

University of Warsaw  
Faculty of Chemistry

## **Rafał Jurczakowski**

## **Załącznik III Appendix III**

### **Summary of Professional Accomplishments**

Warsaw, March 2016



**SUMMARY OF PROFESSIONAL ACCOMPLISHMENTS****1. Name and surname**

Rafał Jurczakowski

**2. Held diploma and scientific degrees – with the name, place and year of acquisition and title of Ph.D. dissertation**

1998 – M. Sc. In chemistry, Faculty of Chemistry, University of Warsaw.

2003 – Ph. D. in chemistry, Faculty of Chemistry, University of Warsaw, title of the dissertation: "Experimental and Theoretical Studies of Oscillations and Multi stability in Electroreduction of Selected Complexes of Nickel(II) at Mercury Electrodes".

**3. Previous employment in scientific institutions:**

March 2005 – to present adjunct, Faculty of Chemistry, University of Warsaw.

**4. Indication of achievements according to Art. 16, item 2, Act of 14 March 2003 – Law on Higher Education, the Law on Academic Degrees and Title and Degrees in Art (Journal of Laws No 65, item 595 with further amendments):****a) Title of the scientific achievement:**

"Applications of Electrochemical Impedance Spectroscopy in Studies of Electrocatalytic Systems and Hydrogen Electrosorption Phenomena"

**b) Publications related to the achievement (habilitation thesis) included in Journal Citation Reports database (star indicates R. Jurczakowski as corresponding author):****1. R. Jurczakowski, A. Lasia**

Limitations of the potential step technique to impedance measurements using discrete time Fourier transform

*Analytical Chemistry*, 2004, 76, 5033-5038

**MNiSW: 45, IF<sub>2012</sub>: 5,695**

(my contribution of ca. 70 % - all numerical modeling, participation in data interpretation, participation in manuscript preparation)

**2. R. Jurczakowski, C. Hitz, A. Lasia**

Impedance of porous Au based electrodes

*Journal of Electroanalytical Chemistry*, 2004, 572, 355-366

**MNiSW: 30, IF<sub>2012</sub>: 2,672**

(my contribution of ca. 50 % - most of experimental results with data analysis and interpretation, manuscript preparation)

**3. R. Jurczakowski, C. Hitz, A. Lasia**

Impedance of porous gold electrodes in the presence of electroactive species

*Journal of Electroanalytical Chemistry*, 2005, 582, 85-96

**MNiSW: 30, IF<sub>2012</sub>: 2,672**

(my contribution of ca. 45 % - experimental results, plan and participation in data analysis, data interpretation, participation in the manuscript preparation)



4. B. Losiewicz, **R. Jurczakowski**, A. Lasia,  
Kinetics of Hydrogen Underpotential Deposition at Polycrystalline Platinum in Acidic  
Solutions  
*Electrochimica Acta*, 2012, 80, 292-301  
**MNiSW: 35, IF<sub>2012</sub>: 3,777**

(my contribution of ca. 25 % - participation in CV measurements, idea and carrying out the impedance measurements in hanging meniscus arrangement, participation in the interpretation of the frequency dispersion)

5. M. Słojewski, J. Kowalska, **R. Jurczakowski\***  
The influence of phosphorous content on hydrogen sorption in Pd-P thin layers  
obtained by electroless deposition  
*Journal of Physical Chemistry C*, 2009, 113, 3707-3712  
**MNiSW: 35, IF<sub>2012</sub>: 4,814**

(my contribution of ca. 75 % - the idea and research supervision, plan and participation in data analysis, data interpretation, manuscript preparation)

6. A. Januszewska, G. Dercz, A. Lewera, **R. Jurczakowski\***  
Spontaneous Chemical Ordering in Bimetallic Nanoparticles  
*Journal of Physical Chemistry C*, 2015, 119, 19817–19825  
**MNiSW: 35, IF<sub>2012</sub>: 4,814**

(my contribution of ca. 65 % - the idea, research supervision, plan and participation in data analysis, data interpretation, participation in manuscript preparation)

7. P. Połczyński, **R. Jurczakowski\***  
Extremely Fast Hydrogen Absorption/Desorption through Platinum Overlayers  
Journal of Power Sources, 2016, 305, 233-239  
**MNiSW: 45, IF<sub>2013</sub>: 6,217**

(my contribution of ca. 70 % - the idea, research supervision, plan and participation in data analysis, manuscript preparation)

8. A. Januszewska, **R. Jurczakowski\***, P. Kulesza  
CO<sub>2</sub> electroreduction at bare and Cu-decorated pd pseudomorphic layers: Catalyst tuning by controlled and indirect supporting onto Au(111)  
*Langmuir*, 2014, 30(47), 14314–14321  
**MNiSW: 35, IF<sub>2012</sub>: 4,384**

(my contribution of ca. 60 % - participation in the idea, research supervision, plan and participation in data analysis, manuscript preparation)

9. **R. Jurczakowski\***, P. Połczyński  
Impedance of Mediated Electrochemical Processes. Novel Impedance Element for Unequal Diffusivities.  
*Journal of Physical Chemistry C*, 2014, 118, 7980–7988  
**MNiSW: 35, IF<sub>2012</sub>: 4,814**

(my contribution of ca. 80 % - the idea, research supervision, analytical model development, participation in the development of new element and manuscript preparation)

10. P. Połczyński, **R. Jurczakowski\***  
Impedance as a Tool for Rapid and Complete Characterization of Electrocatalytic Systems Involving Redox Mediators  
*Electrochimica Acta*, 2016, 188, 882-887  
**MNiSW: 40, IF<sub>2013</sub>: 4,504**

(my contribution of ca. 60 % - the idea, research supervision, participation in data analysis and in simplified model development, manuscript preparation)



c) The aim of the above mentioned papers and description of the scientific achievement.

## Introduction

Impedance spectroscopy (IS) is a relatively new and powerful method allowing for the investigation of electrical properties of matter and relaxation phenomena with time constants ranging over nearly ten orders of magnitude. Impedance measurements can be performed with high accuracy in automatized and remotely controlled systems, however, a broader application of IS in material science and electrochemistry (EIS) is still impeded by the limited number of comprehensive explanations and theories describing different physicochemical systems. The method is often abused in the scientific literature, the major problems are related to an improper assignment of circuits/models to observed impedance features. In general, impedance technique requires also physicochemical information on a given system acquired by using other complementary methods.

The scientific achievement presented in this work is related to the use of impedance spectroscopy for kinetic and thermodynamic studies of (electro)chemical systems under both supported and unsupported (solid state) conditions. First publication [H1] deals with the methods of IS measurements and the application of Fourier analysis to obtain impedance spectra. Further works provide insight into the influence of the electrode geometry [H2, H3] as well as selected electrode reaction mechanisms [H4, H5] on the impedance characteristics of various physicochemical systems. Special emphasis has been placed on the hydrogen sorption phenomena. Electrode reactions involving hydrogen adsorption and absorption in metals are described in [H6] and [H7, H8, H9], respectively. Investigations of the hydrogen absorption in metals with classical electrochemical transient methods was described in separate publications [H7, H8]. Impedance spectroscopy has been used also for the characterization of nonmetallic hydrogen stores by monitoring dielectric properties in solid state during hydrogen release from hydrogen-rich ammonia borane and its alkali metal derivatives [H10].

## Impedance spectroscopy and impedance measurements

Impedance is the proportionality factor between potential and current signals in the frequency domain for a two-terminal circuit, which fulfill the criteria of stationarity, linearity and causality under investigated conditions. Hence, impedance can be defined as:

$$Z^*(\omega) = \frac{F[E(t)]}{F[I(t)]} = \frac{E^*(j\omega)}{I^*(j\omega)} \quad (1)$$

where the operator  $F$  denotes Fourier transform, whereas  $E(t)$  and  $I(t)$  stands for time dependent potential and current functions. In EIS method, usually a small potential perturbation is applied to an electrochemical system and the resulting current response is investigated. The most popular impedance measurements methods are Phase Sensitive Detection (PSD) and Frequency Response Analysis



(FRA). The main drawback of these techniques is related to a relatively long time needed for spectra acquisition in a wide frequency range [1]. This is particularly inappropriate for investigations of nonstationary systems, which evolve in time (e.g. due to adsorption of intermediates poisoning the electrode surface [2]). This problem may be overcome by the use of Dynamic Impedance Spectroscopy (DEIS) [3]. In DEIS technique, a Fourier transformation is used for impedance determination in a short period of time, so that the system can be regarded as stationary during spectrum acquisition. Due to very short acquisition time, DEIS can also be used for local impedance measurements and the investigation of the electrode surface inhomogeneities [4]. Potential perturbation for impedance analysis can be chosen arbitrary, however, usually a sum of sine waves with odd harmonics are used as an excitation signal. The noise is always larger at low frequencies, which means that in order to get better results, one should use larger amplitudes at lower frequencies while they could be lower at higher frequencies [5]. Another possibility is the use of a pulse perturbation because its Fourier transform contains a wide range of frequencies. In particular, Fourier transform of Dirac's  $\delta$  function contains all frequencies of equal amplitudes. Obviously, the application of such potential perturbation would violate the linearity condition, however, it is possible to apply a small potential step and Fourier transform its first derivative versus time, because the derivative of the step is the Dirac's  $\delta$  function [6]. This method enable acquisition of impedance spectra as a function of the electrode potential in potentiodynamic staircase voltammetry. This technique was applied in practice by Mochalov and Kolosnitsyn [7].

