## SPECIFIC PATHWAYS FOR SYNTHESIS OF CARBONACEOUS – NaTi<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> COMPOSITES AND APPLICATION AS NEGATIVE ELECTRODE MATERIALS IN AQUEOUS Na-ION BATTERIES

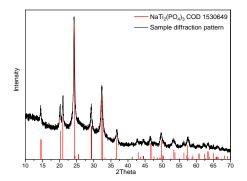
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The most recent Nobel Prize in Chemistry was awarded to John Goodenough, M. Stanley Whittingham and Akira Yoshino "for the development of lithium-ion batteries". Although Li-ion batteries (LIBs) are revolutionary and surely are the main electrochemical energy storage technology today, there are ongoing issues with these batteries, caused by the natural scarcity of lithium and cobalt; furthermore, some issues remain concerning the safety due to the use of organic electrolytes. Therefore, alternative technologies are being intensively explored, one of them being the sodium ion battery (SIB) [1].

The aim of our research is to develop suitable electrode materials for aqueous SIBs that would ensure high capacity, long lifespan and superior cycling performance. One material that stands out in its suitable electrode potential, high theoretical capacity and ionic conductivity, structural and electrochemical stability is the Sodium SuperIonic Conductor (NASICON)-structured  $NaTi_2(PO_4)_3$  (NTP) [2]. We present the detailed aqueous sol-gel synthesis pathway of carbonaceous-NTP composites and a thorough analysis of prepared samples.

The main issue with aqueous sol-gel synthesis of NTP is the stabilization of titanium precursor (in this case-titanium tetraisopropoxide (Ti(<sup>i</sup>OPr)<sub>4</sub>)), as titanium alkoxides are irreversibly hydrolyzed in water. A procedure to make a stable organometallic titanium complex is the reaction of Ti(<sup>i</sup>OPr)<sub>4</sub> with lactic acid [3]. After preparing the stable titanium precursor, different synthesis pathways were employed to prepare the NTP powder. The obtained NTP powder (the active material for electrode preparation) were analyzed by powder X-ray diffraction (XRD) (Fig. 1a), Scanning electron microscopy (SEM) and Fourier Transform infrared spectroscopy (FTIR). The electrodes were prepared by mixing the synthesized active materials (carbonaceous-NTP composites) with conducting carbon filler and polyvinylidene fluoride binder in the ratio 7:2:1 (using N-methyl-2-pyrrolidone as the solvent) and casting the slurry in a uniform layer on aluminum foil. The electrochemical properties of prepared electrodes were characterized in the inhouse built three-electrode bottom mount flat sample cells using cyclic voltammetry and galvanostatic charge/discharge cycling (Fig. 1b).



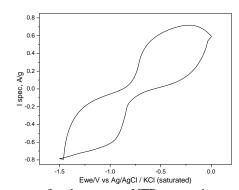


Fig.1 a) XRD pattern; b) Cyclic voltammetry of carbonaceous-NTP composite

In our presentation, we will analyze in detail the pathways of carbonaceous-NTP synthesis, present the obtained material phase-purity and morphology analysis results and further investigate the differences in electrochemical properties and their co-dependence with employed synthesis pathways.

<sup>[1]</sup> Palomares, V., Serras, P., Villaluenga, I., Hueso, K. B., Carretero-González, J., & Rojo, T. Na-ion batteries, recent advances and present challenges to become low cost energy storage systems. Energy Environ. Sci. (2012) 5, 5884.

<sup>[2]</sup> Wu M., Ni W., Hu J. Ma & J. Wei. NASICON-Structured NaTi2(PO4)3 for Sustainable Energy Storage. Nano-Micro Lett. (2019) 11:44.

<sup>[3]</sup> Ohya T., Ito M., Yamada K., Ban T., Ohya Y. & Takahashi Y. Aqueous Titanate Sols from Ti Alkoxide-α-Hydroxycarboxylic Acid System and Preparation of Titania Films from the Sols. Journal od Sol-Gel Science and Technology 30, 71-81, 2004.

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