CORE LEVEL BINDING ENERGY FOR NITROGEN DOPED CHAR: XPS DECONVOLUTION ANALYSIS FROM FIRST PRINCIPLES

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Amorphous carbon produced from lignocellulosic materials has received much attention in recent years because of its applications in environmental and agricultural management with potential to sequester carbon, serve as a soil amendment, and improve soil aggregation. Modern engineered amorphous carbons with promising properties, such as porous structure, surface functionalities (O, N, P, S) and layers with large number of defects, are used in the field of adsorption and catalysis. There is a growing interest in the production of nitrogen-doped carbonaceous materials because of their excellent properties in a variety of applications such carbon electrodes, heterogenous catalysis adsorption and batteries. However, quantifying the surface nitrogen and oxygen content in amorphous nitrogen doped carbons via deconvolution of C 1s x-ray photoelectron (XPS) spectra remains difficult due to limited information in the literature. No suitable method exists to accurately correlate both the nitrogen and oxygen content to the carbon (C 1s) XPS spectrum in the literature. To improve the interpretation of spectra, the C 1s, N 1s and O 1s core level energy shifts have been calculated for various nitrogenated carbon structures from first principles by performing density functional theory (DFT) based calculations. Furthermore, we propose a new method to improve the self-consistency of the XPS interpretation based on a seven-peak C 1s deconvolution (3 C-C peaks, 3 C-N/-O peaks, and π-π* transition peaks). With the DFT calculations, spectral components arising from surface-defect carbons could be distinguished from aromatic sp^2 carbon. The deconvolution method proposed provides C/(N+O) ratios in very good agreement (error less than 5%) with those obtained from total C 1s, N 1s and O 1s peaks. Our deconvolution strategy provides a simple guideline for obtaining high-quality fits to experimental data on the basis of a careful evaluation of experimental conditions and result