More recently, Yoo and Park proposed a new method based on the Discrete Time Fourier transform (DTFT) of the first derivative of the potential step function [8-10]. The authors used a very short data acquisition time and extrapolated the response to a wide frequency range using DTFT.<sup>1</sup> The authors claimed that since potential perturbation contains already all frequencies it is possible to obtain impedance spectrum even for extremely low frequencies within short period of time. Since other methods mentioned above are much more time-consuming, this technique would be very advantageous to instantaneous determination of the full impedance spectra, including information about very low frequencies, for systems changing in time. However, spectra obtained using this technique were different from the regular impedance spectra, as they did not contain any influence of the mass-transfer impedance. Problems related to this technique were discussed in the scientific literature [11, 12].

The aim of the paper [H1] was to verify the validity of the method proposed by Yoo and Park [8-10] and to establish the conditions at which it could be used. Theoretical simulations of current-time curves were performed for a simple ideally polarized electrode and a redox system in the solution. The numerically obtained impedances were compared with those theoretically predicted. It has been shown that the potential step method followed by differentiation of the current and FFT or DTFT may be used to obtain impedance spectra. In [H1] it has been shown that reliable spectra can be obtained only in the frequency range from  $1/(N\Delta t)$  to  $1/(2\Delta t)$ . Interestingly, in some cases, it is possible to continue the extrapolation to lower frequencies, however, no new information is obtained (see Fig. 10 in [H1]). Although the use of DTFT allows for simulation of the data in the whole frequency range, it is not possible to get more information than that already exist in the experimental current-time data. Extrapolation of impedances

<sup>1</sup> DTFT is a form of Fourier analysis that produces continuous functions in the frequency domain.



below the smallest frequency present in the system, that is,  $1/(N\Delta t) = 1/t_{\text{total}}$  introduces artifacts. For example, on the complex plane of impedance for a simple diffusion–kinetic system two semicircles are obtained instead of a typical Randles behavior (semicircle followed by a straight line at 45°), see Fig. 8 in [H1].

The results presented in work [H1] points out that already spectra acquisition is not a trivial task and that experimentally obtained spectra should always be validated before further analysis. EIS is a very sensitive technique and must be used with care already at the stage of data acquisition. In general, various artifacts appears usually at very high and very low frequencies.

### **Electrode geometry – impedance of porous electrodes**

Porous electrodes are in the center of interest of practical applications of electrocatalytic systems, supercapacitors [13], batteries [14] and fuel cells [15]. In industrial use of electrochemistry, smooth electrodes are impractical because the electrodes must have large surface area in order to get high current densities. For this reason porous electrodes are applied in industrial electrolysis [16, 17]. Electrochemical impedance spectroscopy (EIS) is often used to characterize these electrodes and electrochemical reactions taking place on them. There is no strict boundary between rough surfaces and porous electrodes [18]. However, the porous electrodes are different from rough surfaces in the depth,  $l$ , and the pore radius,  $r$ , of the pores, for porous electrodes the ratio  $l/r$  is very large. It should be noticed that the determination of the surface roughness is a challenging problem in electrocatalysis [19].

First theories and models of porous electrodes were developed by Frumkin [20] and later by de Levie [21, 22]. Although, these models were derived for a single cylindrical pore, they were also widely used to describe real physical systems [23, 24]. In the models for porous electrodes, a part of the active surface area located outside the pores have not been taken into account. In [H2] the equivalent circuit has been proposed, in which this external surface has been included (see equivalent circuit - Fig. 8 in [H2]). It has been shown that the area of the external surface have strong influence on the impedance spectra (Fig. 9 in [H2]). Hence, by using equivalent circuit proposed in [H2] it is possible to determine the fraction of the porous electrode that behaves like a flat (or rough) surface and the internal surface fraction. A series of porous gold electrodes obtained by different methods, proposed in the same publication, were analyzed. The fraction of the flat surface was dependent mainly on the electrode geometry, in turn dependent on the preparation method. This model permits for the characterization of such electrodes, determination of the total surface areas with the above mentioned separation into the external and internal part. It should be emphasized, that the evaluation of the apparent (real or active) electrode surface of a solid polycrystalline electrode is a challenging problem in electrochemistry. Different methods of the effective surface area determination were proposed, but usually they lead to different results [19]. In the case of porous electrodes the situation is even more complicated because of the ohmic drops occurring inside the pores. Surface roughness, porosity, surface poisoning and inhomogeneous current distribution are only few other factors that significantly affect the apparent (active) electrode surface.



In the publication [H3], the impedance of the porous gold electrodes was investigated in the presence of the electroactive species. Two model one-electron reactions have been chosen, fast redox processes of hexaamineruthenium(III)  $[\text{Ru}(\text{NH}_3)_6]^{3+}/[\text{Ru}(\text{NH}_3)_6]^{3+}$  and quasi-reversible processes of tris(1,10-phenanthroline)cobalt(II)  $[\text{Co}(\text{phen})_3]^{3+}/[\text{Co}(\text{phen})_3]^{2+}$ . The results were compared to those obtained for polycrystalline gold electrodes.

The theory of the electrochemical processes taking place on the porous electrodes has been developed by Lasia [25]. This model predicts formation of two semicircles on the complex plane plots for a simple one-electron redox system. Basing on this model, it has been concluded in [H3] that for investigated porous electrodes, the electrochemical reaction takes place mainly at the outermost area of the pores and at the external surface outside the pores, while the surface area in depth of the pores remains inactive. This is due to current and concentration distribution within the electrolyte inside the pores. This effect is visible also on the cyclic voltammograms, on which the current peaks are much smaller than those anticipated on the basis of the real surface of the electrodes. For example, the faradaic current peak obtained for the Au electrode from Au-Al pressed powders in the presence of  $[\text{Ru}(\text{NH}_3)_6]^{3+}$  is at least 100 times smaller than expected taking into account roughness factor of this electrode ( $R_f = 410$ ) [H2, H3], see Fig. 2A in [H3]. These results point out that the surface geometry can be frequently a limiting factor in practical applications of porous electrodes in electrocatalytic systems.

### **Complex electrode mechanisms – impedance of electrocatalytic systems involving redox mediators**

Impedance spectroscopy can be very informative in mechanistic studies. The electrode mechanism usually have crucial influence on the impedance characteristics of the electrochemical processes. Usually, even very closely related mechanisms can be distinguished from impedance characteristics, hence, EIS can be used for fine tuning of the mechanism determination. Numerous impedance models have been so far developed for porous electrodes, reactions involving adsorbed species [26, 27], semiconducting systems [28-30], self-assembled monolayers [31-34] or biosensors [35] to name a few. Much less emphasis has been placed on electrochemical systems coupled with chemical homogeneous reactions. Among these, catalytic processes involving redox mediators are of particular relevance since they can be used in various important systems, in which electroinactive molecules are reduced or oxidized by using of the redox mediators carrying the electrons between the electrode and the target molecules. In these processes the electrochemical step (E) is followed by homogeneous chemical reaction regenerating depolarizer (C'). The so called EC' – type reactions are widely employed for mediated electrochemical oxidation of persistent environmental pollutants [36], catalytic hydrogen evolution [37], carbon dioxide electroreduction [38, 39], electrochemistry of nucleic acids [40], amino acids [41], enzyme oxidation [42], as well as for other bioanalytical applications.

In [H4] we have developed, for the first time, an exact analytical model for the impedance of catalytic processes involving redox mediators together with the potential dependence of all impedance parameters. The results obtained for a general case in the rigorous treatment provide a powerful



research tool that can be used in the analysis of catalytic systems with arbitrary kinetics. We have rigorously shown that the resistance of the charge transfer becomes potential-independent at far anodic or far cathodic potentials for catalytic oxidation or reduction, respectively. It should be stressed that the analytical solution will always have an obvious advantage over numerical approach since it represents a closed form of the solution and predicts the model behavior under any circumstances. Furthermore, the fitting procedure by the complex nonlinear least squares routine is far simpler for the impedance in an analytic form. On the contrary numerical solving can be challenging for particular systems. Moreover, the impedance element derived in this study can be easily incorporated into any equivalent circuit so that the impedance analysis can be performed by any electrochemist, not only by a specialist in the impedance spectroscopy. Besides, by using the analytical approach, the dimensionality of the problem is significantly reduced since fewer (minimal) number of parameters are used for the fitting procedure and all of them have explicit physical meaning. In [H4] it has been shown that the key parameters of catalytic systems such as both heterogeneous and homogeneous rate constants, and the ratio of diffusion coefficients may be readily obtained even from a single impedance spectrum.

