**Graphene like carbon-nitride monolayer as the cathode of Al-ion battery**

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In aluminum-ion batteries (AIB), a graphite cathode shows very promising electrochemical performance. [1] However, its low capacity and high volume expansion is a major hindrance. This is due to the limited intercalation of the AlCl4 ion into the graphite layers. [2] Studies have revealed that one of the very effective approaches to enhance the capacity of the cathode is to dope graphite structures with nitrogen atoms. The addition of N atoms into a system certainly increases the number of electrons and the system starts to behave as an electron donating n-type dopant. [3] In addition, Bhauriyal et al [4] reported that the C3N possesses higher theoretical electrical capacity than graphite.

Therefore, in order to test the suitability of the nitrogen doping, pyridinic, pyrrolic and graphitic N-doped graphite structures were all studied and it was revealed that only graphitic nitrogen structures can be the suitable cathode materials for the AIBs. Hence, we have tested two graphitic carbon nitrides with low concentration of nitrogen-doping (C11N) and high concentration of nitrogen doping (C4N) [5] as the potential cathode materials for AIB with the density functional theory (DFT).

From our calculations, it is found that compared to graphene the AlCl4 anions bind more strongly with carbon nitrides with binding energy of 2.21 eV, 3.49 eV and 3.58 eV for graphene, C11N and C4N respectively. Also, the anions gain more charges from the cathode with charge gain of 0.85e, 0.93e and 0.95e per anion from graphene, C11N and C4N respectively. As a consequence the theoretical electrical capacity of the carbon nitrides are found higher compared to graphite. The stage-1 capacity of the graphene, C11N and C4N are 124 mAh/g, 144 mAh/g and 185 mAh/gm in the single layer and 155 mAh/g, 185 mAh/g and 180 mAh/g in the bilayer. Also, the flat potential of the carbon nitrides indicate that there is no barrier for charge transportation; hence the charging of the battery should be similarly as fast as graphite. However, no significant improvement have been observed in the area of volume expansion and voltage profile. Interestingly, low concentration of nitrogen doping is found as equally effective as the high concentration of nitrogen doping.

**References**

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