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## Study of the physicochemical and electrochemical properties of glassy oxides as positive electrode active materials for Sustainable and High Energy Density Li-ion and Na-Ion batteries

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As energy concerns grow and the society needs more sustainable and renewable energy sources, the ability to effectively store and retrieve energy is paramount. Currently, rechargeable lithium-ion batteries (LIBs) seem to be one of the best alternatives to reduce our dependency on fossil fuels. LIBs are using cathodes materials based on polycrystalline oxides or polyanionic compounds [1]. However, their performances can be limited by their crystalline structure when other compounds suffer from metallic dissolution and irreversible phase changes during cycling [2]. To overcome these key shortcomings, implementing glasses or glass-ceramics as cathode materials seems to be an interesting approach [3]. Glasses have a structure composed of more free volume, which can accept a large amount of lithium [4] and easily accommodate structural changes upon lithium ions extraction/insertion [5]. Furthermore, glass production is a scalable process and can be commercially easier to implement than most of synthesis processes of conventional cathode materials.

The main objective of this study is to develop high capacity positive electrodes (up to 300mAh/g) for Li-ion batteries based on glassy oxides without critical transition metals. The ambitious target is to exceed 1000Wh/kg at the active material level and so enable energy densities of 300-350 Wh/kg at the Li-Ion cell level. Additionally, electrochemical behavior of some alkali-free glass compositions will be evaluated in sodium-ion configuration. The main objective is to correlate the most relevant physicochemical properties of glass with the corresponding electrochemical performances. For a better understanding of involved processes, three oxides glass families are studied as promising cathode materials for sustainable and high energy density lithium batteries. These oxides are composed of: i) a glass network-forming element (Si, B, P) leading to the 3 different families, ii) a transition metal (Mn, Fe, ...) and iii) an alkali (Li, Na).

The influence of the nature of the transition metal and the polyanionic group corresponding to the network-forming element (PO<sub>4</sub>, BO<sub>3</sub>, SiO<sub>4</sub>) on the microstructural and electrical properties of the as-prepared glasses will be presented mainly for the lithiated compounds. In that scope, we have: coupled many characterization techniques such as X-Ray Diffraction (XRD), SEM-EDX, DTA/TGA, Raman, IR and UV-Vis spectroscopies, Electrochemical Impedance Spectroscopy (EIS) and chronoamperometry. The effects of the polyanionic group and the transition metal element on the measured electronic and ionic conductivities on the bulky glassy materials as a function of temperature will be discussed. The electrochemical properties in terms of specific capacity, redox potentials, first cycle capacity loss, coulombic and energetic efficiencies of these materials were investigated in coin-cells vs lithium metal electrode.

Finally, to elucidate the reaction mechanisms of the electrochemical processes involved in these materials at room temperature, SEM-EDX, X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS) and Mossbauer Spectroscopy for iron based compounds were coupled and performed on the as-prepared glasses and on ex-situ (after cycling) materials at different electrochemical states of charge. In a next step, machine learning approach and tools will be applied to establish more precisely the relationships between the different glassy oxides structures and their electrochemical performances.

This new study will bring significant elements to optimize both the glass composition and its elaboration conditions to obtain high performance cathode materials without critical materials.

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