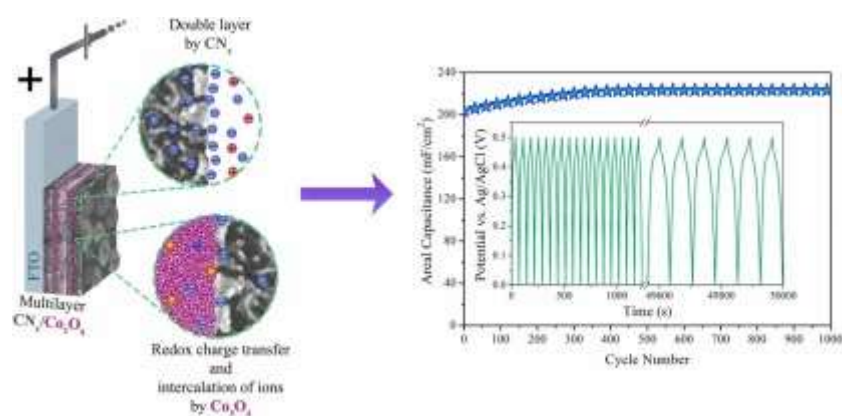


## Abstract

In this work, we report a direct study of the enhanced electrochemical performance of  $\text{CN}_x/\text{Co}_3\text{O}_4$  nano-composite multilayer electrodes which demonstrate great promise as valuable electrode materials for energy storage applications.  $\text{CN}_x/\text{Co}_3\text{O}_4$  electrodes were prepared by a novel hybrid non-thermal plasma/sol-gel deposition technique combining dielectric barrier discharge plasma with a  $\text{C}_2\text{H}_4/\text{N}_2$  gas mixture and sol-gel spin-coating methods. The results indicated that the rough and porous  $\text{CN}_x$  (a-C:H:N) layer consisting of various N species (especially graphitic, pyrrolic and pyridinic N) generated by nitrogen plasma improves electrical conductivity and facilitates ion diffusion in the a-C:H:N/ $\text{Co}_3\text{O}_4$  electrode. Furthermore, the synergistic effects arising from interleaving redox active  $\text{Co}_3\text{O}_4$  interlayers between  $\text{CN}_x$  layers involving numerous nano-pores led to the remarkable electrochemical activity of the composite electrodes. It was found that the  $\text{CN}_x$  layers noticeably overcame the poor cyclic stability of  $\text{Co}_3\text{O}_4$  by protecting it from dissolution in the electrolyte and these composite electrodes remained significantly stable after a very long charge-discharge time. The areal capacitance of  $203.4 \text{ mF}/\text{cm}^2$  at  $3 \text{ mA}/\text{cm}^2$  of the a-C:H:N/ $\text{Co}_3\text{O}_4$  multilayer electrode enhanced by 9.7 % of the initial capacitance after 450 CD cycles and stayed stable afterward. This unique hybrid architecture showed intriguing electrochemical properties including rapid redox reactions, wide potential window of 2.73 V, high surface activity and favorable cyclic stability as well as proper reversibility of a-C:H:N/ $\text{Co}_3\text{O}_4$  multilayer electrode which makes it a valuable low-cost candidate for electrochemical applications such as energy storage electrodes and biosensors.

## Graphical Abstract



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spin-coating methods. The results indicated that the rough and porous  $CN_x$  (a-C:H:N) layer consisting of various N species (especially graphitic, pyrrolic and pyridinic N) generated by nitrogen plasma improves electrical conductivity and facilitates ion diffusion in the a-C:H:N/ $Co_3O_4$  electrode. Furthermore, the synergistic effects arising from interleaving redox active  $Co_3O_4$  interlayers between  $CN_x$  layers involving numerous nano-pores led to the remarkable electrochemical activity of the composite electrodes. It was found that the  $CN_x$  layers noticeably overcame the poor cyclic stability of  $Co_3O_4$  by protecting it from dissolution in the electrolyte and these composite electrodes remained significantly stable after a very long charge-discharge time. The areal capacitance of  $203.4 \text{ mF/cm}^2$  at  $3 \text{ mA/cm}^2$  of the a-C:H:N/ $Co_3O_4$  multilayer electrode enhanced by 9.7 % of the initial capacitance after 450 CD cycles and stayed stable afterward. This unique hybrid architecture showed intriguing electrochemical properties including rapid redox reactions, wide potential window of 2.73 V, high surface activity and favorable cyclic stability as well as proper reversibility of a-C:H:N/ $Co_3O_4$  multilayer electrode which makes it a valuable low-cost candidate for electrochemical applications such as energy storage electrodes and biosensors.

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