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Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Aleksandr Kurilovich

PhD Program: Materials Science and Engineering

Title of Thesis: Oxygen Reduction Reaction on Metal Oxides/Carbon Composite Materials

Supervisor: Professor Keith Stevenson

Name of the Reviewer: Professor Stanislav Fedotov

I confirm the absence of any conflict of interest	**Signature:**  **Date: 31-08-2020**

Reviewer's Report
The thesis entitled "Oxygen Reduction Reaction on Metal Oxides/Carbon Composite Materials" is devoted to multi-scale modelling of experimental data and treatment of experimental uncertainties for the processes of oxygen redox reaction in fuel cells. This work significantly contributes to the field of electrocatalysis since it opens up new insights on the ORR mechanism from the atomistic point of view and presents an integrated modeling approach that allows treating the electrochemical data obtained on a rotating disk electrode augmented by experimental uncertainties quantification. The thesis is of sufficient size, it contains 7 chapters, 41 figures, 6 tables, and the bibliography includes 216 references. Chapter 1 is the introductory part, which provides important general information of global energy demand and challenges tackled by fuel cells, compares briefly the computational approaches used for modelling the mechanism beyond fuel cells operation, describes the proposed modelling approach and postulates the aim of the current work. Chapter 2 represents necessary literature overview regarding different types of fuel cells, underlying physics and chemistry, oxygen reduction reaction (ORR) and its mechanisms, as well as description of computational methods, modelling approaches and uncertainties quantification. Chapters 3 and 4 are devoted to analysis of the experimental electrochemical data and modelling of the ORR for metal-based oxide materials using optimized DFT and MF-MKM approaches. Chapters 5 and 6 presents data and modelling approaches for experimental uncertainties treatment and quantification for the ORR mechanism.

Among key methods used in the current thesis are a group of computational techniques which are based on periodical or cluster DFT, and microkinetic modelling. These methods represent a powerful tool in many branches of energy-related sciences and were well-adopted to fulfil the aims of the current work. The applicability and reliability of these methods are beyond doubts. It is worth noting that such methods were first-time applied for some of the studied metal oxides. A key achievement of this work is seen in the development of the experimental errors' treatment approach for the electrocatalytic type of reaction and formulation of a modelling approach taking into account such uncertainties. Typically, the experimental errors in electrochemical assessment of electrocatalytic reactions are disregarded which might lead to incorrect interpretation of the electrocatalytic mechanism or its elementary steps. In this work, the focus was also placed on the revisiting of this essentially important part of data processing and modelling to better elucidate the ORR mechanism for a series of potential electrocatalytic materials.

The thesis is written with clear and professional scientific English. The contents of the thesis are published in 3 high-quality papers in medium-impact journals of the Q1 quartile in the field of electrochemistry and physical chemistry. One more paper is submitted and expected to be published in due course.

The detailed reading of the thesis provoked some comments listed below:

1. The most significant comment concerns the Chapters 2, 3, and 4 which lack logical connectivity both among them and with the following Chapters 5 and 6. From the first sight they might even look like unrelated parts of different research. The Candidate is encouraged to add more "glue" between the parts during the presentation to show the integrity of the work in whole and significance of each part in particular in addressing the problem posed.
2. The Candidate did not mention who performed the synthesis of different materials in all parts of the work.
3. The manuscript contains some technical misprints (the title of Chapter 4 on p. 74 has an error in the formula) and some grammatical incorrections (*e.g.* informal reduced forms of auxiliaries like "won't" (p. 41), "doesn't" (p. 52) or incorrect use of tenses).

However, these comments are minor and do not underrate the quality and novelty of the work. The Reviewer recommends the Candidate for 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*