The ecological transition we are experiencing in these years involves a gradual shift from fossil fuels to renewable and sustainable energy sources, generally based on electricity. This scenario drives the research and development of electrical energy storage systems. Among different energy storages, lithium-ion batteries are probably the most competitive technology, thanks to their great energy and storage density, acceptable safety and lifespan.

Lithium-ion battery technology is in constant development, both from the side of research on new materials to increase energy and power densities, both on the control and modelling side, to estimate performance parameters, remaining life and strategies to mitigate degradation.

In this last scenario, mechanics plays a significant role: During operation, the electrode microstructure deforms because of the interaction with lithium ions. Such deformation leads to stress and crack propagation, damaging the electrode structure. Fracture propagation in the electrode is one of the main damaging mechanisms in lithium-ion battery, causing the reduction of performances (capacity drop and impedance rise) through life. Mechanics does not have only a bad influence on lithium-ion batteries, indeed in-operando mechanical measurements at macroscopic scale reveal as an interesting indicator for the estimation of battery performance and degradation.

The goals of this work are the following: (a) To understand the physics underpinning the mechanics of lithium-ion batteries and to model deformation and stress arising in the electrode microstructure during operation (Chapter 3, section 1 and 2); (b) To study how fracture propagates in the electrodes, how to design electrode to limit fracture and how to estimate the degradation caused by fractures at battery performance level (Chapter 3, section 3); (c) To create a model giving the battery deformation, temperature and voltage responses to a user-defined current (Chapter 4, sections 1-3), creating the basis for a physics based damaging model, predicting how battery performances and responses change with aging; (d) To experimentally measure the battery responses to different user-defined current rates (Chapter 2, section 2.1-3.1), and to experimentally investigate how the performances and responses change with aging (Chapter 2, section 2.4-3.4); (e) Finally, to validate the battery model with experimental measurements (Chapter 4, section 5).

To develop the experimental approach addressed in Chapter 2, a dedicated battery test bench was designed, allowing the test of 4 battery samples simultaneously according to a user-defined current profile while measuring voltage, temperature and deformation. At first, samples of two battery models (LFP and LCO) were characterized, to know the voltage, temperature and deformation responses at different current rates. It is interesting to highlight that deformation results almost linearly proportional to the battery state of charge, especially in LCO batteries. Subsequently, aging test on a LCO battery sample was performed. During aging, reference performance tests carried out every fixed number of aging cycles allows sensing the battery performance (capacity and internal resistance) and the battery responses. In particular, it is observed a perfectly linear relation between the capacity loss of the battery and the irreversible deformation of the battery.

From the modelling point of view, a micro-mechanics and a macro-mechanics battery model are studied in Chapter 3 and 4.

The micro-mechanics sub-model takes in input the lithium ions flux on the electrode microstructure from the macro-mechanics model as a boundary condition, to compute lithium ions concentration distribution within the electrode microstructure. The inhomogeneous lithium concentration results in stress and consequently fracture propagation. Stress intensity factor is computed to assess electrode design guidelines to limit fracture in the electrodes, and to define a battery damaging model based on the interplay between mechanics and side reactions.

The macroscopic battery model aims to compute the deformation, temperature and voltage responses at user-defined current rates. Focusing on the mechanical sub-model, a multi-scale model is established: lithium concentration and deformation of the electrode microstructure, computed at micro-scale, are the input of the macro-mechanics model, computing the macroscopic battery deformation. Finally, the deformation, temperature and voltage responses are validated with experimental measurements.

In the near future, the damaging model defined at micro-scale and addressed in Chapter 3 will be implemented in the battery model, to predict how the performances (capacity and resistance) and the responses of the battery change with aging. Secondly, state of charge and state of health estimation algorithms based on deformation measurements will be developed, aiming to overcome the current estimation difficulties when using traditional measurements.