

# Development of hierarchically porous materials for high energy –power density green energy technology devices

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Understanding the processes guiding the confinement of adsorbed ions, organic molecules (CH<sub>3</sub>CN, CH<sub>3</sub>OH, C<sub>2</sub>H<sub>5</sub>OH) and gas molecules (H<sub>2</sub>, NH<sub>3</sub>, CH<sub>4</sub>) in different porous structures [1-5] is vital for the development of sustainable green energy technology devices incl. supercapacitors, Li-ion and Na-ion batteries [6-9], polymer membrane fuel cells (PEMFC) and electrolysis cells (PEMEL) [10], as well as porous thin-film absorbents for effective cryo-adsorptive H<sub>2</sub> and CH<sub>4</sub>, NH<sub>3</sub> storage systems. The H<sub>2</sub> mass transfer and absorption characteristics analysed and established by quasi-elastic neutron scattering (QENS) method are important for aqueous electrolytes based supercapacitors, H<sub>2</sub> feeded PEMFC and PEMEL because the reversible/irreversible H<sub>2</sub> adsorption/absorption into catalytically active centres generated under over-polarisation of the above mentioned devices decreases the effective applicable surface area and therefore the energetic efficiency [1-8]. It should be stressed that QENS is applicable over a wide range of timescales (0.2 ps – 150 ps) to determine different self-diffusion (i.e. mass transfer) mechanisms of H<sub>2</sub> adsorbed in a ultramicro-, micro- and mesoporous carbon powders incl. a-SiC, TiC, VC, Mo<sub>2</sub>C, WC, Cr<sub>2</sub>C<sub>3</sub> and sol-gel C(TiC [11]. In addition to QENS, SANS and INES methods the bulk and porous structure of electrodes has been characterized by gas adsorption (N<sub>2</sub>, Ar, H<sub>2</sub>, CO<sub>2</sub>), mercury intrusion porosimetry, Raman spectroscopy, XRD, and wide-angle X-ray scattering (WAXS) [11,12], high-resolution transmission electron microscopy electron diffraction and electron energy loss spectroscopy methods. The characteristics have been tested by cyclic voltammetry, constant current charge/discharge, impedance and constant power methods for demonstration of ultramicro-, micro-, meso- and macroporous aeras role on the electrochemical parameters. To elucidate the sodiation mechanism of hard carbon, *ex situ* and *operando* synchrotron X-ray radiation total scattering experiments were performed at different synchrotrons (SR) [12]. *In situ* STM, *in situ* AFM and quartz crystal micro-balance have been applied for verification of surface and electrical double layer structures at very well ordered graphitic electrodes [6,11]. The ideal polarizability limits of carbon | RTIL interface have been established using SR-XPS method [13]. The role of hierarchically porous structure on the dispersion/ distribution of characteristic time constant (four different values from milliseconds to hundred of minutes) on the shape of C<sub>p</sub>, log frequency and log|Z'| vs. log f, and energy density (E) vs. power density (P) plots will be discussed. *In Situ* nuclear magnetic resonance method has been tested for selective establishment of interactions of ions charged with and electrode surface dependent on the ions' chemical composition. Extremely high P values at fixed E has been observed for materials with optimal micro/meso-porosity. Very low E and P values have been obtained for ultramicroporous and purely meso-macroporous electrodes with very high mesopore volume and surface area. Molecular dynamic and density functional theory calculations have been conducted for detailed analysis of adsorption/sorption layer ionic (RTILs) and molecular-ionic structures. The quartz crystal microbalance has been conducted for visco-elastic analysis of RTILs and electrolytes in porous carbon materials.

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