Unusual impedance characteristics have been found for systems having unequal diffusivities of the oxidized and reduced forms of the redox mediator. Depending on the ratio of diffusion coefficients, at the complex plane the spectra are either markedly deformed or an inductive loop appears at the low frequency range [H4]. It is impossible to fit these spectra in the whole frequency range, neither with the Gerischer impedance nor with any other known transfer function. This behavior is related to a novel impedance element, derived in [H4], which reduces to Gerischer impedance when both diffusion coefficients are equal. Novel transfer function is defined by Equation (2)

$$\hat{Z}_{PJ} = \frac{\hat{Z}_\xi}{\sqrt{j\omega + k}} \quad (2)$$

Similar to Gerischer impedance, in which however the parameter  $\hat{Z}_\xi$  is a complex function described by:

$$\hat{Z}_\xi = Z_1 \frac{\xi + a}{1 + \xi a} + Z_{-1} \quad (3)$$

$$a = \frac{\sqrt{j\omega}}{\sqrt{j\omega + k}} \quad (4)$$

where  $\xi$  is equal to the square root of the diffusion coefficients ratio of both forms of the redox mediator. Equations (2) - (4) are valid for both catalytic reduction and oxidation, but the parameters  $\xi$ ,  $Z_1$  and  $Z_{-1}$  are different and they are defined by Equations 24 and 25 together with Equations 38 and 40 in [H4].

Characteristic frequency for  $\hat{Z}_\xi$  is a complicated function of  $\xi$ . For  $\xi \rightarrow \infty$ ,  $Z_\xi \rightarrow \sqrt{j\omega}$  and the characteristic frequency,  $u \rightarrow 0$ . Whereas for  $\xi \rightarrow 0$  there are two characteristic frequencies  $u \rightarrow 2 + \sqrt{3}$  and  $u \rightarrow 2 - \sqrt{3}$ . Another characteristic point is the limit of the real part of impedance for zero frequency,  $\hat{Z}'_\xi(\omega \rightarrow 0) = \xi$ . However, relatively little information can be drawn from the characteristic points with comparison to the non-linear least squares fitting, hence we have focused on the latter.

It is also important to note that the novel transfer function, by no means, can be decomposed into simpler elements. Moreover, we have shown that the correct values of the kinetic parameters for EC' reaction can be found only by fitting impedance spectra to this novel element (transfer function). The rate constants of the chemical process determined by using presently known transfer functions (impedance elements) are erroneous and differ from the actual (real) values by ~ 20 % assuming usual differences in the diffusion coefficients [H4]. It contains only one additional parameter with respect to Gerischer impedance. This transfer function can also have a broader significance since a variety of physically different systems may exhibit a universal mathematical behavior. For example, characteristic features for the novel element discussed in [H4] and can be found on the complex plane plots reported many times in the solid ionics. It should be noted that their origin is unexplained to date, see e.g. Fig. 11 in [43], other examples are also given in [H4]. Taking into account a rather limited number of currently available transfer functions, the novel impedance element may be applied in many different systems for CNLS fitting of experimental results possessing low frequency distortions from the Gerischer impedance. It should be emphasized that such transfer functions of purely empirical origin have been introduced by Boukamp [44] as a generalization of Gerischer impedance. The shape of the impedance spectra for  $\xi > 1$  is quite similar to the double fractal Gerischer introduced by Boukamp et al. [44], described by impedance equivalent to phenomenological Havriliak-Negami dielectric dispersion.

Figure 6 in publication [H4] represents results of fitting of the Gerischer and Havriliak-Negami (HN) impedance to theoretical impedance spectrum for  $\xi = 1,414$ . Figure 6 in work [H4] shows By using this element, some improvement in CNLS fitting has been achieved at lower frequencies down to 70 mHz; however, below this frequency, it is impossible to fully reproduce the impedance by using HN element.

The novel element has been recently added to commercially available software used for CNLS fitting ZView® software developed by Scribner Scribner Associates Inc. This software is based on fitting routine developed by Macdonald [45], however all functions are defined by the producer. The novel element has been implemented by Scribner Associates Inc. in extended elements as Polczynski-Jurczakowski impedance. The new version of the ZView® containing this element will be released in 2016.

The model for the EC' type processes has been used for investigation of Fenton reaction [H5], which is frequently used for industrial wastewater treatment containing phenols, dyes, and other organic substances [46, 47]. Despite its chemical complexity, Fenton reaction have been widely used as a model system for the EC' – type processes. To date, many of important physicochemical parameters for this reaction, such as heterogeneous rate constant,  $k_0$ , diffusion coefficients  $D_{\text{ox}}$ ,  $D_{\text{red}}$ , electron transfer coefficients,  $\alpha$  and  $\beta$ , have been determined only in the absence of hydrogen peroxide in the solution. However, these parameters may significantly change in a real EC' system (that is in the presence of  $\text{H}_2\text{O}_2$  and other species).

By using a simplified model, the homogeneous reaction rate constant, cathodic and anodic electron transfer coefficient, and both diffusion coefficients of the redox mediator were precisely determined by using the impedance spectroscopy solely. The majority of these parameters have been determined for the first time in the Fenton solution while others are in good agreement with previous literature reports.



Although a particular case of the Fenton reaction was discussed in [H5], the analysis provided in this work has more general meaning and can be applied to any electrocatalytic system, in which a redox mediator is used as an electron carrier.

### Hydrogen electrosorption

At platinum group metals (Ru, Rh, Pd, Os, Ir, and Pt) hydrogen reduction takes place at the potentials more positive than the hydrogen evolution equilibrium potential,  $E_{\text{H}^+/\frac{1}{2}\text{H}_2}^0$ . This so-called hydrogen underpotential deposition (H UPD) results mainly from strong interaction between hydrogen and the metal surface. Similar UPD reactions were observed for deposition of metals on metals, for thermodynamic and structural considerations see Ref. [48]. H UPD reaction in acid solution may be written as:



where M denote atom at the metal surface. The kinetics of these processes were studied by us for rhodium [49], platinum [H6] and iridium [50] by using impedance spectroscopy. From the literature review it is clear that contradictory results were obtained by different groups for hydrogen adsorption at platinum. This reaction is very fast and must be studied at high frequencies, which are inaccessible for potentiostats. Kinetics of hydrogen underpotential deposition (H UPD) reaction at polycrystalline Pt has been studied in 0.1 M  $\text{H}_2\text{SO}_4$  and  $\text{HClO}_4$  by using a twin electrode configuration in a large frequency domain up to 1 MHz. Impedance between twin electrodes was measured directly using the frequency response analyzer (FRA) while the potentiostat was used only for a dc polarization in external circuit (Fig. Fig. 2 in [H6]). Two resistors  $R$  of 100 k $\Omega$  were used to assure dc polarization of two electrodes at the same dc potential by the potentiostat (there is no dc current in the circuit). The amount of ac current possible in this case is  $10^5$  times smaller than that flowing through the current measuring circuit. Calibrated resistor  $R_{\text{sr}}$  of 1  $\Omega$  served for current measurements from the ohmic drop. Potentiometer  $R_d$  was used to divide the amplitude of the potential perturbation generated by the FRA (which in the usual EIS measurements is divided by the potentiostat). All the resistances were placed on the top of the electrodes. This electrical set-up allowed for elimination of the potentiostat time constant, which limits the impedance measurements at high frequencies, and minimized the solution resistance. These measurements allowed for determination of the charge transfer resistance,  $R_{\text{ct}}$ , double layer,  $C_{\text{dl}}$ , and adsorption,  $C_{\text{ad}}$ , capacitances as functions of the electrode potential. The directly measured and potential-dependent charge transfer resistances due to hydrogen electrosorption were very small. Taking into account the surface roughness, they ranged from  $0.0337 \pm 0.0007 \Omega \cdot \text{cm}^2$  at 0 mV to  $9.4988 \pm 3.3016 \Omega \cdot \text{cm}^2$  at 360 mV in 0.1 M  $\text{H}_2\text{SO}_4$  solution. The kinetics of the H UPD is very fast at polycrystalline Pt with the slope  $dE/d\log R_{\text{ct}}$  of  $0.122 \pm 0.009 \text{ V dec}^{-1}$ . Slightly higher values of  $R_{\text{ct}}$  were observed for Pt electrode in 0.1 M  $\text{HClO}_4$  solution. It has been found that the kinetics of hydrogen UPD is slightly faster in sulfuric than perchloric acid.

### Hydrogen absorption in metals

Palladium – hydrogen is a model system of hydrogen absorption in metals described for the first time by Graham 150 years ago [51]. Palladium exhibits a number of exceptional properties, which enable its application in a myriad of hydrogen technologies. Its applications include hydrogen purification, storage, detection, and fuel cell electrocatalysis. Palladium has the ability to absorb large volumetric quantities of hydrogen at room temperature and atmospheric pressure. The hydrogen absorption mechanism is still not fully understood. Two mechanisms are generally considered, namely, direct (one step) [52]:



or indirect (two steps) in which the adsorption process, described by Eq. (5), is followed by:



In this model, hydrogen adsorption at the metal surface is an independent process of hydrogen absorption. At similar potentials hydrogen evolution takes place according to Tafel or Heyrovsky mechanism. It should be emphasized that this is only the most general classification of the hydrogen electrosorption mechanism, which does not distinguish between not only adsorption sites H<sub>UPD(tetr)</sub> H<sub>UPD(otk)</sub> but also between under- and overpotentially adsorbed hydrogen H<sub>UPD</sub> i H<sub>OPD</sub>. The existence of the hydrogen absorbed in subsurface sites has also been reported [53]. Subsurface absorbed hydrogen differs in both properties and concentration from both *alpha* and *beta* phase hydrogen in the metal bulk. The amount of hydrogen in subsurface layer is dependent on the electrode potential (or partial H<sub>2</sub> pressure) but is greater than that in the bulk. Additionally it has been proposed that in the two stage mechanism the hydrogen adsorbed in UPD and OPD process were the precursors for hydrogen adsorption in alfa and beta phase, respectively [54].

