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Review

on the doctoral dissertation of Mrs. Yuhi, "Design and Characterization of $CsPbCl_3$ Perovskite-Based Anodes and Artificial SEI in Lithium Batteries", in the chemical sciences discipline

supervised by Prof. Władysław Wieczorek (Warsaw University of Technology, Poland)
and co-supervised by Prof. Robert Dominko (National Institute of Chemistry, Ljubljana, Slovenia

The report is based on the hard copy received on 12 November 2025, containing 106 pages, written mostly in English (formally required Polish translations included where necessary).

Manuscript of the doctoral thesis subjected to review aims to represent significant scientific and research achievements, demonstrating Mrs. Yuhi ability to conduct independent research and contribute original knowledge to the fields of applied electrochemistry and materials chemistry.

This report aims to provide a structured evaluation of the thesis, highlighting obvious strengths, identifying potential weaknesses, and offering constructive feedback for further improvement. The discussion in relevant chapters is more akin to peer advice than to criticism, as the manuscript presents the findings well-supported by the data and discussed with reference to relevant literature. Some of the results are already published, thus – reviewed.

I need to admit that the manuscript is very concise which is quite unusual for dissertations written in Poland, however, all necessary details and supporting information are included. In my opinion, this is advantageous in the context of thesis clarity and has a positive impact on its scientific value.

The structure of the thesis follows a typical pattern in Poland, with acknowledgements, a general introduction, a presentation of the hypothesis (objectives and scope), a literature review, and chapters discussing the relevant results. These chapters finish with conclusions and perspectives.

Briefly, the thesis is generally well-structured, with a logical flow from the introduction to the conclusion. Again, the research scope is clearly articulated, and the objectives are well defined. Each sub-chapter of the literature review builds upon the previous one, providing a coherent narrative that guides the reader through the research process in further parts.

In overall, Mrs. Yuhi dissertation focuses on the development and optimization of electrode materials for energy storage applications, mostly Li-ion batteries.

Key findings, claimed by Mrs. Yuhi, include:

1. **$CsPbCl_3$ perovskite as electrode materials**:

    - Mechanochemical synthesis of $CsPbCl_3$ allowed for preparing the active material, further processed to formulate the electrodes by coating method;
    - The material loading was 1.0 – 1.3 mg/cm^2 and varied dependently on the final electrode composition (ratio between active material, binder and conductive agent);
    - The structural analysis (XRD) demonstrated that the major phase is tetragonal perovskite with minor contribution of orthorhombic phase;
    - SEM/EDX analysis demonstrated that perovskite powder consist of agglomerate partcles with irregular shapes, with broad diamater spectrum (a few hundred nanometers to a few microns).

2. **$CsPbCl_3$ perovskite as electrode materials for Li batteries**:

    - Li||$CsPbCl_3$ half-cells with two different electrode compositions (active material content varying as 60:30:10 and 70:20:10) demonstrated the impact on electrode specific capacity (first discharge capacity ~600 mAh/g vs. ~500 mAh/g);
    - Selected potential window (0.05 V – 2.0 V vs. Li/Li$^+$) indicated that the material undergoes phase changes, further evaluated by XRD at certain potential values;
    - Decomposition of perovskite at high voltages (above 2.0 V) was confirmed by XRD and conversion/alloying mechanism was identified;
    - Subsequent charging/discharging cycles confirmed further decrease of the electrode capacity, however, after SEI stabilization the capacity decrease is less pronounced (~110 mAh/g);
    - In the long-term cycling, reversible Pb alloying/dealloying is found as governing mechanism.

3. **$CsPbCl_3$ -based artificial SEI as protector for Li-anode**:
    - Solution-based method for perovskite powder syntheiss has been proposed and structural characterisation confirmed the targeted compound presence;
    - Li-foil anodes have been coated by the perovskite layer with various loadings; electrodes with 2 mg/cm^2 loading demonstrated the lowest overpotentials and were used in further studies;
    - SEM/EDX analysis demonstrated that the thickness of protective layer is ~5 microns with homogenious distribution of the elements;
    - Electrochemical measurements (impedance) demonstrated lower impedance for perovskite-coated electrodes vs. bare Li-electrodes;
    - *Operando* EIS measurement demonstrated substantial differences in overpotential evolution during cycling, in favor to electrodes coated by perovskite layer;
    - Asymmetric configuration, with Li||Cu cells (with limited Li content, calculated to 4 mAh/cm^2) demonstarted that $CsPbCl_3$ layer contributes to improved Coulombic efficiency of the cells containing perovskite;

