Understanding and predicting heteroatom dopants for carbon anodes.

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The introduction of heteroatoms, like N, to carbon can enhance the capacity of the material.[1] For example, the addition of less than 3 at% N to graphene type carbon resulted in a reversible capacity increase of at least 200 mAh/g over pure graphene.[2,3] Similar capacity and cycleability enhancements are reported for other carbon structures such as carbon nitride nanobelts (~480 mAh/g, 10 at% N)[4], bamboo-like carbon nanotubes (~493 mAh/g, 29 at% N)[5], nitrogen doped carbon nanotubes (~494 mAh/g, 16.4 at% N)[6], carbon nanofiber webs (~940 mAh/g, 16 wt% N)[7], and mesoporous carbons (<1200 mAh/g, ~20 wt% N).[8] The exact origin of the Li-ion insertion mechanism or enhanced capacity with N addition is not clear.

To identify structural motifs and the role of N in the electrochemical capacity of N-doped carbons we investigated the lithiation of nitrogen rich carbon nitride phases C_3N_4 (> 52 at% N). These results show that the Li irreversibly reacts with graphitic C_3N species but not with pyridinic C_2NH.[9] Synthesizing carbons with only C_2NH result in stable and durable anode materials.[10]

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