Comprehension of the nature of the hydrogen absorption is of great fundamental and practical relevance. In work [H7] we have described the influence of the nonmetallic element on the hydrogen absorption in Pd-P alloys. We have reported electrosorption isotherms for hydrogen absorption in amorphous Pd-P alloys obtained by electroless deposition. The results show that the phosphorous markedly influences the hydrogen solubility. Increasing phosphorous content diminish hydrogen absorption at higher hydrogen pressures. In contrast, at lower hydrogen pressures, in the *alpha* phase, the hydrogen insertion in the alloy, when compared to both Pd and crystalline palladium phosphides, was enhanced nearly by order of magnitude [H7]. Impedance spectroscopy has been used to evaluate the surface roughness of the electrode. Because at low hydrogen concentrations the contribution from the adsorbed hydrogen can be considerable in the overall hydrogen content measured by electrochemical methods.

We have also investigated the influence of the crystal violet on the hydrogen absorption in the alloys. Crystal violet blocks hydrogen absorption but accelerated the hydrogen absorption. Enhanced hydrogen absorption in the alloy can find applications in electrocatalysis and hydrogenation reactions.

### **Enhanced hydrogen solubility in PdPt nanoparticles, stability of nanostructures**

Recently we have found that platinum nanoparticles can be prepared without surfactant additions [55]. This gave us an opportunity to study palladium-platinum nanoalloys in an “as received” state without oxidative decontamination procedures. We have investigated the hydrogen solubility enhancement in nanoparticles reported by Kobayashi et al. for Pd-Pt [56] and Pd-Rh [57] systems. The authors explained the solubility enhancement in terms of “nanosize-induced” effects for both systems despite rather large nanoparticles size in case of Pd-Pt alloys (~7 nm). Alloy homogeneity “at the atomic scale level” was claimed basing on X-ray powder diffraction and EDX imaging. The atomic structure of bimetallic catalysts influences their physicochemical properties and largely determine their activity in virtually all applications. Systematic design of bimetallic catalysts require a proper recognition of structural stability and chemical ordering at the atomic level. Currently in the scientific literature there is a large discrepancy between various segregation scenarios predicted theoretically and experimental results in which homogeneous nanoalloying is readily reported or a priori assumed. The work [H8] deals with spontaneous segregation and chemical ordering of Pd-Pt nanoalloys, which was evidenced by hydrogen absorption enhanced nearly by three orders of magnitude. A departure from the homogeneous state was also confirmed in XPS studies and by comparison of hydrogen absorption isotherms for bulk segregated alloys. The latter were previously described by Flanagan et al. [58-64]. In [H8] we have also discussed the experimental difficulties related to the segregation assessment on the atomic scale and we have point out the most common sources of related misconceptions.

In [H8] we have also reported the highest hydrogen sorption in Pd<sub>80</sub>Pt<sub>20</sub> alloy exceeding H/(Pt+Pd) = 0.5 at the ambient temperature and hydrogen pressure, which additionally emphasizes the importance of both surface purity and the segregation in electrocatalysis and hydrogen storage. Our results clearly show that phase segregation is an extremely important factor that is frequently overlooked in experimental studies yet quite crucial for practical applications.

Basing on the results obtained for palladium-platinum nanoparticles two important hypotheses can be made, namely: (i) platinum thin films at least at the nanoparticles surface are permeable for hydrogen and (ii) platinum overlayers on the segregated Pd-Pt surface are stable against hydrogen induced lattice migration. For testing these hypotheses, in work [H9] the hydrogen electrosorption in thin palladium films (50-1000 nm) was investigated at palladium electrodes covered with platinum monolayers. Similar catalysts have been previously used by Adzic as catalysts for oxygen reduction (ORR) [65-69] and fuel oxidation [70]. The authors emphasized unusual stability of such systems [71], however hydrogen electrosorption has not been investigated.



The results presented in [H9] for this model system show that the rates of the hydrogen sorption/desorption are orders of magnitude higher for platinized samples with respect to pure palladium. The highest absorption kinetics have been observed for Pd electrodes fully covered with 1 – 3 platinum monolayers. We have shown that the observed enhancement of the hydrogen absorption rate occurs also in the *alpha* phase and is not related to overpotentially adsorbed hydrogen because the surface coverage of the H-OPD at potentials +0.1V vs. RHE is negligible [72].

By means of electrochemical impedance spectroscopy (EIS) we have also shown that the process is diffusion limited at platinized Pd layers. Diffusion coefficient,  $D_H$ , determined in EIS, is two orders of magnitude higher than that previously reported for thin palladium films and approaches  $D_H$  for bulk palladium. The system stability after hydrogen absorption was assessed and the sorption mechanism was discussed. Surprisingly high durability of the platinized palladium enables its use in a variety of applications where fast and selective response in the presence of hydrogen is required.

EIS measurements in twin electrode configuration, similar as described in [H6], in a large frequency domain up to 1 MHz have been used to investigate the kinetics of hydrogen absorption in Pd-Pt systems. Impedance between twin electrodes was measured directly using the frequency response analyzer (FRA). The resistance of the charge transfer was of the order of  $0.02 \Omega \text{ cm}^2$  (Figure 1). It should be noted that a distinct Warburg impedance between 1 and 10 kHz has been observed for layers as thin as 50 nm (Figure 1).

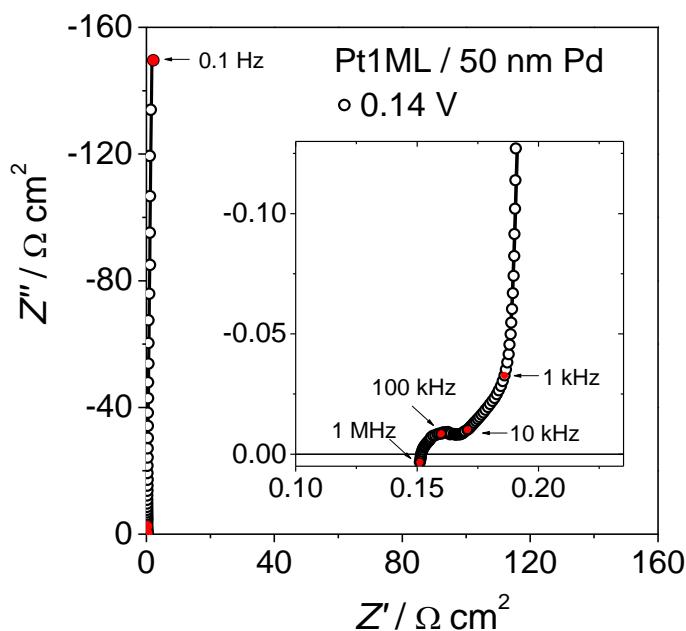


Fig. 1. Complex plane plot of impedance of 50 nm Pd electrode covered by one Pt monolayer measured in twin electrode configuration.

It should be noticed that the resistance of the charge transfer  $R_{ct} = 0.018 \pm 0.001 \Omega \text{ cm}^2$  at the potential +0.130 V is nearly two times smaller than that observed in the hydrogen UPD at the platinum electrode at the same potential [H6]. This testify for extremely fast rate of hydrogen absorption reaction. Similar

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enhancement in the kinetics of the hydrogen sorption phenomena have never been reported despite the fact that platinum has been already used as catalyst in hydrogen absorption reactions [73, 74]. However, the layer thickness used in these studies probably too large (at least several nanometers, i.e. > 20 ML) to enable hydrogen permeation.

### **Electrical properties of nonmetallic hydrogen stores studied by impedance spectroscopy**

Amidoboranes ( $MNH_2BH_3$ ) constitute an important group of solid state hydrogen stores with low temperatures of hydrogen desorption (onset at 70–100°C) and very high hydrogen content (7–14 wt.%) [75]. Amidoboranes are also considered as a promising source of hydrogen for low temperature fuel cells. Amidoboranes of numerous metals (Li, Na, K, Mg, Ca, Sr, Y) have been so far characterized. Solely potassium amidoborane evolve hydrogen without impurities [76]. Heating of other derivatives containing light alkali metals results in desorption of hydrogen polluted with ammonia. The ammonia desorption can be correlated with an endothermic process (40–60°C) preceding evolution of hydrogen (70–100°C).

We performed first solid-state impedance ammonia borane and its three alkali metal amidoborane derivatives. In work [H10] impedance measurements were combined with thermogravimetric analysis, scanning calorimetry, analysis of evolved gases, infrared absorption spectroscopy as well as  $^{11}B$  and  $^1H$  MAS NMR. We have performed EIS experiments by using a novel cell designed for the investigation of highly reactive powder samples without an exposure to atmospheric air and moisture. The powder sample is placed directly between the electrodes without any modifications (Fig. 1 in [H10]). By using the novel experimental setup it is possible to register high quality impedance spectra for very reactive samples under simultaneous control of both temperature and pressure.

