

Supporting Information

Effect of KOH on the Energy Storage Performance of Molasses-based Phosphorus and Nitrogen Co-doped Carbon

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Synthesis of Phosphorus and Nitrogen Co-doped composites:

Prior to microwave carbonization, molasses and ammonium polyphosphate were weighed in the amounts mentioned in Table S1 and added with addition of 100 μ L of deionized water into a boron nitride crucible (and cover) and hand-stirred into visible homogenization. Once mixed, the crucible containing pre-carbonized solution was covered with another crucible placed into an aluminum oxide box with cover. Precursor materials were microwaved for 30 minutes in a conventional, high-powered, microwave oven. Product was then left to cool to room temperature, at which point it was removed from the oven, powdered with mortar and pestle, and utilized for electrochemical studies and characterization.

Table S1. Composition of PNDC1 and PNDC2 prepared prior to carbonization.

| Material | Amt. Molasses (g) | Amt. APP (g) | Amt. KOH (mL) | PNDC Yield (g) |
|----------|-------------------|--------------|---------------|----------------|
| PNDC1 | 1 | 0.6 | 0 | 0.165 |
| PNDC2 | 1 | 0.6 | 0.1 | 0.28 |

X-ray Diffraction (XRD)

The morphology for PNDC-2 was confirmed via XRD. The resulting broad and fuzzy peaks demonstrates the amorphous nature of this material, as shown by Figure S2. Furthermore, use of Bragg's law demonstrates to us that the multilayered PNDC-2 is very widely spaced out. Bragg's law demonstrates the relationship between x-ray diffraction and reflection off a presumably crystalline structure. The relationship is shown by the following equation:

$$\lambda = 2d\sin\Theta \quad (2)$$

When substituting in appropriate values to the following equation, λ corresponds to wavelength of the x-ray beam used for analysis for which $\lambda = 0.15$ nm. Θ (theta) is the diffraction angle between the x-ray beam and XRD detector. Solving for 'd', where 'd' corresponds to the separation length of multilayered planes, the d-spacing value is determined to be 0.44 nm separation between co-doped layers of PNDC2. Compared to the d-spacing value of pure graphitic carbon of 0.34 nm, this difference is mainly due to

presence of matrix-incorporated doping atoms and functional groups, further supportive of carbon doping [1,2].

The additional peaks in XRD spectra indicate carbon when chemically activated/functionalized with an activating agent such as KOH. The region that the sharp peaks are shown support highly disordered carbon along with the broad peak that encompasses them, indicating amorphousness as mentioned in line 136. The sharpness of the peaks shown in the XRD spectra are indicative of some crystallite graphene-like properties being present, but surrounded by other layers of graphene-like carbon that are still poorly formed, thus the broad, staticky, larger peaks.

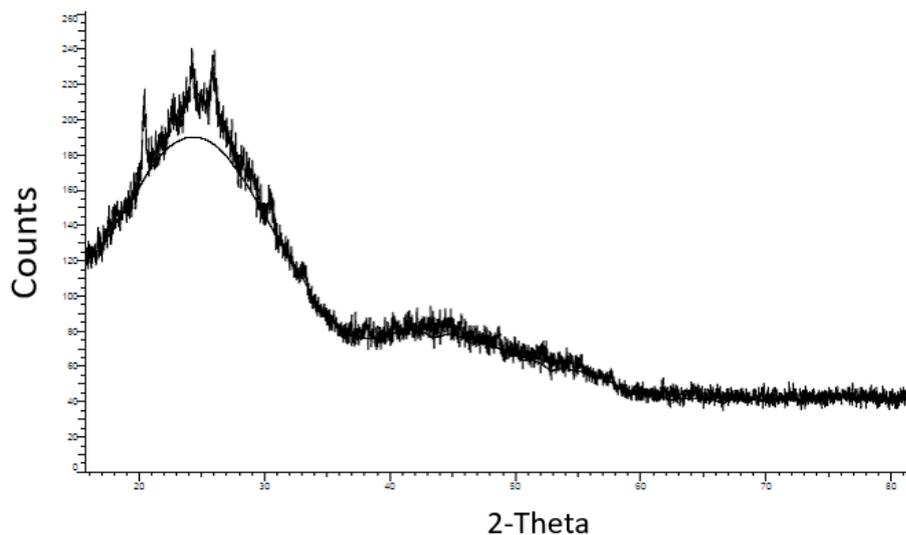


Figure S1. X-ray diffraction spectra for PNDC-2

Surface area analysis (BET)

