

DESCRIPTION FOR THE GENERAL PUBLIC

Energy can be stored electrochemically either through redox reactions, e.g., in accumulators (called also batteries), or by electrostatic forces in electrochemical capacitors (called also supercapacitors). In the latter case, the charges are stored in the form of an electrical double-layer (EDL) created at the interface between an activated carbon (AC) electrode and an organic electrolytic solution (the most often tetraethylammonium tetrafluoroborate in acetonitrile – $\text{Et}_4\text{NBF}_4/\text{ACN}$), thus the systems are also called electrical double-layer capacitors (EDLCs). Owing to this particular mechanism, EDLCs represent one of the most appealing strategies for fast storage of energy and, consequently, for the realization of high power energy storage devices. Owing to these particular properties, EDLCs are among others implemented in hybrid and electric vehicles, and represent a unique solution to reduce urban pollution. Nevertheless, their application could be widespread by improving energy density, while retaining high power density and long cycle life. Taking into account that the stored energy (E) is equal to $E = \frac{1}{2} CU^2$, research effort is essentially oriented to enhancing the capacitance C and the cell potential U . To reach this objective for both parameters, it requires to better figure out the structure of the EDL in the pores of AC electrodes, which by essence is very different from the traditional EDL models established for flat electrodes.

Most of the studies performed on polarized AC electrodes have attempted to determine the composition of the EDL in the pores at a given potential and the molecular fluxes when varying the potential. It has been demonstrated that ions are at least partially desolvated in pores smaller than 1 nm, e.g., that they lost a part of their solvating molecules as compared to the bulk. *In-situ* experiments by Nuclear Magnetic Resonance, Electrochemical Quartz Crystal Microbalance (EQCM) and InfraRed (IR) spectroscopy demonstrate a rearrangement of the electrolyte species inside the carbon nanopores under polarization. Nonetheless, the EDL charge storage mechanism has not yet been carefully analyzed by considering both the microscopic properties of the electrode/electrolyte system (pore size distribution of carbon electrodes, relative sizes of ions and their solvation sphere, solvent nature and electrolyte concentration) and the macroscopic ones (electrodes performance in terms of capacitance and resistance, cathodic and anodic decomposition potential of the electrolytic solution).

In this context, the objective of the **EDLstruct** project is to implement various organic electrolytes and carbons to study the relationship between the porous texture of carbons and the ion/solvent ratio in the porosity for various values of positive and negative potential, and to find correlations with the decay of electrochemical performance of carbon electrodes depending on temperature and electrodes potential.

The Friedrich Schiller University Jena group (FSU Jena) will design a range of organic electrolytes by varying the solvent, the salt and its concentration. The Poznan University of Technology group (PUT) will design ACs of controlled porous structure and surface functionality. In order to acquire information about the EDL formation/composition under the effect of polarization, PUT will measure the molecular fluxes by *in-operando* electrochemical dilatometry and realize post-mortem analyses of polarized electrodes by thermoprogrammed desorption. The relationship between the EDL composition in the pores of carbon (especially solvent amount) and its thermal stability will be investigated by thermal analyses - TGA/DSC (FSU Jena) and electrochemical on-line mass spectrometry (PUT). Finally, PUT and FSU Jena will sum up the research results and establish a model of EDL formation/composition in porous carbons; based on this model, the decomposition mechanisms occurring in the pores of carbon under polarization will be interpreted.

As already stated above, extending the concept of EDL formation/composition from a flat surface to the surface of pores in a confined system will represent an important scientific breakthrough. Moreover, since AC electrodes are widely applied in energy storage systems, a better knowledge of the EDL structure in these materials should enable to improve the capacitive performance and cell potential of EDLCs, thus representing an important challenge from the societal and economic point of view. The combination of the know-how of PUT for carbons and FSU Jena for electrolytes represents a unique opportunity to conduct this research which optimal efficiency and chances of success, especially taking into account that the two principal investigators already worked together during writing of a highly cited review in *Advanced Materials*.