Similar shapes of the complex plane plots of impedances were observed for all samples; the normalized impedance spectra are shown in Fig. 2 in [H10]. One semicircle followed by a spur at lower frequencies related to the charge accumulation at blocking electrodes was observed. This is typical for samples with predominating ionic and not electronic conductivity. The radius of the semicircle is strongly dependent on the temperature indicating changes of ionic conductivity. Complex permittivity  $\varepsilon^*(\omega)$  depends on linearly decoupled processes of instantaneous electronic polarization,  $\varepsilon_\infty$ , long range charge transport,  $\varepsilon_\sigma$ , and dielectric relaxation process,  $\varepsilon_{\text{relax}}$ . Assuming Cole-Cole relaxation function for the latter [77], the complex permittivity may be described by Equation (8).

$$\varepsilon^*(\omega) = \varepsilon_\infty + \varepsilon_\sigma + \varepsilon_{\text{relax}} = \varepsilon_\infty + \frac{\sigma_0}{i\omega\varepsilon_0} + \frac{\Delta\varepsilon}{1 + (i\omega\tau)^{1-n}} \quad (8)$$

This relaxation function can be represented by equivalent circuit shown in Fig. 1 in [H10]. An additional CPE described by parameters  $A_d$ ,  $n_d$ , represents impedance of the electrodes  $Z_{\text{el}} = [A_d(j\omega)^{n_d}]^{-1}$  (Warburg impedance for  $n_d = 0.5$ ) [78].

The dielectric properties of  $\text{NH}_3\text{BH}_3$  and freshly prepared alkali metal amidoboranes are plotted in Fig. 3. The dielectric loss factor,  $\epsilon''$ , is inversely proportional to the frequency, which indicates that dielectric response is dominated by ionic conductivity (the second term in the equation 4). In order to confirm this conclusion, a complex electric modulus formalism,  $M^*(\omega) = [\epsilon^*(\omega)]^{-1}$ , has been used since it allows for easier interpretation of the microscopic processes responsible for the measured a.c. response [79]. In the absence of dipolar relaxation, the introducing of a conductivity relaxation time  $\tau_\sigma = \epsilon_0 \epsilon_s \sigma_0$ , the electric modulus  $M^*(\omega)$  can be described as:

$$M^*(\omega) = [\epsilon^*(\omega)]^{-1} = M_s \frac{j\omega\tau_\sigma}{1 + j\omega\tau_\sigma} \quad (9)$$

Under these conditions  $M'$  exhibits a relaxation with a single relaxation time,  $\tau_\sigma$ , in the same way that dielectric loss  $\epsilon''$  for dielectric relaxation. Hence in this representation, the difference between localized (i.e. dielectric relaxation) and non-localized conduction (i.e. long range conductivity) processes within the bulk of the material can be easily detected. Indeed, this relaxation manifests as a peak in the plot of  $M'$  vs.  $\log(f)$  (Fig. 3 in [H10]). Importantly, the peak maximum shifts towards higher frequency as temperature is raised (ESI). In this way we have discovered high ionic conductivity of these materials ( $\sigma = 2.3 \text{ mS/cm}$ ,  $E_a = 158 \text{ kJ/mol}$  for NaAB) [H10].

Conductivity and dielectric properties undergo further significant change upon thermal decomposition of amidoboranes providing a deep insight into the chemical nature of this process. The studies clearly demonstrate that powder impedance spectroscopy is a powerful tool for monitoring processes related to hydrogen release from solid state hydrogen stores.

**Summary**

The most important scientific achievements related to habilitation can be summarized as follows:

- (i) Development of impedance model for catalytic reactions involving redox mediators together with potential dependence of all impedance parameters.
- (ii) Introduction of new impedance element for catalytic processes and demonstration of its potential applications in determination of all kinetic and thermodynamic parameters for the EC' type processes on the example of the Fenton reaction.
- (iii) Observation of and explanation of superior hydrogen absorption in the *alpha* phase of  $\alpha$ -Pd-P-H with respect to Pd-H system.
- (iv) Revealing of spontaneous segregation in nanostructured bimetallic Pd-Pt alloys and the elucidation of the exceptionally high hydrogen dissolution in the Pd-Pt nanoparticles.
- (v) Design and characterization of stable catalytic system, in which the kinetics of the hydrogen sorption phenomena in palladium was enhanced by nearly four orders of magnitude.
- (vi) Investigation of electric properties of hydrogen stores and mechanistic information acquired by using IS for thermal decomposition of ammonia borane and of its alkali metal derivatives (amidoboranes).



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## 5. Other scientific achievements

### a) Before Ph.D. title (total 5 items)

1. R. Jurczakowski, M. Orlik  
Experimental and Theoretical Studies of the Multistability in the Electroreduction of the Ni(II) Complexes at a Streaming Mercury Electrode.  
*Journal of Physical Chemistry B*, 2003, 107, 10148-10158  
**MNiSW: 35, IF2012: 3.696**
2. M. Orlik, R. Jurczakowski  
On the Stability of Nonequilibrium Steady-States for the Electrode Processes at a Streaming Mercury Electrode  
*Journal of Physical Chemistry B*, 2002, 106, 7527-7536  
**MNiSW: 35, IF2012: 3.607**
3. R. Jurczakowski, M. Orlik  
Bistable/Oscillatory System Based on the Electroreduction of Thiocyanate Complexes of Nickel(II) at a Streaming Mercury Electrode  
*Journal of Physical Chemistry B*, 2002, 106, 1058-1065  
**MNiSW: 35, IF2012: 3.607**
4. R. Jurczakowski and M. Orlik  
The oscillatory course of the electroreduction of thiocyanate complexes of nickel(II) at mercury electrodes - experiment and simulation  
*Journal of Electroanalytical Chemistry*, 2000, 486, 65-74  
**MNiSW: 30, IF2012: 2.672**
5. R. Jurczakowski and M. Orlik  
On the source of oscillatory instabilities in the electroreduction of thiocyanate complexes of nickel(II) at mercury electrodes  
*Journal of Electroanalytical Chemistry*, 1999, 478, 118-127  
**MNiSW: 30, IF2012: 2.672**

### b) After Ph.D. title (total 39 items including 9 in habilitation thesis, star indicates R. Jurczakowski as corresponding author)

1. P. Połczyński, R. Jurczakowski\*  
Extremely Fast Hydrogen Absorption/Desorption through Platinum Overlayers  
*Journal of Power Sources*, 2016  
**MNiSW: 45, IF<sub>2013</sub>: 6,217**

(my contribution of ca. 70 % - the idea, research supervision, plan and participation in data analysis, manuscript preparation)

2. P. Połczyński, R. Jurczakowski\*  
Impedance as a Tool for Rapid and Complete Characterization of Electrocatalytic Systems Involving Redox Mediators  
*Electrochimica Acta*, 2016, 188, 882-887  
**MNiSW: 40, IF<sub>2013</sub>: 4,504**

(my contribution of ca. 60 % - the idea, research supervision, participation in data analysis and in simplified model development, manuscript preparation)

3. A. Januszewska, G. Dercz, A. Lewera, R. Jurczakowski\*  
Spontaneous Chemical Ordering in Bimetallic Nanoparticles  
*Journal of Physical Chemistry C*, 2015, 119, 19817–19825  
**MNiSW: 35, IF<sub>2012</sub>: 4,814**

(my contribution of ca. 65 % - the idea, research supervision, plan and participation in data analysis, data interpretation, participation in manuscript preparation)

4. W. Adamczyk, P. Połczyński, P. Mika, T. Jaroń, Z. Mazej, K.J. Fijalkowski, R. Jurczakowski, W. Grochala,  
New Ag( $F_{1-x}Cl_x$ ) phases for energy storage applications  
*Journal of Fluorine Chemistry*, 2015, 174, 22–29  
**MNiSW: 25, IF2012: 1.952**

(my contribution of ca. 5 % - supervision and interpretation of dielectric measurements, participation in the manuscript preparation related to these results)

5. K. Klucińska, R. Jurczakowski, K. Maksymiuk, A. Michalska  
Ultrasensitive 4-methylumbelliferon fluorimetric determination of water contents in aprotic solvents  
*Talanta*, 2015, 132, 392–397  
**MNiSW: 40, IF2012: 3.511**

(my contribution of ca. 5 % - supervision and participation in Karl Fischer (KF) titration, participation in the manuscript preparation related to KF results)

6. A. Januszewska, R. Jurczakowski\*, Kulesza P.  
CO<sub>2</sub> electroreduction at bare and Cu-decorated Pd pseudomorphic layers: Catalyst tuning by controlled and indirect supporting onto Au(111)  
*Langmuir*, 2014, 30(47), 14314–14321  
**MNiSW: 35, IF2012: 4.384**

(my contribution of ca. 60 % - participation in the idea, research supervision, plan and participation in data analysis, manuscript preparation)

7. R. Jurczakowski\*, P. Połczyński  
Impedance of Mediated Electrochemical Processes. Novel Impedance Element for Unequal Diffusivities.  
*Journal of Physical Chemistry C*, 2014, 118, 7980–7988  
**MNiSW: 35, IF2012: 4.814**

(my contribution of ca. 80 % - the idea, research supervision, analytical model development, participation in the development of new element and manuscript preparation)

8. A. Januszewska, G. Dercz, J. Piwowar, R. Jurczakowski\*, A. Lewera  
Outstanding catalytic activity of ultra-pure platinum nanoparticles.  
*Chemistry – A European Journal*, 2013, 19, 17159–17164  
**MNiSW: 40, IF2012: 6.378**

