

**Referee's comment of doctoral thesis submitted by
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***Fabrication of flexible supercapacitors with high
electrochemical performance***

The thesis aim was preparation of flexible supercapacitors with high electrochemical performance. Flexible electrochemical capacitors or ultracapacitors have attracted increasing attention of research community for developing advanced energy storage with high capability, long cycle life, low cost, and easy fabrication. Recent effort was concentrated on developing flexible, carbon-based and all-solid-state supercapacitors with different structure and also performance characteristics. From this point of view is aim of the thesis actual and attractive. Flexible energy storage devices are essential for the development of flexible and wearable electronics. The developed supercapacitor contained flexible electrodes based on reduced graphene oxide (RGO) hydrogel films modified by pseudo-active materials such as conducting polymer - polyaniline (PANI) and manganese dioxide (MnO_2). Suitable gel electrolytes were prepared at the point of view to match the construction and electrochemical performance of flexible supercapacitors. Finally, the construction of flexible supercapacitor was designed and investigated using various methods and techniques.

The presented PhD thesis has a standard content, after short introduction general properties of electrochemical capacitors and flexible supercapacitors (SC), including mechanical theory, structural design and optimization of such devices are discussed. This part summarized recent progresses in the development of high-performance supercapacitors based on carbon nanomaterials. Also various rational concepts for SC engineering to improve the device performance for a large variety of potential applications, ranging from consumer electronics through wearable optoelectronics is introduced. This is illustrated by a comprehensive list of references, which contains 162 papers. From this, 90 items are cited in the introduction part. The other references are used in discussions, including one reference no. 98 of the PhD thesis author from previous work.

Aims of the work are explicitly formulated.

Experimental part is described adequately. Formally, the PhD thesis has a very good level, including using graphic layout, illustrations, microphotographs and graphs, mainly presented in colour with precious explanation of all Figures. It is all written in English, but contains the title, summary, and keywords in Czech. The English text is well understood with only minor stylistic or grammatical errors, e.g. *red lacer* in Fig. 6.13.

Conclusions are composed with main findings of thesis.

I have also some comments and questions to PhD thesis.

1. In the Experimental part of the thesis it is described, that fabrication of flexible supercapacitor using PANI/RGO/G hydrogel films was done using polyethylene terephthalate (PET) substrate, but fabrication of the flexible supercapacitor using MnO₂/RGO/G hydrogel film on polyethylene (PE) films (40 µm, thickness). What is the main reason of two polymer substrate using? Did you used specially pre-treated PE and PET films at it is usual for ink-jet printing electronic devices as organic field effect transistors and others?
2. In part 6.2, X-ray photoelectron spectroscopy (XPS) results are presented, including *Figure 6-2. XPS spectra of GO, C1s region and survey scan*, only for prepared graphene oxide with the results that in GO percentage of carbon and oxygen are 70.7 % and 25.8 %, respectively. First of all it should be emphasized that it is atomic %. For the supercapacitor preparation also information about reduced GO will be useful, as well as comparison with original graphite and content of C and O. Have you done this experiment?
3. Ionic conductivity of hydrogel based on PAA/SA-Ca/BC was measured by electrochemical impedance spectroscopy (EIS). Did you measured electrical conductivity of RGO and RGO/G and modified RGO/G with polyaniline?
4. One of PhD thesis conclusion is for the future work to find compromise between flexibility and the electrochemical performance of a flexible SC. What is your plan for the continuation of SC research?

Conclusions

Finally, I would like to summarize that the author of this dissertation has demonstrated the ability of independent creative scientific work. The work contains a large number of interesting original experimental results which are clearly presented and discussed. The aims of submitted PhD thesis have been fulfilled. H. Fei published two scientific publications in international scientific journals, he also presented obtained results in the international conferences. Therefore, after answering questions and successful defence of the thesis, I recommend to award **Haojie Fei** the degree

“Philosophiae doctor”.

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