

# Plasma-Tailored Graphitic Nanocarbons for High-Performance Aluminium Batteries

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Existing electrochemical energy storage systems, including Li-ion batteries, continue to face critical limitations, such as unsatisfactory energy performance, limited availability of raw materials, high manufacturing costs, and safety concerns. These challenges have intensified the search for alternative energy storage systems, and multivalent-ion batteries have emerged as a promising option offering better cycling stability, lower cost, and improved safety. Among them, aluminium-ion batteries (AIBs) stand out due to the high abundance (~8 wt.% in the Earth's crust), exceptionally high volumetric capacity (8046 mAh mL<sup>-1</sup>), intrinsic safety, and economic feasibility. Despite these advantages, AIBs still fall short of achieving energy and power densities comparable to those of LIBs due to their lower redox potentials and higher ionic atomic weights. Therefore, fabricating advanced cathode materials capable of accommodating a larger number of Al<sup>3+</sup> ions, alongside compatible metal anodes, is crucial for realising high-performance AIBs. Plasma-enabled techniques are widely known for engineering materials at the nanoscale for various applications [1,2]. This work discusses improving the aluminium storage capacity of 2D graphitic nanomaterials through low-pressure plasma synthesis and surface engineering, with the aim of using them as efficient cathodes for AIBs. Graphitic nanocarbons synthesised and engineered via plasma-enabled techniques in different gases: nitrogen, argon and hydrogen were used to engineer the surface and controllably tailor the graphitic structure. The optimised plasma discharge conditions demonstrate a significant improvement in aluminium storage, delivering almost double the discharge capacity (123 mA h/g) compared to non-plasma-treated graphitic structures (67 mA h/g). In addition, the plasma-treated electrode materials demonstrated a 95% rate capability at higher current densities and remarkable long-term stability, maintaining the initial capacity after 1000 cycles. In addition, the charge-storage mechanism was evaluated using ex situ Raman characterisation of the electrodes at the charged states, and a clearly observed peak shift corresponding to the intercalation of chloroaluminate ions was observed. These findings provide valuable insights into the charging/discharging processes and intercalation chemistry of AIBs and highlight the potential of plasma-engineered graphitic materials as promising alternatives to conventional lithium-based battery technologies.

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## References

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## Acknowledgement

This work is supported by the EU Graphene Flagship FLAG-ERA III JTC 2021 project "VEGA" (MIZS-PR-11938) and M-ERA. NET 3 project ANGSTROM (MVZI, Slovenia (PR-13503)).

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