

Investigation of structural evolution and hydrogen sorption properties of $Y_{0.9}Ni_{2-y}Mn_y$ ($0.1 \leq y \leq 0.5$) alloys with C15 Laves phase structure

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AB_2 -type alloys (A = Rare Earth or Early Transition Metal, B = Last Transition Metal) with the C15 Laves phase structure have more tetrahedral sites than AB_5 and AB_x ($3 \leq x \leq 3.8$) alloys. It allows to accommodate more hydrogen and provide high hydrogen storage capacity [1]. YNi_2 does not crystallize in the classic C15 Laves phase structure but in a vacancy-ordered superstructure with composition $Y_{0.95}Ni_2$. Upon hydrogenation YNi_2 suffers from hydrogen-induced amorphization or disproportionation [2] and becomes amorphous upon hydrogen absorption of 3 H/mol [3]. Thus, its reversible capacity and cycle life are low.

This work aims at stabilizing YNi_2 upon hydrogen cycling by substituting Mn for Ni in pseudo-binary $Y_{0.9}Ni_{2-y}Mn_y$ ($0.1 \leq y \leq 0.5$) compounds. Surprisingly, all these compounds crystallize in the classic C15 structure while maintaining Y sub-stoichiometry. As $Y_{0.95}Ni_2$ is stabilized by vacancy ordering of Y sites, one may expect that $Y_{0.9}Ni_{2-y}Mn_y$ compounds crystallize in the C15 phase with higher content of disordered Y-site vacancies.

Neutron diffraction measurements have been performed to determine atom occupancy in $Y_{0.9}Ni_{2-y}Mn_y$ compounds. Rietveld refinement of neutron data evidences no vacancies in the A -site and Mn occupation not only in the B site but, surprisingly, also in the A site. The fact that Mn can behave as an A -type atom has been previously observed in the $ZrMn_{2+x}$ and $TbNi_2Mn$ systems [4,5]. The hydrogenation properties of $Y_{0.9}Ni_{2-y}Mn_y$ compounds and their relationship with the crystal structure will be discussed.

References:

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