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REPORT ON THE DISSERTATION ENTITLED "CARBON-BASED MATERIALES FOR HIGH PERFORMANCE ENERGY STORAGE DEVICES"

The basis for issuing the opinion on the doctoral dissertation of Jiaxin Li is the letter of Prof. Dr. Mirosława El Fray, Chairman of the Scientific Council of the Material Engineering Discipline of West Pomeranian University of Technology in Szczecin on August 16, 2023 (ZUT/RDIMat/12/2023)

A dissertation submitted by Mrs. Jiaxin Li *(hereafter referred to as the Candidate)* consists of a basis for awarding the Doctoral degree. The thesis has been conducted under the supervision of Dr. Hab. Xuecheng Chen, and it correlates well with the research activities carried out successfully in this team. The report, which I had a pleasure to provide, will be divided into separate sections describing various aspects of the thesis.

1) Thesis content and contribution to the field

The *Thesis* consists of 152 pages, and it has been written using a standard layout. It begins with the Declaration, followed by the Abstract, Streszczenie, Contents, the List of abbreviations, nine Chapters, the List of scientistic publications, Participation in scientific conferences and Patents. The thesis ends with list of References (a massive number of 320 items!!!).

The Chapter 1 is a literary essay containing a thematically relevant and concise introduction that guides the reader through the topic of electrochemical energy storage in a very accessible way. *The Candidate* describes lithium-ion batteries, their discovery, ingredients, mechanism of action, cathode and anode materials used. Subchapter 1.2 provides a detailed description of zinc-ion capacitors, as well as examples of such materials. The following subsections describe supercapacitors and porous carbon materials. The introduction ends with an announcement of the research conducted during the doctoral thesis. The promising properties of carbon nanomaterials and their composites (with metal oxides) were investigated and linked to a sustainable renewable fuels strategy to meet the demand for LIBs, ZICs and supercapacitors.

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Chapter 2 contains the results of the conducted research presented in the form of general descriptions of experimental procedures. Noteworthy, *The Candidate* used various analytical techniques such as SEM, HRTEM, XRD, TGA, XPS, BET method and DFT calculations was used to calculate specific surface area and pore size distribution of the carbon materials. The electrochemical measurements on LIBs, ZICs and supercapacitors were deeply discussed in experiments described in this *Thesis*. Moreover, these techniques are typically used in various scientific settings, which demonstrates the high commitment of the *Candidate* to carry out the research at the highest level.

Chapter 3 of this *Thesis* describes results and discussion of the recycling of real-world plastic wastes into Co_3O_4@void@C towards high performance lithium-ion batteries. The *Candidate's* research work aimed to find a highly efficient and effective strategy to convert real world mixed plastics (waste mask, waste plastic jar and foam sheets) into Co_3O_4@void@C in large scale. The research results have been published in a prestigious journal such as *ACS Applied Nano Materials* and provides an easy method for the design of high-performance anode materials of LIBs by recycling of the plastic wastes.

Chapter 4 narrates the search for high performance anode materials of lithium-ion batteries from N-doped carbon coated CNT@SnO_2 sword-sheath structures. It has been observed that the N-doped carbon layer significantly improve the electrochemical stability, but also effectively increase the conductivity of composites for better lithium storage. CNT@SnO2@CNx also has excellent performance and cycle stability.

In chapter 5 Jiaxin Li also detailed reports to recycle waste poly(ethylene terephthalate) (PET) into high-value PET-based metal organic frameworks (MOFs) and further convert it into porous carbon for green energy storage applications. In the study, a facile and cost-effective hydrothermal process was developed to direct recycle waste PET bottles into MIL-53(Al) with a 100% conversation, then the MOF-derived porous carbon was assembled into electrodes for

high-performance supercapacitors. It is with real pleasure that I note that the results of these studies described have already been published in reputable journal: *Green Energy & Environment*.