(my contribution of ca. 30 % - research co-supervision and participation in data interpretation, participation in the manuscript preparation)

9. P. Połczyński, R. Jurczakowski\*, W. Grochala  
Strong and long-lived free-radical oxidizer based on silver(II). Mechanism of Ag(I) electrooxidation in concentrated H<sub>2</sub>SO<sub>4</sub>.  
*Journal of Physical Chemistry C*, 2013, 117, 20689–20696  
**MNiSW: 35, IF2012: 4.814**

(my contribution of ca. 30 % - research supervision and participation in data interpretation, manuscript preparation)

10. E. Van Wenum, R. Jurczakowski, G. Litwinienko  
Media effects on the mechanism of antioxidant action of silybin and 2,3-dehydrosilybin: Role of the enol group.  
*Journal of Organic Chemistry*, 2013, 78, 9102–9112  
**MNiSW: 35, IF2012: 4.564**



(my contribution of ca. 10 % - electrochemical measurements supervision, interpretation of the electrochemical results, participation in manuscript preparation related to these results)

11. Połczyński P., Jurczakowski R., Grochala W.  
Stabilization and strong oxidizing properties of Ag(II) in a fluorine-free solvent.  
*Chemical Communications*, 2013, 49, 7480-7482  
**MNiSW: 40, IF2012: 6.378**

(my contribution of ca. 30 % - electrochemical research supervision and participation in data interpretation, participation in the manuscript preparation)

12. B. Losiewicz, R. Jurczakowski, A. Lasia,  
Kinetics of Hydrogen Underpotential Deposition at Polycrystalline Platinum in Acidic Solutions  
*Electrochimica Acta*, 2012, 80, 292-301  
**MNiSW: 35, IF2012: 3.777**

(my contribution of ca. 25 % - participation in CV measurements, idea and carrying out the impedance measurements in hanging meniscus arrangement, participation in the interpretation of the frequency distribution)

13. M. Sosnowska, M. Goral-Kurbiel, M. Skunik-Nuckowska, R. Jurczakowski, P.J. Kulesza  
Hybrid materials utilizing polyelectrolyte-derivatized carbon nanotubes and vanadium-mixed addenda heteropolytungstate for efficient electrochemical charging and electrocatalysis  
*J. Solid State Electrochem.*, 2013, 17, 1631-1640  
**MNiSW: 25, IF2012: 2.010**

(my contribution of ca. 5 % - participation in data interpretation, participation in the manuscript preparation)

14. K.J. Fijalkowski, R. Jurczakowski, W. Koźmiński, W. Grochala,  
Insights from impedance spectroscopy into the mechanism of thermal decomposition of M(NH<sub>2</sub>BH<sub>3</sub>), M = H, Li, Na, Li<sub>0.5</sub>Na<sub>0.5</sub>, hydrogen stores  
*Physical Chemistry Chemical Physics*, 2012, 14, 5778-5784  
**MNiSW: 40, IF2012: 3.829**

(my contribution of ca. 30 % - electrochemical research supervision and participation in data interpretation, participation in the manuscript preparation)

15. R. Solarska, R. Jurczakowski, J. Augustynski  
A highly stable, efficient visible-light driven water photoelectrolysis system using a nanocrystalline WO<sub>3</sub> photoanode and a methane sulfonic acid electrolyte  
*Nanoscale*, 2012, 4, 1553-1556  
**MNiSW: 40, IF2012: 6.233**

(my contribution of ca. 20 % - impedance measurements and impedance data interpretation, participation in the manuscript preparation related to these results)

16. P. Kietlinska, M. Królikowski, A. Dobrzeniecka, R. Jurczakowski\*, K. Miecznikowski, I.A. Rutkowska, J. Stroka, P.J. Kulesza, Z. Galus  
The preparation and electrochemical properties of nanostructured nickel hexacyanoruthenate deposited on multiwalled carbon nanotubes  
*Electrochimica Acta*, 2011, 58, 474-480  
**MNiSW: 35, IF2012: 3.777**

(my contribution of ca. 5 % - SEM micrographs, preparation of the final version of the manuscript)

17. A. Królikowska, R. Jurczakowski\*, P. Barczuk, J. Augustynski



The core-shell nature of nanostructured WO<sub>3</sub> photoelectrodes demonstrated in Spectroelectrochemical studies  
*Journal of Electroanalytical Chemistry*, 2011, 662, 229-239  
**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 20 % - impedance measurements and impedance data interpretation, participation in the manuscript preparation related to these results)

18. M. Gorzkowski, A. Wesołowska, R. Jurczakowski, P. Ślepski, K. Darowicki, M. Orlik  
Electrochemical oscillations and bistability during anodic dissolution of vanadium electrode in acidic media-part I. Experiment  
*J. Solid State Electrochem.*, 2011, 15 (11-12), 2311-2320  
**MNiSW: 25, IF2012: 2.010**

(my contribution of ca. 5 % - the participation in the idea of the studies on vanadium anodic dissolution)

19. B. Łosiewicz, R. Jurczakowski, A. Lasia,  
Kinetics of hydrogen underpotential deposition at polycrystalline rhodium in acidic solutions  
*Electrochimica Acta*, 2011, 56, 5746-5753  
**MNiSW: 35, IF2012: 3.777**

(my contribution of ca. 10 % - participation in cyclic voltammetry experiments and data interpretation, participation in the manuscript preparation)

20. Kowalczyk, A. Nowicka, R. Jurczakowski, M. Fau, A. Krolikowska, Z. Stojek  
Construction of DNA biosensor at glassy carbon surface modified with 4-aminoethylbenzenediazonium salt  
*Biosensors and Bioelectronics*, 2011, 26, 2506-2512  
**MNiSW: 45, IF2012: 5.437**

(my contribution of ca. 5 % - impedance measurements and impedance data interpretation, participation in the manuscript preparation related to these results)

21. R. Solarska, B. D. Alexander, A. Braun, R. Jurczakowski, G. Fortunato, M. Stiefel, T. Graule and J. Augustynski  
Tailoring the morphology of WO<sub>3</sub> films with substitutional cation doping: Effect on the photoelectrochemical properties  
*Electrochimica Acta*, 2010, 55, 7780-7787  
**MNiSW: 35, IF2012: 3.777**

(my contribution of ca. 5 % - impedance measurements and impedance data interpretation, participation in the manuscript preparation related to these results)

22. K. Pekala, A. Wiśniewski, R. Jurczakowski, T. Wiśniewski, M. Wojdyga, M. Orlik  
Monitoring of Spatiotemporal Patterns in the Oscillatory Chemical Reactions with the Infrared Camera: Experiments and Model Interpretation  
*Journal of Physical Chemistry A*, 2010, 114, 7903–7911  
**MNiSW: 35, IF2012: 2.771**

(my contribution of ca. 5 % - participation in the experimental studies)

23. A. Kowalczyk, A.M. Nowicka, R. Jurczakowski, P. Niedzialekowski, T. Ossowski, Z. Stojek.  
New anthraquinone derivatives as electrochemical redox indicators for the visualization of the DNA hybridization process  
*Electroanalysis*, 2010, 22 49-59  
**MNiSW: 32, IF2012: 2.817**



(my contribution of ca. 5 % - impedance measurements and impedance data interpretation, participation in the manuscript preparation related to these results)

24. K. Pekala, R. Jurczakowski, M. Orlik

On the interpretation of the potentiometric response of the inert solid electrodes in the monitoring of the oscillatory processes involving hydrogen peroxide

*J. Solid State Electrochem.*, 2010, 14, 27-34

**MNiSW: 25, IF2012: 2.010**

(my contribution of ca. 10 % - the idea of the use of different electrode materials for potentiometric experiments in this system, participation in the experimental studies)

25. M. Słojewski, J. Kowalska, R. Jurczakowski\*

The influence of phosphorous content on hydrogen sorption in Pd–P thin layers obtained by electroless deposition

*Journal of Physical Chemistry C*, 2009, 113, 3707-3712

**MNiSW: 35, IF2012: 4.814**

(my contribution of ca. 75 % - the idea and research supervision, plan and participation in data analysis, data interpretation, manuscript preparation)

26. M. Gorzkowski, R. Jurczakowski, M. Orlik

Electrochemical oscillations and bistability in the redox processes of mercury ions, coupled with the self-induced convection of Hg surface

*Journal of Electroanalytical Chemistry*, 2008, 615, 135-144

**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 10 % - participation in the experimental studies)

27. M. Orlik, R. Jurczakowski

The kinetic parameters of the electrocatalytic reduction of the coordinated azide anions found from comparison of dc and ac measurements for the tristable Ni(II)-N3-system

*Journal of Electroanalytical Chemistry*, 2008, 614, 139-148

**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 30 % - chronoamperometric measurements, voltammetric studies, impedance measurements, participation in the data interpretation, participation in Figures preparation)

28. K. Pekala, R. Jurczakowski, A. Lewera, M. Orlik

Luminescent chemical waves in the Cu(II)-catalyzed oscillatory oxidation of SCN- ions with hydrogen peroxide

*Journal of Physical Chemistry A*, 2007, 111, 3439-3442

**MNiSW: 35, IF2012: 2.771**

(my contribution of ca. 25 % - participation in the idea of temperature measurements, participation in the experimental studies)

