Enhanced metal-ion adsorption and diffusion in two-dimensional C_3N/Black phosphorene heterostructures: A multidisciplinary computational-experimental approach for next-generation batteries

Akhil K. Nair[1,*], Brohath Amrithraj[2], Carlos M. Da Silva[1], Gisele Azimi[2] and Cristina H. Amon[1,2]
[1]Department of Mechanical and Industrial Engineering, University of Toronto
[2]Department of Chemical Engineering and Applied Chemistry, University of Toronto
*akhil.nair@mail.utoronto.ca

Mechanical & Industrial Engineering
UNIVERSITY OF TORONTO

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Introduction

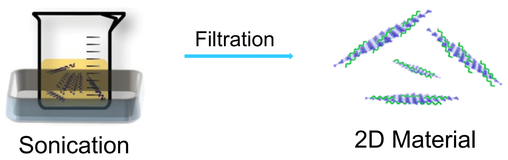
- Developing two-dimensional (2D) van der Waals (vdW) heterostructures by combining different 2D materials offers new possibilities for battery electrodes by integrating desirable properties and overcoming the limitations of the constituent monolayers.
- This work is a multidisciplinary collaboration between ATOMS Lab in MIE and LSM Lab in MSE/ChemEng.
- We are comprehensively investigating C3N/black phosphorene (C3N/BlackP) heterostructures as anode materials for metal-ion batteries based on DFT calculations and cutting-edge synthesis and characterization approaches.

Computational details

- Density Functional Theory (DFT) calculations were implemented with the Quantum ESPRESSO and Quantum ATK packages, utilizing GGA with the PBE parameterization technique.
- For our investigation, we considered a C3N/Black P heterostructure supercell comprised of 3 × 3 × 1 supercells of C_3N and B.P, with a total of 24 C, 8 N, and 24 P atoms.

Experimental details

- The objective is to synthesize few layer heterostructure with minimal defects to enable high performance in aluminum-ion batteries.
- Liquid phase exfoliation was chosen as the preferred synthesis method due to its scalability and simplicity.



- The resulting material was characterized using Scanning Electron Microscopy (SEM) imaging; the data obtained will be used for tuning the synthesis process.

Lattice structure and adsorption of Al ions on C_3N/BP heterostructure

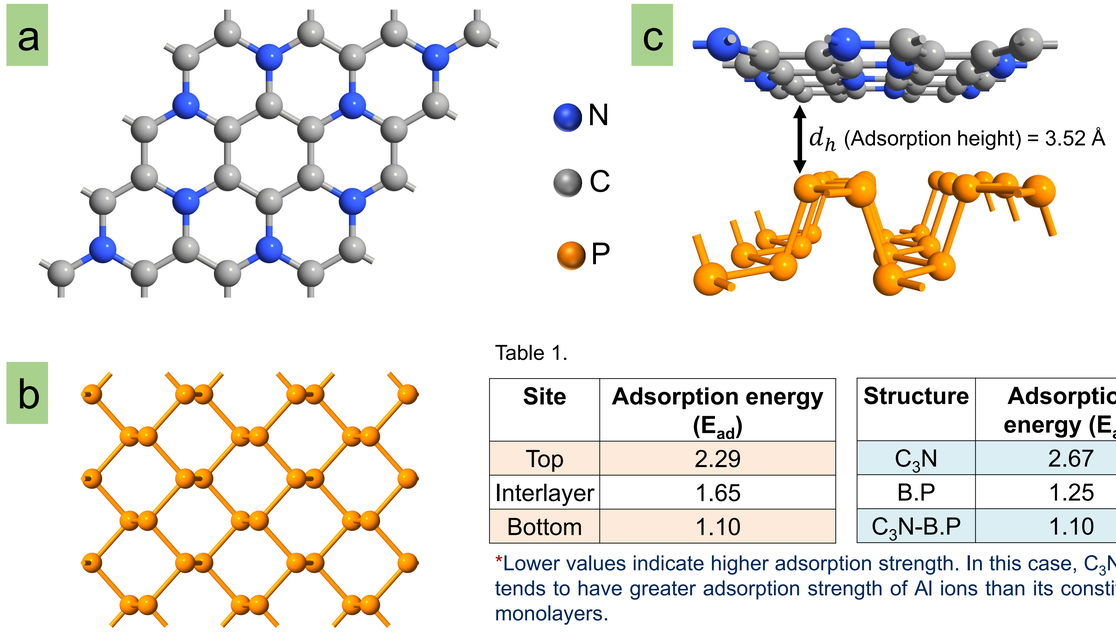


Table 1.

