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Skolkovo Institute of Science and Technology

Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Natalia Katorova

PhD Program: Materials Science and Engineering

Title of Thesis: The effect of selected electrode-solution interactions on the potassium-ion battery electrochemical performance

Supervisor: Professor Keith Stevenson
Co-supervisor: Professor Artem Abakumov

Name of the Reviewer: Aksenov Dmitry

I confirm the absence of any conflict of interest (Alternatively, Reviewer can formulate a possible conflict)	**Date: 02-10-2022**

The purpose of this report is to obtain an independent review from the members of PhD defense Jury before the thesis defense. The members of PhD defense Jury are asked to submit signed copy of the report at least 30 days prior the thesis defense. The Reviewers are asked to bring a copy of the completed report to the thesis defense and to discuss the contents of each report with each other before the thesis defense.

If the reviewers have any queries about the thesis which they wish to raise in advance, please contact the Chair of the Jury.

Reviewer's Report

The thesis is well written, the used methodology is well described and justified. Overall, the goal and tasks are clear, but I suggest to write them out explicitly in the Introduction section. The obtained results are highly important from the practical point of view paving the way to commercial K-ion technology. At the same time the in-depth investigation of the reasons for improved performance of high-concentrated electrolytes, mechanism of K storage in hard carbon and impact of VC on cyclability have fundamental value. All results are published in high-impact international journals highlighting the significance and quality of the thesis work.

The work has minor issues that should be further clarified and answered:

- A summary for section "Effect of synthesis conditions on Hard Carbon properties" should be added at the end of the section.

- What is the best combination of synthesis conditions for hard carbon? Please add in summary.

- Some conclusions or connection with the next section should be added for section "Aluminum current collector passivation" otherwise it is not clear for what reason the measurements were performed

- What is the solubility of KPF6 in diglyme-based electrolytes?

- The performance of electrodes in high-concentrated electrolytes is improved, but what about the conductivity of such electrolyte and its performance at lower temperatures?

- Fig. 5.11 What was the c-rate during cycling?

- p 76. The KVOPO4 cathode, exhibiting the KTiOPO 4 -type structure, could serve as the cathode in K cells.

The capacity of this cathode is 50 mAh/g. It it too small for realistic cells, please rewrite.

- p89. - However, the specific reasons for the observed HC behavior in the concentrated

electrolytes should be clarified in the future research.

What kind of specific reasons? Please clarify.

- p90 No significant difference was observed for K half cells comprising HC negative electrode

and diglyme-based electrolyte with various KPF 6 salt contents in Chapter 5. However, the full cell

demonstrated the improved performance with pre-loaded HC electrodes in electrolytes with

enhanced concentrations that could arise from the anion-derived solid electrolyte interface (SEI)

layers 172

Why was the effect observed only in full cells?

- p92 The HAADF-STEM characterization coupled with STEM-EDX mapping of 30 times cycled

HC electrode surface reveals the absence of thick SEI formation (Figure 6.4).

Could it be that the layer was washed out during specimen treatment? Usually, such SEI are observed with cryo-TEM. If there is no SEI, how to explain the improved behaviour in high-concentrated electrolyte?

- p111 What is the optimal concentration of VC? Why it is so? Please add to the summary.

Please correct typos:

- p77 The presence of ““nanosphere”” particles

- p113 in K-ion cells цшер the highest electrolyte concentrations

The mentioned issues do not reduce the quality of the thesis work. Therefore I recommend that the candidate should defend the thesis by means of a formal thesis defense.

Provisional Recommendation

☒ *I recommend that the candidate should defend the thesis by means of a formal thesis defense*

☐ *I recommend that the candidate should defend the thesis by means of a formal thesis defense only after appropriate changes would be introduced in candidate's thesis according to the recommendations of the present report*

☐ *The thesis is not acceptable and I recommend that the candidate be exempt from the formal thesis defense*

Assistant Professor of Skolkovo Institute of Science and Technology, Dr. Aksyonov D.A.		02.10.2022