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Vanadium-free oxide glasses: Promising positive electrode material for Li-Ion and Na-Ion Technologies

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The development of new materials for battery positive electrodes is required to increase the batteries performances. Currently, best commercial positive electrode materials are crystalline oxides, and reach 150 to 190 mAh/g (so less than 700 Wh/kg at material level) for Li-ion technology (LCO, NCA or NMC material), and 120 mAh/g (or less than 450 Wh/kg at material level) for Na-ion technologies (NVPF or Lamellar Oxide materials). Nevertheless, glasses are promising materials as their vitreous structure can undergo structural changes like alkaline insertion/disinsertion mechanisms. Furthermore, depending on their composition, they can be easy to processed and produced on an industrial scale.

Most of the papers on glass positive electrodes reported in the literature focus on Vanadium-containing glass. Furthermore being a glass forming element that helps the glass network formation, Vanadium presents multiple oxidation states that can lead to high electrochemical performances through the exchange of several electrons per Vanadium. The reported glasses are generally elaborated in reducing conditions and reach specific capacities around 300 mAh/g (i.e. 1000 Wh/kg) for Li-ion cells [1]. Such performances in energy density make glass a highly interesting material family for positive electrode. Nevertheless, Vanadium is considered by European Union as a critical raw materials. So there is a strong interest in developing Vanadium-free glasses for positive electrode. These materials are less investigated in the literature, reaching around 60 mAh/g for iron silicates glasses in Li-ion technology [2]. Similarly, glasses for Na-ion battery positive electrode remain poorly studied. Specific capacity of 115 mAh/g was reported by Nakata *et al.* [3] for sodium iron phosphate glass.

The present work investigates new oxide glass compositions for Li-ion and Na-ion technologies. The glasses were synthesized and characterized to investigate their structures (SEM, EDS, Raman spectroscopy, UV-vis, XRD, ...). Positive electrodes were prepared with ball milling technique, assembled in coin cells using either Li-metal or Na-metal configuration. Then the positive electrodes were electrochemically evaluated by galvanostatic cycling. The electrode were characterized *post-mortem*. Data collection from structural, chemical and electrochemical characterizations are used to develop a database to predict best promising new compositions by using machine learning techniques. This work will also give first insights relative to the links between glass composition, elaboration conditions, structural properties, and electrochemical performances.

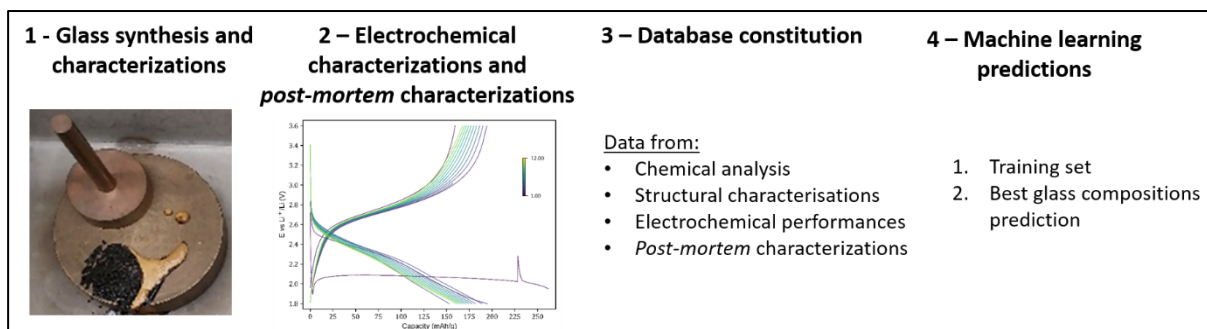


Figure 1: Methodology employed in our study

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