- Full cell investigation (with LFP cathode) demonstrated improved capacity retention over cycles, with ~99% capacity retained over 250 cycles at 1C current load

4. **$CsPbCl_3$ -based artificial SEI as a protector for graphite and Si anode**:

   - Graphite anode covered by perovskite layer demonstrated improved cyclability and capacitance retention; interestingly, SEM observation inidcated that bare graphite degradation does not directly correspond to the structural changes;
   - Silicon anode, despite enormous volumetric change during lithiation/delithiation, mainatined the capacity over 500 mAh/g over 150 cycles, with remarkable (yet less pronounced) drop in the initail cycles (up to 20 loops).

5. **General insights from all chapters**:
   - $CsPbCl_3$ could be obtained in various methods and its electrochemical activity is mainatained regardless the synthesis pathway;
   - $CsPbCl_3$ could be considered as interesting material for anode protection or so-called articificial SEI, to improve the electrode/cell electrochemical performance in long-term cycling.

Overall, the dissertation emphasizes the importance of synthesis control of perovskite powder, careful evaluation of measurement effects (employment of operando EIS), as well as advanced diagnostics to enhance the performance and understanding of perovskite-based electrodes for energy storage. It provides material-specific guidelines for Li- (and to some extent - Si) based electrodes and general insights for future electrochemical energy storage materials. I like and highly value these findings, as they demonstrate careful analysis of the obtained data and their discussion with relevant literature. Interesting insights into the 'artificial SEI' concept have been made.

Literature review

The literature review is comprehensive (122 items cited) and demonstrates a good understanding of the state of the art in the field. It spans electrochemistry, electroanalytics, and materials engineering, with smooth interconnections and very logical correlations between structure and performance. Mrs. Yuhi critically engages with relevant sources, identifying gaps in the literature and positioning their research accordingly.

However, the manuscript could benefit from a more explicit discussion of recent advancements to ensure the research remains at the cutting edge, given that the field has a large number of papers and advancements may not be readily apparent.

I also have certain remarks/questions on the thesis:

- SEI formation and its properties depend strongly on the current regime; I did not find relevant discussion on the optimization of current load or at least the comment on tentative impact of it on the perovskite performance
- How the perovskite presence impacts the electrode/electrolyte interplay? Is there any impact on electrolyte?
- Is there any volumetric change of the perovskite expected?

- What was the motivation for using $CsPbCl_3$?
- What is the targeted operating potential range for electrodes coated with perovskite?

Obviously, these remarks are of little significance in the light of the enormous and high-level work done here. I would also like to point out a very aesthetic way of presenting the information, with a set of good figures and self-explanatory graphics.

Methodology

The methodology is always very detailed and appropriate to the research objectives outlined in the relevant sections of the manuscript. The candidate provides clear justifications for their chosen methods (either in the summary or the chapter body that presents the part of the work), and the procedures are described with sufficient detail to allow reproducibility.

Results and analysis

The results are presented clearly, with effective use of tables and figures to illustrate key findings. The analysis demonstrates a high level of critical thinking, and the candidate draws meaningful conclusions from the data.

Discussion and conclusions

The discussion thoughtfully interprets the results in light of the research questions and the broader literature. The candidate successfully highlights the implications of their findings and suggests a direction for future research. The conclusions are well-supported by the data and analysis presented throughout the thesis. Supplementary materials are rich and really meaningful.

Originality and contribution

This thesis makes a significant original contribution to the field. The research addresses an important problem and offers novel insights that advance current understanding of charge storage processes in Li-based electrode materials.

The originality of the work is evident in both the research design and the interpretation of results.

Presentation and style

The writing is clear, concise, and generally free of errors. The thesis is well-formatted, and references are consistently cited.

Conclusion

Overall, this PhD thesis demonstrates a high level of scholarly competence and makes a valuable contribution to the field of electrochemical energy conversion and storage.

With minor revisions, the work will be further strengthened and ready for final defence.

The scientific accomplishments presented by Mrs. Yuhi are evident and deserve to be pointed out. I think the candidate is ready to take the next steps in a scientific career.

Finally, the candidate is commended for their dedication and the quality of their research.

Formally, I would like to claim that the thesis **fills all necessary criteria of Polish law valid at the time of review submission** (13.01.2026), and I recommend the file to be further considered at public defence for awarding the doctoral degree.

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