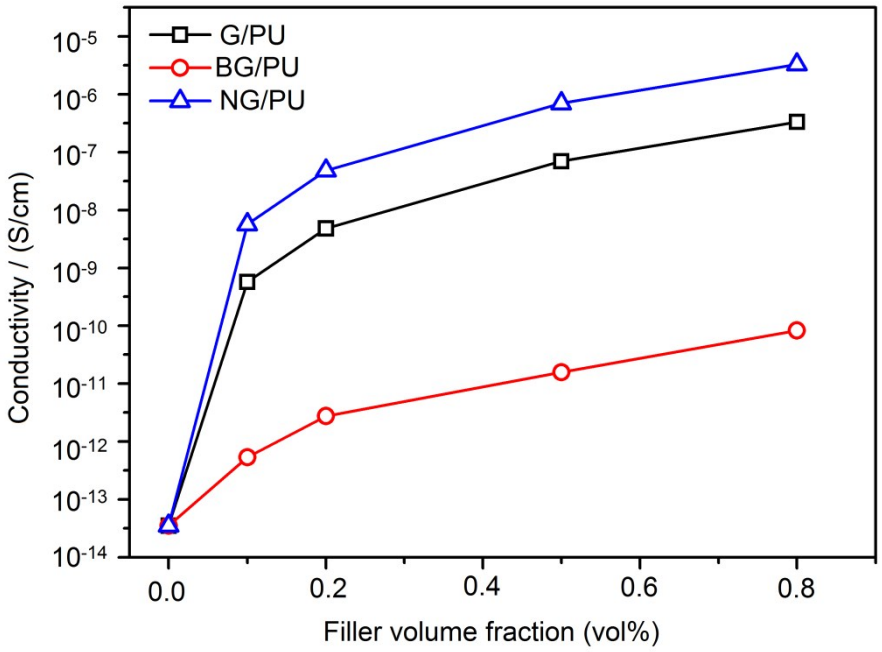

Figure S7. Electrical conductivity of the PU nanocomposits coatings.

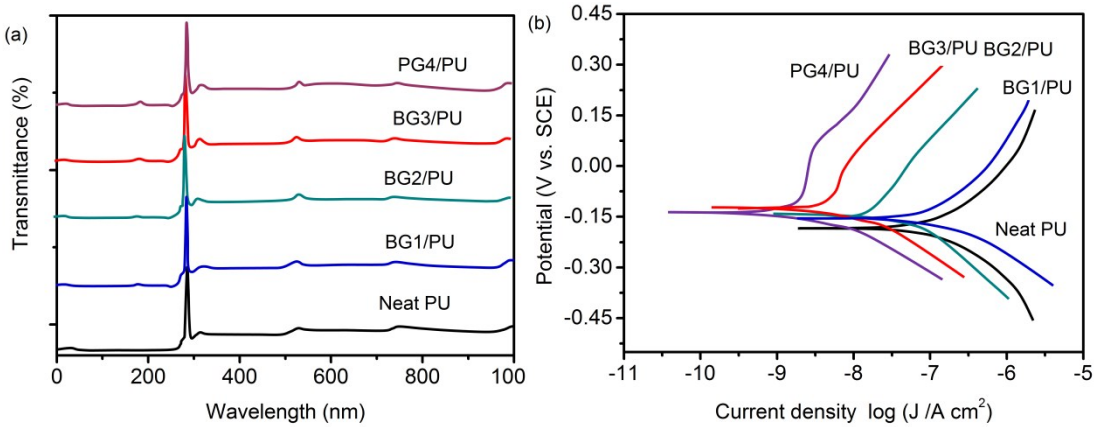

Figure S8. The XPS spectra and Polarization curves of neat PU and their nanocomposite coatings with different B doping concentration graphene.

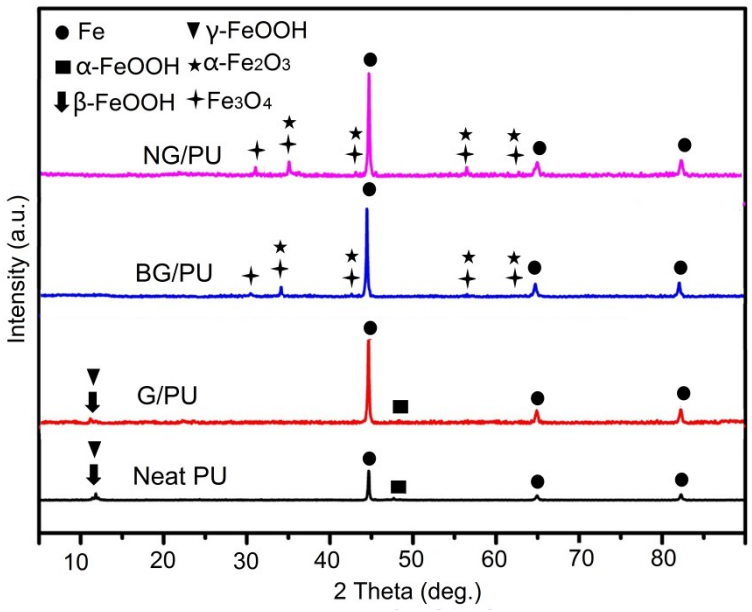

Figure S9. XRD patterns spectra for rust regions on the steel beneath the neat PU and their nanocomposite coatings.