29. R. Jurczakowski, M. Orlik

Experimental and theoretical impedance studies of oscillations and bistability in the Ni(II)-SCN- electroreduction at the streaming mercury electrode

*Journal of Electroanalytical Chemistry*, 2007, 605, 41-52

**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 20 % - voltammetric experiments and impedance measurements)

30. R. Jurczakowski, B. Łosiewicz, A. Lasia,

Kinetic and Thermodynamic Parameters of Hydrogen Sorption in Pd, Pd-Pt and on Pt

*The Electrochemical Society Transactions*, 2007, 2 (17), 11-19

**MNiSW: n.a., IF2012: n.a.**



31. F. Vigier, R. Jurczakowski, A. Lasia  
Reply to the letter of P. Zoltowski "Remarks on "Determination of hydrogen absorption isotherm and diffusion coefficient in Pd81Pt19 alloy"  
*Journal of Electroanalytical Chemistry*, 2007, 602 (1) 145-148  
**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 40 % - determination of the hydrogen absorption isotherm, manuscript preparation)

32. R. Jurczakowski, G. Litwinienko, M. Orlik  
Studies on the formation and kinetics of self decomposition of Ni(IV)-N3- complexes in aqueous media  
*Zeitschrift für Physikalische Chemie*, 2006, 220, 1-17  
**MNiSW: 20, IF2012: 1.361**

(my contribution of ca. 30 % - the idea, voltammetric and UV-Vis experiments, data analysis, participation in data interpretation)

33. F. Vigier, R. Jurczakowski, A. Lasia  
Determination of hydrogen absorption isotherm and diffusion coefficient in Pd81Pt19 alloy.  
*Journal of Electroanalytical Chemistry*, 2006, 588, 32-43  
**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 10 % - determination of the hydrogen absorption isotherm)

34. M. Orlik, R. Jurczakowski  
On the role of specific characteristics of the streaming mercury electrode in the generation of the nonlinear electrochemical dynamical phenomena  
*Polish Journal of Chemistry*, 2004, 78, 1221-1234  
**MNiSW: 20, IF2012: 0.440**

(my contribution of ca. 10 % - experimental results)

35. R. Jurczakowski, C. Hitz, A. Lasia  
Impedance of porous gold electrodes in the presence of electroactive species  
*Journal of Electroanalytical Chemistry*, 2005, 582, 85-96  
**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 45 % - experimental results, plan and participation in data analysis, data interpretation, participation in the manuscript preparation)

36. R. Jurczakowski, M. Orlik  
The electroreduction of azides bound to nickel(II) ions in weak acidic media  
*Journal of Electroanalytical Chemistry*, 2005, 574, 311-320  
**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 30 % - chronoamperometric measurements and participation in data interpretation)

37. R. Jurczakowski, A. Lasia  
Limitations of the potential step technique to impedance measurements using discrete time Fourier transform  
*Analytical Chemistry*, 2004, 76, 5033-5038  
**MNiSW: 45, IF2012: 5.695**

(my contribution of ca. 70 % - all numerical modeling, participation in data interpretation, participation in manuscript preparation)

38. R. Jurczakowski, C. Hitz, A. Lasia  
Impedance of Porous Au based electrodes



Journal of Electroanalytical Chemistry, 2004, 572, 355-366  
**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 50 % - experimental results, plan and participation in data analysis, data interpretation, manuscript preparation)

39. R. Jurczakowski, M. Orlik  
The Apparent Faradaic Response in the Impedance Characteristics of the Capacitive Current at a Streaming Mercury Electrode  
Journal of Electroanalytical Chemistry, 2004, 562, 205-215  
**MNiSW: 30, IF2012: 2.672**

(my contribution of ca. 45 % - impedance measurements and impedance data interpretation)

c) **Bibliographic data**

Total Impact Factor 2013: **162.197**  
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Number of all citations: **380** (06.01.2016)

d) **Conferences and meetings**

*oral presentation given by the author of self-presentation*

1. P. Polczynski, R. Jurczakowski,  
**Impedance of Mediated Electrochemical Processes: Fenton Reaction**  
228 ECS Meeting, Phoenix, USA, 2015
2. P. Polczynski, R. Jurczakowski,  
**Exceptionally Fast Hydrogen Absorption and Desorption through Platinum Overlays**  
227 ECS Meeting, Chicago, USA, 2015
3. A. Januszewska, G. Dercz, A. Lewera  
**Spontaneous Phase Segregation in Bare Palladium-Platinum Nanoparticles Evidenced By Superior Hydrogen Absorption**  
225 ECS Meeting, Orlando, USA, 2014
4. Aneta Januszewska, Paweł Kulboka, Adam Lewera, Rafał Jurczakowski  
**Synthesis of ultrapure nanoparticles and its applications to electrocatalysis: Ethanol and Ethylene glycol oxidation on Pt**  
223rd ECS Meeting, Toronto, Kanada, 2013
5. R. Jurczakowski, A.Królikowska, P. Barczuk, J.Augustynski  
**The Influence of Solution Conductivity on the Impedance of mesoporous WO<sub>3</sub> layers**  
European Material Research Society Meeting, Francja, 2011
6. Aneta Januszewska, Adam Lewera, Rafał Jurczakowski,  
**Kinetics of Hydrogen Sorption in Pt-Pd Alloy Nanoparticles**  
218th ECS Meeting, USA, 2010
7. P. Polczynski, G. Dercz, E. Rowinski, M. Słojewski, R. Jurczakowski,  
**Kinetics and Thermodynamics of Hydrogen Sorption in PdP and PdN alloys**  
60th Annual Meeting of the International Society of Electrochemistry, Beijing, China, 2009
8. R. Jurczakowski, B. Łosiewicz, A. Lasia  
**Kinetics and Thermodynamics of Hydrogen UPD on Iridium in Perchloric and Sulfuric Acids**  
59th Annual Meeting of the International Society of Electrochemistry, Seville, Spain, 2008
9. R. Jurczakowski, A. Lasia,  
**Study of hydrogen electrosorption in Pd, Pt and Pd<sub>81</sub>Pt<sub>19</sub> in acidic media**,  
XLIX Meeting of Polish Chemical Society, Gdansk, Poland, 2006



10. R. Jurczakowski, C. Hitz, A. Lasia,  
**Impedance of porous Au based electrodes** Impedance Spectroscopy for Characterisation of Materials and Structures, Warsaw, Poland, 2003
11. A. Lasia, R. Jurczakowski,  
**Limitations of a New Potential Step Technique of Impedance Measurements Using Fourier Transform**  
203rd Meeting of the Electrochemical Society, Paris, France, 2003  
Student Travel Grant Award from Electrochemical Society

*Presentations given by other co-authors and posters*

12. P. Połczyński, R. Jurczakowski, W. Grochala;  
**Electrochemical Properties of Ag(II) in Concentrated Sulfuric Acid Solutions**  
223rd ECS Meeting, Kanada, 2013 (oral)
13. P. Połczyński, G. Dercz, R. Jurczakowski  
**Hydrogen electrosorption into palladium-nitrogen Alloys**  
223rd ECS Meeting, Kanada, 2013 (poster)
14. Aneta Januszewska, Rafał Jurczakowski, Paweł Kulesza  
**CO<sub>2</sub> Electroreduction at Pd/Au(111) and Pd/Pt(111) Multilayers**  
223rd ECS Meeting, Kanada, 2013 (oral)
15. A.Królikowska, P. Barczuk, R. Jurczakowski, J. Augustynski  
**Probing the interface structure of mesoporous WO<sub>3</sub> film photoanodes,**  
European Material Research Society Meeting, Francja, 2011 (oral)
16. R. Jurczakowski, B. Łosiewicz, A. Lasia  
**Kinetics and Thermodynamics of Hydrogen UPD on Rhodium in Perchloric and Sulfuric Acids** 59th Annual Meeting of the International Society of Electrochemistry, Seville, Spain, 2008, (oral)
17. A. Lasia and R. Jurczakowski  
**Kinetic and Thermodynamic Parameters of Hydrogen Sorption in Pd, Pd-Pt and on Pt,**  
209th ECS Meeting - Denver, Colorado, USA, 2006 (oral)
18. Rafał Jurczakowski, Marek Orlik,  
**Experimental and Theoretical Impedance Studies of Electrochemical Oscillations and ultistability in the Electrode Processes of Thiocyanate and Azide Complexes of Nickel(II)**  
Gordon Research Conference - Oscillations & Dynamic Instabilities in Chemical Systems, England, (poster)
19. A. Lasia, R. Jurczakowski, H. Duncan, B. Łosiewicz  
**Mechanism and the Kinetics of Hydrogen Adsorption and Absorption at Pt, Pd and PdPt Alloy** 5th International Conference on Electrocatalysis 5th ECS'06, Kotor, Montenegro, 2006 (oral)
20. A. Lasia, R. Jurczakowski, R., Duncan H.,  
**Mechanism and kinetics of hydrogen adsorption and absorption into metals**  
39<sup>th</sup> Heyrovsky Discussion and 7<sup>th</sup> International Symposium on Electrochemical Impedance Analysis, Castle Trest, Czech Republic, 2006 (oral)
21. M. Orlik, R. Jurczakowski  
**Oscillations and multistability in electrode reactions of pseudohalogenede complexes of Nickel(II) at mercury electrodes**  
XLIX Meeting of Polish Chemical Society, Gdansk, Poland, 2006 (lecture)
22. A. Tymosiak-Zielińska, R. Jurczakowski,  
**Catalytic activity of rough gold electrode in electrooxidation of methanol**  
XLIX Meeting of Polish Chemical Society, Gdansk, Poland, 2006 (poster)
23. M. T. Gorzkowski, R. Jurczakowski, M. Orlik  
**Study of convective instabilities at mercury surface as a example of dynamical selforganization** XLIX Meeting of Polish Chemical Society, Gdansk, Poland, 2006 (poster)
24. K. Pękala, R. Jurczakowski, M. Orlik  
**Luminescent chemical waves in the Cu(II)-catalyzed oscillatory oxidation of SCN<sup>-</sup> ions with hydrogen peroxide**  
XLIX Meeting of Polish Chemical Society, Gdansk, Poland, 2006 (poster)