Site	Adsorption energy (E_{ad})
Top	2.29
Interlayer	1.65
Bottom	1.10

Structure	Adsorption energy (E_{ad})
C_3N	2.67
B.P	1.25
C_3N-B.P	1.10

*Lower values indicate higher adsorption strength. In this case, C_3N-B.P tends to have greater adsorption strength of Al ions than its constituent monolayers.

Fig. 1. Diagrammatic representation of monolayers of (a) C_3N, (b) black phosphorene (B.P) and 2D heterostructure of (c) C_3N/B.P

Our initial findings indicate that C3N/BlackP heterostructures exhibit improved metal-ion adsorption compared to their monolayer counterparts.

Synthesis of C_3N/BP heterostructure

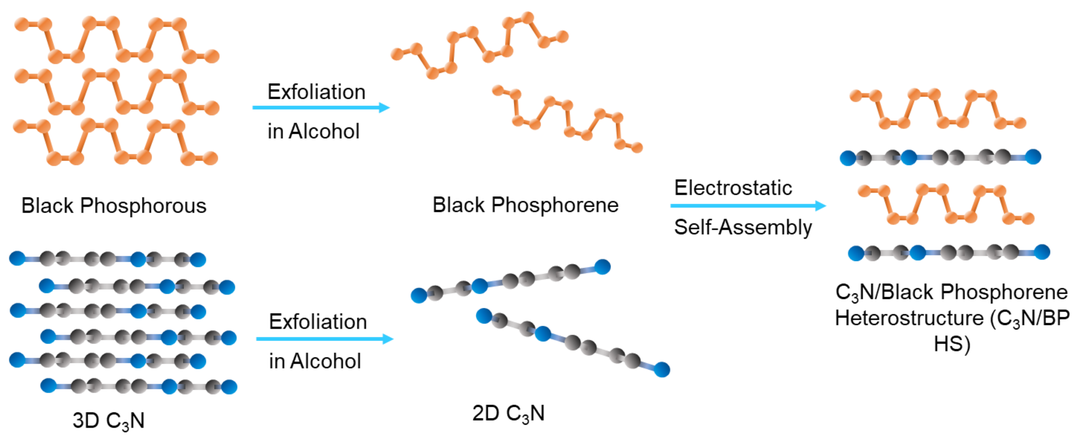


Fig. 2. Synthesis of heterostructure using a 2-step liquid phase exfoliation process

- This synthesis process can be tuned using the solvent, sonication power and duration of sonication as the parameters.
- Ideally, the chosen solvent must not react with C_3N and Black Phosphorous, and the combination of sonication power and duration must deliver enough energy to exfoliate layers of the parent materials.
- A prototype of the heterostructure was synthesized using a sonication duration of 1 hour at a power output of ~60 W.

Preliminary Results

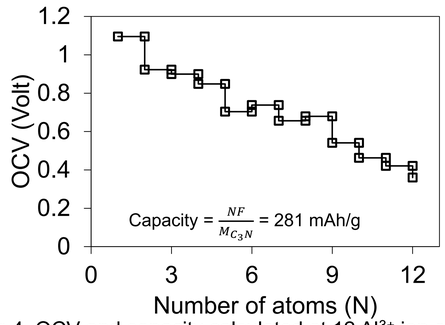


Fig 4. OCV and capacity calculated at 12 Al^{3+} ion adsorption

- DFT predicts an OCV of 0.4 V with a capacity of 281 mAh g^{-1}.

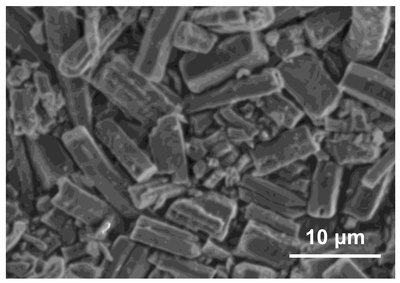


Fig 3. SEM image of synthesised prototype

- Imaging shows the size of particles is less than 10 µm.
- Exfoliation must be performed at a higher power output to achieve smaller particle dimensions.

Conclusion

- DFT simulations suggest a high-performing heterostructure cathode for Al-ion batteries. However, practical limitations need to be overcome to achieve this performance.
- The C_3N/Phosphorene heterostructure prototype was synthesized using liquid phase exfoliation and characterized using SEM imaging.
- Future work will optimize the synthesis process and use DFT to predict other electronic properties such as the density of states.

Acknowledgements

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