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High quality crystalline boron doped diamond growth on spherical monodisperse silica particles and impact of seeding density on the microstructure

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CO_2 conversion to chemical fuels such as alcohol and hydrocarbons has been found to be one of the most promising renewable energy source. It has urged the scientific community to develop efficient material in favor of CO_2 reduction. Boron-doped diamond (BDD) electrodes have aroused an interest among scientists for its outstanding electrochemical properties in particular its wide potential window, making it a good candidate for the electrochemical reduction of CO_2 [1].Moreover, hydrogenated BDD films, under UV illumination, could generate solvated electrons in aqueous solutions, making it highly relevant for photo(electro)catalytic CO_2 reduction [2].

In this study, we report an innovative approach that aims to grow a BDD coating onto monodisperse silica cores to obtain SiO_2@BDD core-shell particles by microwave plasma chemical vapor deposition (MPCVD). After removal of the core, hollow BDD could be used in the development of new electrode architectures which allows a significant gain in electro/photo-active surface. Currently, BDD particles are obtained by grinding massive BDD films, expensive process that produces in small amounts highly polydisperse particles.

Several investigations have correlated the electrochemical behavior of BDD with its crystallinity [3]. We therefore dedicated our work on the influence of several parameters on the crystalline quality, the morphology and the sp^2/sp^3 carbon ratio of diamond coating. These parameters are (i) the methane concentration in the gas phase, (ii) the nature of seeds (milled or detonation nanodiamonds), (iii) their density and (iv) the oxygen species removed from seeded silica cores during MPCVD process. The morphology of diamond coatings was characterized by scanning electron microscopy (SEM). Results showed that the lower initial seeds density and methane concentration, the larger grain size we obtain with faceted structure. Raman measurements on diamond coatings allowed to compare diamond crystalline quality and sp^2/sp^3 carbon ratio for the different growth conditions. Transmission electron microscopy (TEM), coupled with EELS, EDS spectroscopies were performed in order to access the diamond microstructure. The associated growth mode and the nature of the SiO_2 / diamond interface were studied in cross-sections obtained by Focused ion beam (FIB).

[1] Natsui et al. (2018), Angewandte Chemie, 130, 2669

[2] Zhang et al. (2014), Angewandte Chemie, 126, 9904

[3] Garcia-Segura et al. (2015), Electrochemistry Communications, 59, 52