Chapter 6 reported the preparation and characterization a series of monodisperse ZIF-8 derived N-doped porous carbon materials by reaction temperature control and focus on the effects of particle size on the pore size distribution (PSD), specific surface area (SSA), functional group as well as the electrochemical performance for SC and ZICs. Among all the ZIF-8 based porous carbon, CZ-150 possessed the optimal particle size that can effectively increase the ion diffusion coefficient for fast electrolyte ion transport caused by the shorten ion transport distance and fast ion diffusion rate.

Chapter 7 focuses on the preparation of 2D nanosheets with adjustment pore size distribution and N, P co-doped designed and achieved by one-step process. The enhanced porosity originates from its hierarchical nanostructure, where mesopores serves as the effective channels for the fast mass transport and storage to micropores. *Ad interim*, its N, P co-doped could further lower the resistance between the electrolyte and carbon materials and boost the chemical adsorption of Zn ions on the electrode surfaces, leading to an enhanced electrochemical performance.

The high quality of the research tasks described in Chapter 8 was recognized by the reviewers of the *Diamond and Related Materials* journal, which is commendable for the Doctoral Student, not forgetting, of course, the leading role of the research Supervisor. The last part of the work was devoted to effective and simple a method for selective production of N-doped porous carbon from freeze-drying was proposed freeze-dried banana sheets with adjustable pore size to match electrolyte ions for high performance supercapacitor.

The research results described in chapters 4, 6 and 7 constitute ready material for new publications. Perhaps one of them has already been published?

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2) Quality of the work

The dissertation showcases the *Candidate's* deep comprehension of the fundamental principles of electrochemistry of carbon-based materials, as demonstrated by an in-depth analysis. The scientific novelty and effectiveness of the dissertation's objectives are supported by three experimental publications. The quality and originality of the results underwent meticulous assessment by external reviewers and the editorial board. Therefore, it is unnecessary to reiterate and re-evaluate the data's quality by summarizing them again.

3) Presentation

Despite not being a native speaker, the *Candidate's* presentation is excellent. The schematic representations of the obtained results are of high quality, with well-designed figures and graphs. The thesis descriptions and format are clear, and the experimental data are presented briefly and precisely. It is worth mentioning that the candidate has presented their research findings orally at international conferences on two occasions, indicating their extensive experience as a chemist with a curious and open-minded approach to research.

4) Originality

The scientific novelty presented in this dissertation undoubtedly lies in the development of a simple and highly effective electrochemical energy storage materials.

5) Summary

The *Thesis* contains original and valuable scientific results of development of new modificated carbon-based ensembles, which is an important contribution to material chemistry. The thesis represents a great deal of work. I find the dissertation of Mrs. Jiaxin Li as conceptually very interesting. The work was properly conducted and reported with due care and

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competence, and the analysis of the experimental results is insightful. *The Thesis* represents a substantial amount of work.

The exceptional scientific activity of the Candidate deserves to be emphasized. In addition to three publications included in the dissertation, he is the co-author of three other papers in recognized journals (*Environmental Sciences Europe* (2) and *Renewable Energy*).

The excellent reputation of the *Candidate's* Supervisor, and the research team in which the work was performed had set the expectation for this *Thesis* at the impressive level. And indeed, as expected, the quality of this *Thesis* is not disappointing at any point. I believe that the scope of work presented in this *Thesis* is extremely important and useful not only from a purely scientific point-of-view, but also due to the high application potential of renewable materials, the industrial use of which will probably increase in the coming years.

In view of the above, I believe that the dissertation meets all the requirements and I recommend the Scientific Council of the Material Engineering Discipline of to the West Pomeranian University of Technology in Szczecin to award the candidate the PhD degree. At the same time, due to the high quality of experimental results contained in the doctoral dissertation and the innovative ideas, which will certainly impact the development of the studied field, I recommend for its distinction. It is important to note that the design of smart carbon nanocomposite architectures can provide practical guidance for the preparation of high-performance materials for electrochemical energy storage, and the dissertation under review includes a wide spectrum of such examples.

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