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Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Vahid Ramezankhani

PhD Program: Materials Science and Engineering

Title of Thesis: Design of potassium-ion batteries using novel organic electrode materials

Supervisor: Assistant Professor Stanislav S. Fedotov

Co-supervisor:

Name of the Reviewer: Evgeny V. Antipov, Full Professor

I confirm the absence of any conflict of interest	 **Date: 30-09-2023**

Reviewer's Report

The dissertation "Design of potassium-ion batteries using novel organic electrode materials" tackles a pertinent issue in the field of battery technology, which is the development of novel organic electrode materials for potassium ion batteries. It demonstrates a high level of quality and sophistication in terms of its research and presentation and shows a clear understanding of current industry needs on environmentally benign, low-cost materials and an attempt to address them through research.

The thesis is well-structured, consisting of six chapters that flow logically from one to the next. Here's an evaluation of the thesis quality and overall structure. The introductory chapter effectively highlights the novelty of the work and the importance of introducing novel electrode materials for potassium ion batteries. It also draws attention to the economic implications of this technology. The inclusion of Chapter 2 - Background is dedicated to battery specifications and a discussion of both organic and inorganic electrode materials, which is crucial for providing the necessary context for the research. This chapter helps to understand the state of the art before delving into the new materials being proposed. Chapter 3 - Research Objectives gives a clear elaboration of research objectives. It provides the specific goals and aims of the study, which is essential for maintaining focus throughout the research. Chapter 4 - Research Methodology describes the research methodology, including cell design and characterization tools, it ensures transparency in the research process to reproduce and/or

understand the experiments conducted. Chapter 5 - Results and Discussions presents results and engages in discussions allowing for a detailed exploration of findings, separating the raw data from interpretation. Chapter 6 - Conclusion provides a concise summary of the entire thesis and its implications, grasping the significance of the research. Appendices present extra, supplementary information without cluttering the main narrative. The inclusion of appendices further enhances the overall quality of the thesis.

In summary, the dissertation demonstrates a strong structure, beginning with a compelling introduction and progressing through well-organized chapters that cover all essential aspects of the research. It appears to be a well-thought-out and well-executed piece of research.

The relevance of the dissertation's topic to its actual content is highly evident and well-maintained the research. The title of the dissertation, which focuses on exploring K-ion storage using novel organic electrode materials, accurately reflects the core theme and objectives of the study. Here's a breakdown of the alignment between the topic and the content:

1. Title and Core Theme: The dissertation's title centers on the exploration of potassium-ion (K-ion) storage, which establishes the primary subject matter of the research. The K-ion storage is a significant and growing area of interest in the field of energy storage, particularly for its potential applications in batteries. This matches well with the contemporary relevance of sustainable energy solutions.
2. Specific Material Selection: The author's choice to delve into a specific type of organic electrode material, namely carbonyl-containing organic redox-active materials, shows a clear connection between the topic and content. This specific material selection suggests a deep exploration of the K-ion storage intricacies within this particular context.
3. Challenges Addressed: The relevance of the topic is further confirmed by the author's acknowledgment of the challenges associated with the chosen family of materials. This recognition demonstrates a thorough understanding of the complexities and limitations associated with these materials in the context of energy storage.
4. Innovative Solutions: Importantly, the dissertation's content goes beyond merely acknowledging challenges. The author systematically addresses these challenges by proposing the development of more advanced molecular structures or the implementation of new electrolyte formulations. The dissertation demonstrates a proactive approach to overcoming

the obstacles related to carbonyl-containing organic redox-active materials. This aligns perfectly with the broader goal of advancing the field of K-ion storage for practical applications.

In summary, the dissertation's topic and content are closely aligned, demonstrating a clear and logical transition from the overarching theme of K-ion storage to the specific focus on carbonyl-containing organic redox-active materials. The author's deep understanding of the challenges associated with this material family and the innovative solutions proposed to mitigate these challenges underline the dissertation's relevance and significance in the field of energy storage and battery technology.

The methods discussed in the dissertation, particularly those related to battery assembly, electrode preparation, and mechanism investigation, are highly relevant and integral to the overall research. They facilitate the exploration of the novel organic electrode materials' applicability for K-ion batteries. This methodological rigor contributes to the dissertation's strength and its potential impact on the field of energy storage and battery technology.

The scientific significance of the results obtained in this dissertation is substantial and in line with the international standards and the current state-of-the-art in the field of energy storage. Chapter 6, which compares the obtained results with the record-breaking outcomes reported in scientific publications, serves as an evidence of the research's high quality. The author's conclusion that the obtained results rank among the best in terms of cyclability, energy density, and discharge capacity is particularly noteworthy. Such findings hold great scientific significance as they not only validate the effectiveness of the novel organic electrode materials for potassium-ion batteries but also contribute to advancing the field by pushing the boundaries of performance. Achieving results that compete with or surpass those reported in leading scientific publications emphasizes the dissertation's relevance and its potential to influence the development of sustainable energy storage solutions on an international scale.

The obtained results hold significant relevance to practical applications, particularly in the field of potassium-ion batteries. The discovery of an optimal molecular structure for use as an anode material in these batteries is a crucial step towards the development of more efficient and sustainable energy storage solutions. Furthermore, the successful optimization of the electrode composition, with 85% by weight dedicated to active materials, is a notable achievement as it aligns with the practical requirements for real-world battery applications. Author’s recommendations for further optimization of this anode material family in potassium-ion batteries also signifies the practical applicability of the research, as they offer valuable insights for future advancements in energy storage technology. Overall,

the obtained results are not only scientifically valuable but also hold great promise for real-world applications in the development of more efficient and cost-effective potassium-ion batteries.

Worth noting, that the quality of Vahid's publications is exceptionally high, as evidenced by their placement in top Q1 journals, including "J. Power Sources" and "J. Materials Chemistry A." These prestigious journals are well-regarded in the scientific community and known for their rigorous peer-review processes and strict publication standards.

In anticipation of the upcoming thesis defense, it is evident that the dissertation has been meticulously prepared. However, as we approach the defense, there are a few noticeable issues within the results and discussion that merit further attention.

- Firstly, the rationale behind the selection of a G2-based electrolyte with a concentration of 2.2 M warrants clarification, as understanding the choice of electrolyte concentration is pivotal to the overall research context.
- Secondly, it would be valuable to explore the possibility of crystallization of the polymers investigated throughout the dissertation, as this could significantly impact their performance and applications.
- Lastly, addressing the question of whether OHTAP operates effectively when implemented as a cathode in a real full-cell battery is crucial.

These comments and questions are mostly minor and do not diminish the quality or undermine the novelty of the work. Considering the performance in original research achievements, I recommend the acceptance of his PhD thesis for formal defense.

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*