Abstract

Climate change and environmental degradation represent a huge threat to our society, and to limit them, switching to more sustainable energy sources is essential. The intermittent nature of these sources and the growing energy demand make it crucial to research new energy storage technologies for their reliable integration into the energy grid. In this context, a critical roadblock is the design of advanced materials suitable for this application, namely with a high capacity to store and transfer energy and reduced aging over time. To this end, improving the understanding of dynamic and complex phenomena occurring within materials or at the interfaces between two or more materials is crucial. Thus, in this thesis, relevant transport and aggregation phenomena at the nanoscale solid interfaces are investigated by combining various computational approaches and theoretical considerations.

In the first part of this work, a gold-based nanofluid is studied as a possible interesting candidate for advanced thermal energy storage and transfer systems, among other applications. It is evaluated how the interfacial morphology of a gold nanoparticle (AuNP) in an aqueous solution affects the interaction mechanisms with biocompatible polymers (Poly(Lactic-co-Glycolic Acid) - PLGA). This investigation is essential to provide guidelines for the rational design of a gold nanoparticle that facilitates the encapsulation process in a PLGA matrix. The latter holds the potential to improve the stability of gold-based nanofluid and reduce its potential health and environmental risks. Molecular dynamics simulations supported by an unsupervised machine learning algorithm are employed to show the temporal evolution of the aggregation mechanisms of PLGAs on AuNP by tuning the polymer concentration in the aqueous solution. Then, a detailed analysis of nanoparticle surface coverage coupled with free energy landscape calculations highlights the anisotropic nature of PLGA aggregation on the AuNP. This result demonstrates that the shape and surface morphology of nanoparticles may impact polymer aggregation and preferentially guide adsorption on specific sites.

In the second part of this work, the attention is focused on rechargeable batteries, which represent a key technology for electrochemical energy storage. The passivation layer that forms at the anode-electrolyte interface in current lithium-ion batteries and significantly affects their performance, called solid electrolyte interphase (SEI), is analyzed. Hence, understanding the phenomena controlling the formation and growth of SEI and its ion transport properties is critical to developing next-generation rechargeable batteries. The SEI consists of two layers: one near the anode interface, mainly formed of inorganic components, and the other near the electrolyte interface, primarily composed of organic components. First, non-reactive molecular dynamics simulations combined with an unsupervised machine learning algorithm are used to determine the rates of self-aggregation processes of the main organic SEI component. These phenomena drive the early stages of organic layer growth. Then, reactive molecular dynamics simulations associated with theoretical considerations are employed to analyze the lithium ion transport properties in one of the main inorganic SEI components. Such investigations provide an accurate understanding of the aggregation and transport phenomena under study and the foundational information to implement a larger-scale model. Thus, a new mesoscale model, namely a kinetic Monte Carlo, is proposed to connect the nanoscale features to the heterogeneous morphology of the SEI and the resulting macroscopic transport properties.

The modelling study conducted in this dissertation represents a first step towards the development of a virtual platform to design advanced materials for energy storage applications, as recommended by European roadmaps.