

Effect of Na/Li concentration in ionic liquid electrolytes for ion batteries

In recent years, the emergence of wireless gadgets used in our daily life as for example smartphones, laptops, tablets, shavers, electric toothbrushes, among many others makes essential the improvement of the technology that involves the energy storage. One of the most common and useful devices used to store energy are rechargeable batteries. Among different kind of batteries, lithium-ion (Li-ion) batteries are the most used in home electronics gadgets. In this kind of batteries, lithium ions are dissolved in a liquid electrolyte which allows the movement of ions from one electrode to the other during the charge and discharge process. The nature of the liquid electrolyte limits the efficiency, stability, and consequently the duration of the battery. Recently it has been proved as good candidates the so-called room temperature ionic liquid-based electrolytes due to their non-flammable and non-volatile nature. Room temperature ionic liquids (RTILs) are salt in the liquid state at ambient conditions. They are formed by a combination of organic cations and organic or inorganic anions which show excellent thermal and electrochemical stability and relatively high ionic conductivity.

Nowadays the lithium resources are enough to supply the demand of the current ion batteries; however the increasing requirements and power of the emerging technology could make it difficult in a short or medium period of time. For that reason it is necessary to find alternatives to lithium-ion batteries that can surmount this issue. However, replacing Li with Na can be a suitable choice in terms of battery cost, safety, and raw material abundance.

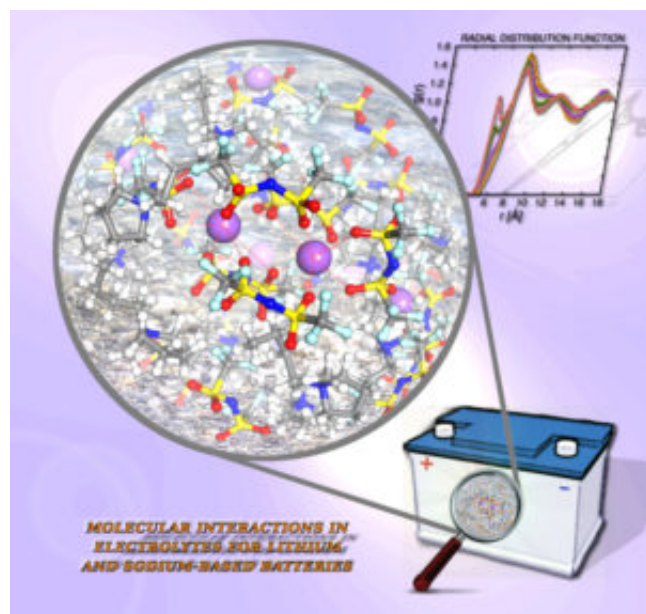


Fig. 1.

In our work, we investigate at microscopic level the behavior of electrolytes containing sodium or lithium salts (Fig. 1). We carried out Molecular Dynamics (MD) simulations with classical potentials that describe properly the physicochemical properties of the electrolytes reproducing experimental density, diffusion coefficients, and ionic conductivity. Electrolytes are composed by N-alkyl-N-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide $[C_4PYR]^+[Tf_2N]^-$ ionic liquid with the addition of different amounts of Na/Li salts ($Na^+/Li^+[Tf_2N]^-$).

It is well studied that increasing the amount of sodium and lithium salt dissolved in the ionic liquid, decreases the conductivity of the electrolyte. Ionic conductivity is the key property in the batteries field, so it is the best way to measure the ability of an electrolyte to conduct electricity. On the basis of MD results the decline in the conductivity can be related with the structural organization of the electrolytes. The contribution to the total conductivity of the sodium or lithium is negligible compared with the contribution of the ionic liquid compounds, so the decrease in the conductivity by adding sodium or lithium salt is not a direct effect. When sodium or lithium are dissolved in the aforementioned ionic liquid, these metals interact strongly with the $[Tf_2N]^-$ anion forming local clusters of 2 or 3 anions surrounding each metal. In addition to MD simulations, we performed quantum mechanics calculations to describe the arrangement of the clusters. On the one hand, quantum calculations support the findings of the MD simulations showing the same structural properties. On the other hand, they indicate that clusters are highly stable since they exhibit large binding energies between the metallic atoms and the $[Tf_2N]^-$ anion. These complexes affect to the microscopic structure of the electrolytes because the negative ions are brought closer to each other limiting slightly the mobility of the systems. The consequence of the reduction of the mobility of the species that form the electrolyte is a noticeable depletion in the total ionic conductivity.

The molecular simulations that we exploit in this work can provide a useful microscopic description to understand the molecular mechanism involved in the ionic transport of electrolytes for novel concepts in batteries among many others applications.

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Publication

[Quantum and Classical Molecular Dynamics of Ionic Liquid Electrolytes for Na/Li-based Batteries: Molecular Origins of the Conductivity Behavior.](#)

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