

Microstructure-performance correlations of disordered carbons as negative electrode materials for Na-ion batteries

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With the recent renewal of interest for Na-ion batteries as a potential low cost alternative to Li-ion technology, the study of sodium insertion into carbonaceous materials experiences a revival in recent years [1]. Although many candidate positive electrode active materials have been discovered with fairly good performance [1], the choice is very limited on the negative electrode side [2,3]. Mimicking the Li-ion technology shows its limit in this domain as graphite presents negligible Na insertion [4,5,6] (figure 1a). Although promising efforts have been recently dedicated to circumvent this issue [6,7], it does not match the best reports made on disordered carbons [8,9,10,11,12].

Disordered carbons, being “soft” or “hard”, present a typical sloping voltage-composition curve below 1V traditionally ascribed to defects assisted insertion of Na between the graphene layers [13] (figure 1b), while the specificity of hard carbons is to present an additional low voltage plateau ascribed to the packing of sodium within the micro-pores formed by cross-linked graphitic layers (figure 1c). Due to this extra capacity, the efforts of the community have been mainly focused on hard carbons. However, reported attempts to improve the capacity of hard carbons by increasing the micro-porosity, via ball milling [9] or physical activation [10], actually resulted in lower capacities than the pristine material after pyrolysis, demonstrating that the correlation between the microstructure of these carbons and their electrochemical performance is not yet fully understood.

Within this perspective, we underwent a comparative electrochemical and microstructural study of various soft and hard carbons. By coupling gas adsorption, powder X-ray diffraction (in-situ and ex-situ PXRD) and Small Angle X-ray Scattering (SAXS) we were able to depict the microstructure and morphology of these carbons. It allowed us to get new insights into the mechanism of sodium insertion into disordered soft and hard carbons and identify key microstructural features to be considered for best electrochemical performance.

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