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UNIVERSITÄT
JENA

Institute for Technical Chemistry and Environmental Chemistry Center for Energy and Environmental Chemistry Jena (CEEC Jena)

Universität Jena · Einrichtung · 07737 Jena

Prof. Dr. Andrea Balducci
PROF. für Angewandte Elektrochemie

Philosophenweg 7A
07743 Jena

Telefon: +49 (0) 3641 948464
Telefax: +49 (0) 3641 948402
E-Mail: andrea.balducci@uni-jena.de

Dr. Tomasz Śliwa
Poznan University of Technology
Faculty of Chemical Technology
ul. Berdychowo 4, 60-965
Poznan (Poland)

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Report on the doctoral thesis of Ms. MSc eng. Paulina Bujewska
Redox activity of pseudohalides in electrochemical capacitor application

The work described in the thesis of Ms. Paulina Bujewska, is dedicated to the investigation of redox activity of pseudohalides in view of their use as electrolytes in electrochemical capacitors (EC). EC are nowadays considered among the most important electrochemical energy storage devices, and they are used in a large number of applications. However, in order to further extend the use of these devices, an increase of their energy density, which is significantly lower than that of lithium-ion batteries (LIBs), appears needed. In this context, the development of innovative electrolytes appears of great importance, as this component is determining the operative voltage of EC an, thus, their energy. Taking these points into account, the PhD of Ms. Bujewska is therefore addressing a very actual research topic.

The thesis, which is cumulative, is divided in 3 chapters. The first chapter is reporting an overview about the components and properties of EC. Particular attention is dedicated to the electrolyte, which is the main subject of this work. The chapter is well written, and it is supplying important and clear information about these devices. This updated overview is very important to understand the motivation of the work carried out by Ms. Bujewska and the results reported in the following chapters. The second chapter is discussing the investigations about aqueous solutions and ionic liquid (ILs) containing pseudohalide anions (thiocyanate and selenocyanate). Ms. Bujewska investigated salts of various alkali metals, and considered in detail the influence of the salt concentration on the chemical-physical and electrochemical properties of the electrolytic solutions. She investigated the electrochemical behavior of lab-scale EC

containing two different types of electrode materials (entirely microporous and micro/mesoporous). Furthermore, she investigated to impact of these innovative electrolytes on different types of current collectors (stainless steel vs. gold for aqueous solutions and stainless steel vs. aluminum for the ionic liquid). She also modified potassium thiocyanate-based aqueous electrolytes with gold nanoparticles with the aim to improve the electrochemical capacitor performance. Ms. Bujewska was able to show that all considered electrolytes can be successfully used in EC, and that their use can contribute to increase the electrochemical performance of devices containing aqueous electrolytes. This can be clearly seen when the energy and power of the EC containing the standard lithium sulfate solution are compared to that of EC considered in this work. These studies have been published in 4 manuscripts (indicated as P1-P4). The third chapter is describing a study about the storage mechanism of EC containing the IL 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide as well as aqueous electrolyte based on lithium sulfate solution. These studies were performed utilizing an electrochemical dilatometer (coupled with the step potential electrochemical spectroscopy). In this case Ms. Bujewska used two carbon materials, having different physical properties, to gain information about the impact of material porosity on the electrical double-layer formation. These investigations on ILs suggested that the mechanism of charge accumulation depends on the electrode polarization (whether negatively or positively). The studies carried out utilizing the aqueous electrolyte aimed to understand how the pH of the electrolyte impacts the charge accumulation in porous carbon electrodes. These latter investigations suggested that the higher pH of the electrolyte, the bigger the contribution of hydroxide anions in the electric double-layer formation at the electrode polarized positively. The thesis is concluded by a clear summary and outlook.

I considering the thesis of Ms. Bujewska a very interesting work. Ms. Bujewska investigated several important aspects related to the development of innovative redox active electrolytes suitable for EC. The work that she carried out gave novel and important information about this important component, and these studies represent important contributions in the field of energy storage. Taking these points into account, and considering the fact that Ms. Bujewska published 5 papers, I consider this work as excellent. I would like to congratulate the student as well as her supervisor for the the work that has been carry out during the time of the thesis.

Sincerely yours,

Andrea Balducci