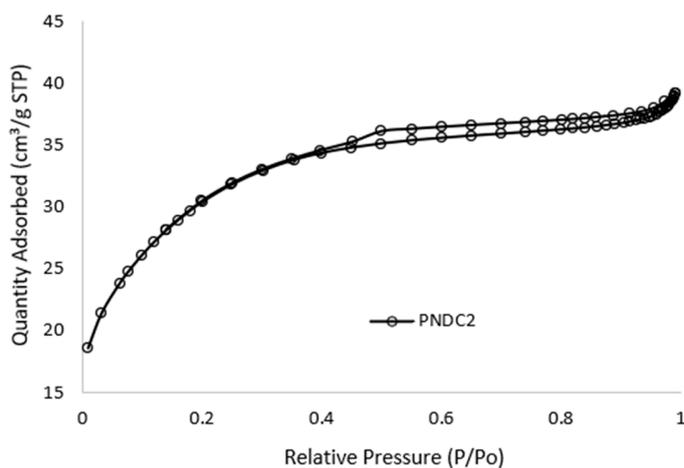


Figure S2. BET isotherm plots for PNDC1

X-ray Photoelectron Spectroscopy (XPS)

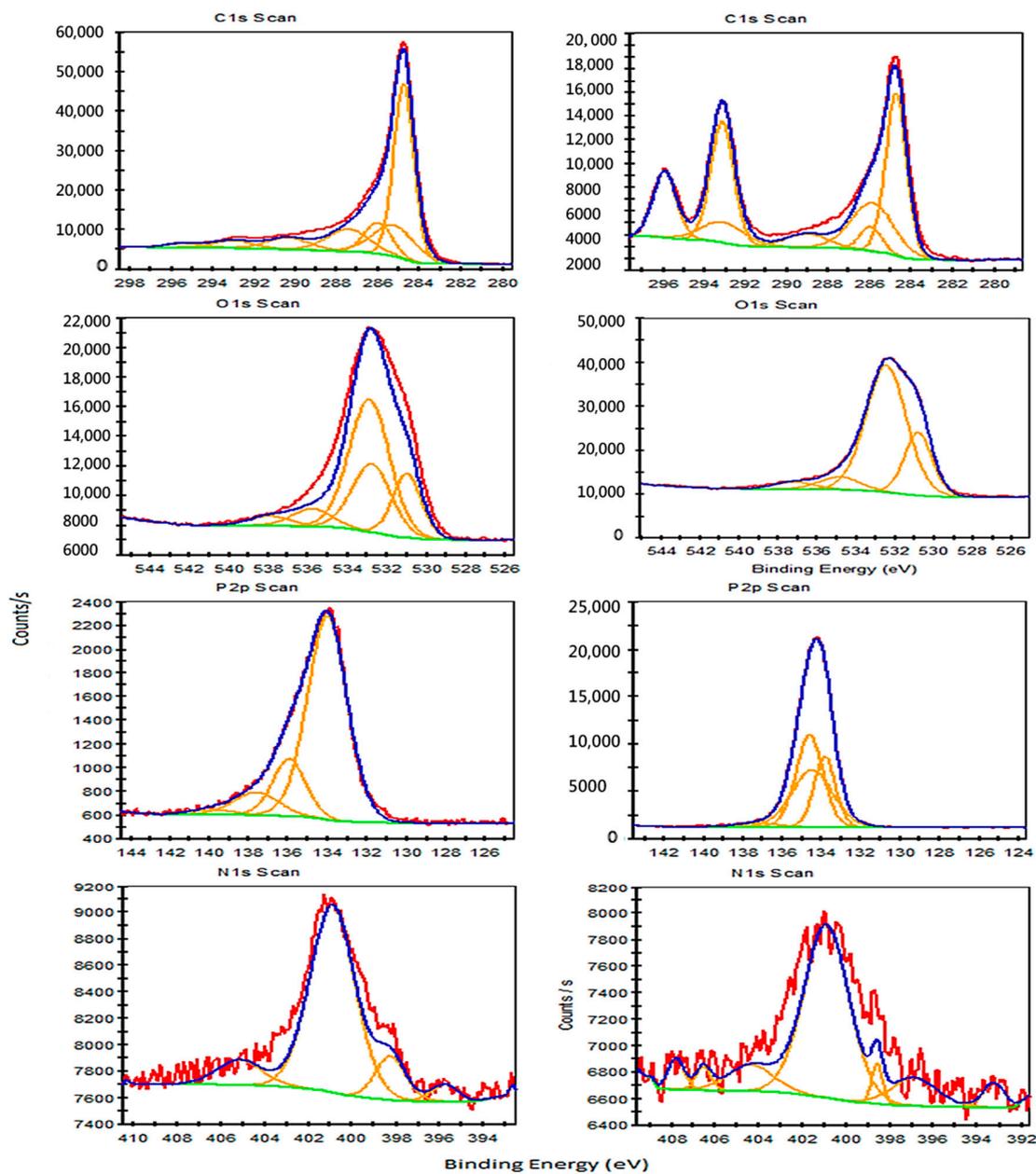


Figure S3. XPS narrow scan single trace views of PNDC1 (left) vs. PNDC2 (right). XPS spectra are of C1s, O1s, P2p and N1s. For all spectra, red lines indicate experimental (raw) data, blue lines represent fitted data, orange peaks are representative of functional groups on PNDC surface, and green line is background.

