

## DESCRIPTION FOR THE GENERAL PUBLIC

The project concerns synthesis of synergetic molecular hybrids called IoCarboNanoFluids ('ICON Fluids') containing chemically designable, zippable covalently modified by ionic carbon nanotubes (ICNTs) and ionic liquids (ILs). From the chemist's point-of-view, the 'teeth of the zip' play the key role as they should fit one into the other. Importantly, such nanosystems should reveal excellent, smart, and functional characteristics toward for miniaturization of flexible electronics. All of those required properties will be unraveled using an exceptional combination of analytical techniques exploring the fate of ultrasounds and alternative current upon passing through the title hybrids. This analysis will be accompanied by in-depth, three-dimensional atom-scale imaging of various localizations of differently assembling ions and nanotubes. Overall, the molecular arrangements will be translated into the unique properties of ICON Fluids which can resemble liquids, as well as plastic or elastic gels.

And now more about the components themselves. Single-walled (SW-) and multi-walled CNTs (MWCNTs) are characterized by high surface area, excellent mechanical properties, high electric conductivity, outstanding chemical and electrochemical stability. In turn, ionic liquids (ILs), so-called 'designer' compounds, consist of organic cations presenting a low symmetry and inorganic or organic anions. ILs' properties are dominated by strong electrostatic interactions between ions. Hence, ILs have exciting electrical and electrochemical properties as well as high thermal and chemical stability, low vapor pressure, and non-flammability. Statistics-driven surface functionalization of CNTs with simple moieties such as hydroxyl (-OH) or carboxylic (-COOH) groups is straightforward, but also very destructive for nanotube structure and therefore properties. On the other hand, robust and long-lasting functionalization of CNTs with ionic moieties resembling components of ILs (cations and anions) is related to strong covalent modifications while also weaker, non-covalent modifications (achievable by simple mixing) such as based on the interactions between the carbon-based  $\pi$ -systems and highly polarizable  $\pi$ -electrons in ILs. The structure of the latter, i.e. ionanofluids – non-covalently functionalized CNTs – is not well defined. Those systems are composed of CNTs dispersed in ILs with interfacial nanolayer of IL on the CNT surface. However, how the nanolayers are organized and how the nanolayers can be projected and controlled in the fluid state and what are their nanoscale properties still remain open and challenging questions.

This project is about to bring the answers for the above questions by the molecular design of the ICON Fluids – a new class of 'properties-by-design' CNT-functionalized multi-ionic systems which will form the assembled molecular architectures. The control over functionalization of defectless CNTs will be achieved by directing functionalizing agents – trapped in a template – and hence guiding the agent to the nanotube surface while upon the reaction with nanotube the template will be removed. By designing covalently ion-functionalized carbon nanotubes (ICNT<sup>+</sup> and ICNT<sup>-</sup>) and the appropriate counterions, it would be possible to qualitatively and quantitatively control of organization of molecular architecture, i.e. both solid-liquid interface structure and the bulk properties of ICON Fluids. The IoCarboNanoFluids will be hence composed of CNT<sup>+</sup> 'carbon nanotubium', CNT<sup>-</sup> 'carbon nanotubate', and the other two types of ions. In some cases, the 'zipping–unzipping' mechanism should be expressed between ICNT<sup>+</sup>/ICNT<sup>-</sup> contacts like in case of CNT–CNT in so-called CNT buckypaper (based only on non-ionic interactions). The four-ionic systems composed of ICNTs or ionically-zippable ICNTs and ILs have not been synthesized to date.

The proposed idea found the interest of many leading centers in the world. Analysis of chemical structure of ICON Fluids will be collaboratively performed using cryogenic transmission microscopy (cryo-TEM), combined with scanning transmission electron microscopy (S/TEM) and energy dispersive X-ray spectroscopy (EDS) with Department of Chemistry at the University of Cambridge. The cryo-TEM method enables direct imaging and characterizing the system in its 'natural state'. ICON Fluids will be made glassy by rapid cooling and examined in the form of an amorphous film. The 3D-nanoscale morphology and distribution of nanostructures down to atomic imaging will be characterized using S/TEM tomography and EDS tomography enabling to obtain atomic elemental maps. Additionally, our idea of the ultrasound absorption spectroscopy (UAS) coupled with electrochemical impedance spectroscopy (EIS) will allow the unique sonoelectrochemical structural studies and the insights into the complex structure of the ICON Fluids. The unique quantitative information about the timescale of various relaxation processes that occur in ICON Fluids can be obtained and subtle sample differences can be distinguished. The molecular dynamics calculations which will be performed in collaboration with Centro de Química Estrutural at the Universidade de Lisboa helping to answer the questions how molecular nanoarchitecture and physicochemistry of ICNTs and ILs governs the electrical, thermal and rheological properties of ICON Fluids. Further, the unique isochoric thermal characteristics at extremally high temperatures will be realized in collaboration with Geothermal & Renewal Energy Institute, High Temperature Joint Institute of the Russian Academy of Sciences. The expected effect of this project is to create the protocol of automatic, precise control of structure and interrelated properties of ICON Fluids enabling the precise adjustment of counter ions as puzzles toward switchable properties and molecular 'domino effect'.