We are currently witnessing the rapid development of many energy-storage technologies with a great impact in our daily lives. This development is visible in a wide range of industrial sectors ranging from portable applications, such as consumer electronics, through autonomous wireless sensor networks and structural health monitoring, to large-scale applications, such as environmentally-friendly electric vehicles. Critical technical specifications regarding fast delivery of energy on minute or even second timescale and long operational lifespan are required to make these applications possible. Electrical double-layer capacitors (EDLCs), or supercapacitors, have emerged as promising electrochemical energy storage devices. They satisfy the mentioned requirements due to their excellent capacitive properties, high power density, excellent cycling stability, and reversibility. Nowadays, the market of EDLCs already involves numerous commercial applications, for example, in electric mobility for stop/start systems in cars, electric drive buses, *etc*. Despite the tremendous research effort, the low energy density and the limited electrochemical performance at low temperatures are currently one of the main bottlenecks, hindering the wide-spread application of these ecologically-friendly energy storage systems. Therefore, new perspectives from the material science point of view are crucial to accomplish tomorrow's technological challenges in this field.

In recent years, the synergistic effect produced between novel electrolytes based on ionic liquids (ILs) and the design of porous carbonaceous nanostructure electrodes has paved the way to conceive high-performance EDLCs. This was due to the outstanding features of electrodes: a large operating cell voltage, wide working temperature range, and excellent capacitive properties. However, the energy storage mechanisms involved at the electrode/electrolyte interface during the charging and discharging processes have not yet been fully understood. A selection of different IL mixtures by tuning their anion/cation size, shape, valence, and molecular flexibility will be investigated in nanoporous carbons at different working temperatures and operating cell voltages to reach this challenging goal. The correlation of the device's electrochemical performance as a function of the employed IL mixture and the pore size distribution of carbon electrodes will be scrutinized using a multi-disciplinary approach based on electrochemistry, modeling, and in-situ nuclear magnetic resonance measurements. As a result, the knowledge acquired in this project will enlighten new horizons to design reliable, high-performance EDLCs in the near future.