

Structural and computational analysis of $Y_{0.88}Ni_{3-x}Mn_x$ compounds to understand the localization of Mn atoms

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Abstract

Intermetallic compounds AB_n (A =rare earth, Mg; B =early transition metal, $2 \leq n \leq 5$) can store large amount of hydrogen. They have been investigated as negative electrode materials for Ni-MH batteries due to their outstanding electrochemical properties, including in new protic ionic liquid (PIL) electrolyte^[1-3].

Within this family, AB_3 compounds ($n=3$) are of primary importance due to their high capacity. In this work, we investigate the influence of the Mn substitution on hydrogenation and electrochemical properties of $Y_{0.88}Ni_{3-x}Mn_x$ ($0 \leq x \leq 1$) compounds. Indeed, for the close A_2B_7 family ($n=3.5$), Mn substitution for Ni flattens the plateau pressure, and increases the electrochemical capacity^[4]. Moreover, AB_3 system structural study is interesting, as a sub-stoichiometry of Y is needed for obtaining single-phase AB_3 compounds. To understand this, density functional theory calculations have been coupled to advanced structural characterization using synchrotron X-ray diffraction at SOLEIL/Cristal beamline and neutron diffraction at ILL/D1b beamline. Joint Rietveld refinement have been performed to determine the location of Mn atoms. The lattice parameter a keeps almost unchanged while c decreases with Mn content up to $x=0.3$. This anisotropic behavior is directly correlated to the site-occupation of Mn atoms.

The electrochemical capacity of these alloys as negative electrode material was tested in pyrrolidinium acetate [Pyr][Ac] and KOH electrolytes. A capacity of 275 mAh.g^{-1} was obtained for $x=0.44$, higher than that obtained in KOH for this material. For all compositions, the capacity decreases drastically over cycling in KOH while is stable in [Pyr][Ac], proving the interest of coupling AB_3 compounds and PILs. The correlation between structural and electrochemical properties will be discussed.

References:

- [1] T. Kohno *et al.*, J.Alloys Compds 311(2000) 5-7
- [2] V. Charbonnier *et al.*, J.Phys.ChemC 119(2015) 12218-12225
- [3] N. Chaabene *et al.*, J.Power Sources 574(2023) 233176_1-7
- [4] N. Madern *et al.*, Energies 13(2020) 3470_1-17

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