Electrochemical Observation

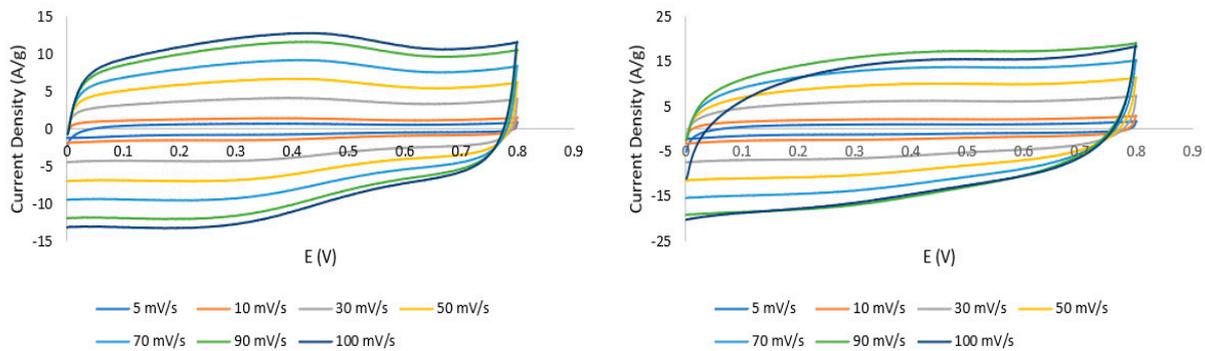


Figure S4. Cyclic voltammogram comparison in 1M H₂SO₄ electrolyte for PNDC1 (left) and PNDC2 (right) at varying scan rates.

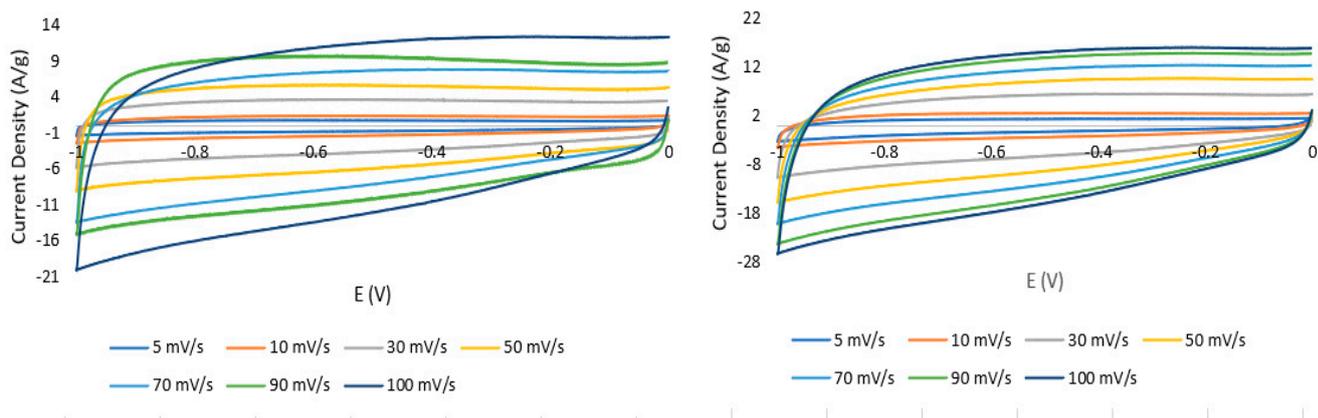


Figure S5. Cyclic voltammograms in 6M KOH electrolyte for PNDC1 (left) and PNDC2 (right) at varying scan rates.

Electrochemical recyclability

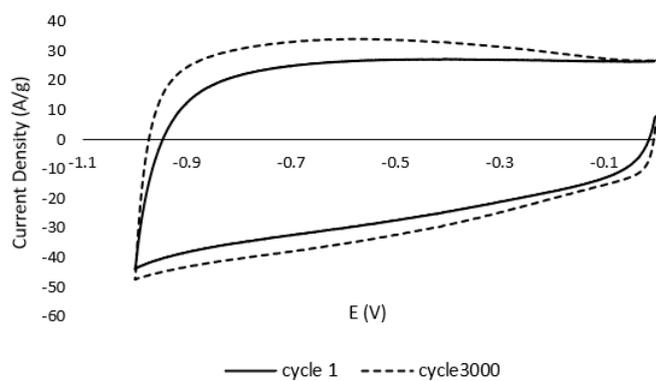


Figure S6. PNDC2 stability for 3000 cycles at 50 mV/s vs. 6M KOH electrolyte using three-electrode system.

References

1. Seehra, M. S., Narang, V., Geddam, U. K., & Stefaniak, A. B. Correlation between X-ray diffraction and Raman spectra of 16 commercial graphene-based materials and their resulting classification. *Carbon* **2017**, *111*. <https://doi.org/10.1016/j.carbon.2016.10.010>
2. Seehra, M. S., Geddam, U. K., Schwegler-Berry, D., & Stefaniak, A. B. Detection and quantification of 2H and 3R phases in commercial graphene-based materials. *Carbon* **2015**, *95*. <https://doi.org/10.1016/j.carbon.2015.08.109>