25. R. Jurczakowski, A. Lasia,  
***Étude d'électrosorption d'hydrogène dans des films minces de l'alliage Pd<sub>0.81</sub>Pt<sub>0.19</sub> dans le milieu acide***  
Journées d'électrochimie, Saint Malo, Francja, 2005, (oral)
26. R. Jurczakowski, C. Hitz, A. Lasia,  
***L'impédance des électrodes poreuses à base de l'or***  
Journees d'électrochimie, Poitiers, Francja, 2003, (oral)
27. R. Jurczakowski, M.Orlik,  
***Oscillations in the Ni(II)-SCN<sup>-</sup> electroreduction – experiment and simulation***  
XLIII Meeting of Polish Chemical Society (PTCh), Lodz 2000, Poland (poster)
28. R. Jurczakowski, M. Orlik,  
***Electrochemical oscillations in the electroreduction of thiocyanate complexes of nickel(II) at mercury electrodes. Experiment and simulation*** 51- st Meeting of the International Electrochemical Society (ISE), Warszawa, Polska, 2000 (poster)
29. R. Jurczakowski, M. Orlik, ***On the source of oscillatory instabilities in the electroreduction of thiocyanate complexes of nickel(II) at mercury electrodes*** XLII Meeting of Polish Chemical Society (PTCh), Rzeszow, Poland, 1999 (poster)
30. R. Jurczakowski, M. Orlik, ***The Apparent Faradaic Response in the Impedance Characteristics of the Capacitive Current at a Streaming Mercury Electrode*** 3-rd Baltic Conference on Electrochemistry, Gdańsk, Polska, 2003, (poster)
31. R. Jurczakowski, M. Orlik, ***Application of the Streaming Mercury Electrode to the Studies of Oscillations and Multistability in Electrochemical Processes*** 3-rd Baltic Conference on Electrochemistry, Gdańsk, Polska, 2003, (oral)
32. Marek Orlik, R. Jurczakowski, ***Complex Dynamical Behaviors: Bistability and Tristability in the Electroreduction of the Azide Complexes of Ni(II) at a Streaming Mercury Electrode*** 53- rd Meeting of the International Electrochemical Society (ISE), Düsseldorf, Niemcy, 2002 (poster)
33. R. Jurczakowski, Marek Orlik,  
***Construction and application of the streaming mercury electrode to the study of oscillations and bistability in the electroreduction of Ni(II)-SCN<sup>-</sup> complexes. Experiment and simulations*** 53- rd Meeting of the International Electrochemical Society (ISE), Düsseldorf, Niemcy, 2002 (oral)

e) Patent pendings (domestic)

1. A. Lewera, R. Jurczakowski, J. Piwowar, B. Mielcarek, *A Method of Preparing Pure Precious Metal Nanoparticles With Large Fraction of (100) Facets, Nanoparticles Obtained by This Method and Their Use*, **P.407178** (2014)
2. K. Fiąkowski, R. Jurczakowski, *Cell for electrical measurements of highly reactive powder and liquid samples and method for electrical measurements of highly reactive powder and liquid samples*. **P.396725** (2011)
3. A. Januszewska, R. Jurczakowski, P. Kulesza, A. Lewera, *A Method of Preparing Pure Platinum Group Metal Nanoparticles and Microparticles*, **P.397444** (2011)
4. R. Jurczakowski, P. Kulesza, A. Lewera, P. Połczyński, *Stop pallad-azot, stabilny w warunkach normalnych, oraz jego zastosowanie*, **P.400169** (2012)
5. R. Jurczakowski, P. Kulboka, A. Lewera, *Sposób wytwarzania zasadniczo czystych nanoczastek w układzie przepływowym, nanocząstki otrzymane tym sposobem oraz ich zastosowanie*. **P.399505** (2012)
6. Piotr Połczyński, Aneta Januszewska, Krzysztof Strzałkowski, Rafał Jurczakowski *Materiał elektrodowy, sposób jego wytwarzania oraz zastosowanie, sposób prowadzenia fotoelektrosorpcji wodoru*, **P.405483** (2013)

f) Patent pendings (international)

1. A. Lewera, R. Jurczakowski, J. Piwowar, B. Mielcarek *A Method of Preparing Pure Precious Metal Nanoparticles With Large Fraction of (100) Facets, Nanoparticles Obtained by This Method and Their Use*, **PCT/IB2014/062831**
2. K. Fiąkowski, R. Jurczakowski, *Cell for electrical measurements of highly reactive powder and liquid samples and method for electrical measurements of highly reactive powder and liquid samples*, **PCT/IB2012/002111**.

3. Piotr Połczyński, Adam Lewera, Rafał Jurczakowski  
Selective catalyst especially for oxygen electroreduction, electrochemical system comprising thereof, palladium-inert gas alloy and its use and cell comprising the same, especially photo-fuel-cell, **PCT/IB2013/056260**
4. Paweł Kulboka, Adam Lewera, Rafał Jurczakowski  
A flow system method for preparing substantially pure nanoparticles, nanoparticles obtained by this method and use of thereof, **PCT/IB2013/054857**
5. Hubert Szabat, Aneta Januszewska, Piotr Połczyński, Adam Lewera, Rafał Jurczakowski  
Means for carrying out electroless metal deposition with atomic sub-monolayer precision, **PCT/IB2015/059969**
6. Piotr Połczyński, Adam Lewera, Rafał Jurczakowski  
Palladium-platinum system for use as hydrogen storage material and/or electrocatalyst, preferably in fuel-cells, **PCT/EP2015/079028**
7. A. Lewera, R. Jurczakowski, J. Piwowar, B. Mielcarek  
A Method of Preparing Pure Precious Metal Nanoparticles With Large Fraction of (100) Facets, Nanoparticles Obtained by This Method and Their Use,  
**PCT/IB2014/062831**
8. Maciej Słojewski, Agnieszka Tymosiak-Zielinska, Gregoire Auge, Gaetan Bigoin, Rafal Jurczakowski, Porous support covered by multilayers of different transition metals cyanometalates, preparation method and use of thereof, Francja, **No. 1561977**

**g) Granted patents**

1. Piotr Połczyński, Adam Lewera, Rafał Jurczakowski  
Selective catalyst especially for oxygen electroreduction, electrochemical system comprising thereof, palladium-inert gas alloy and its use and cell comprising the same, especially photo-fuel-cell, **PCT/IB2013/056260**
  - Polish patent **PAT.221643 (2016)**
  - European patent **EP 2 785 483 B1**
  - United States patent **US 9221044**
  - Japanese patent **JP 5807129**
2. Piotr Połczyński, Adam Lewera, Rafał Jurczakowski  
Selective catalyst especially for oxygen electroreduction, electrochemical system comprising thereof, palladium-inert gas alloy and its use and cell comprising the same, especially photo-fuel-cell, **PCT/IB2013/056260**
  - European patent **EP 2 785 483 B1**
  - United States patent **US 9221044**
  - Japanese patent **JP 5807129**
3. Paweł Kulboka, Adam Lewera, Rafał Jurczakowski  
A flow system method for preparing substantially pure nanoparticles, nanoparticles obtained by this method and use of thereof, **PCT/IB2013/054857**
  - European patent **EP 2803101**
  - Japanese patent **JP 5782579**



**6. Other information****a) Teaching:**

2005 - present	Course “ <b>General Chemistry</b> ” for 1 <sup>st</sup> year students, University of Warsaw
2006 - present	Supervising M.Sc. students during experimental work and preparation of thesis
2005 - present	Laboratory of <b>Inorganic Chemistry</b> for 3 <sup>rd</sup> year students, University of Warsaw
2005 - present	Laboratory of <b>Electrochemistry</b> for 4 <sup>th</sup> year students, University of Warsaw
2004	Occasional lectures in “ <b>Instrumental Analytical Chemistry</b> ” for undergraduate students, University of Sherbrooke (in French)
2000 - present	Calculus in “ <b>Analytical Chemistry</b> ” for 1 <sup>st</sup> year students, University of Warsaw
1999 - 2001	Laboratory in “ <b>Analytical Chemistry</b> ” for 2 <sup>nd</sup> year students, University of Warsaw

**b) Leadership in research projects:**

1. The influence of non-metallic element on the hydrogen electrosorption in Pd-P and Pd-N thin layers. Grant MNiSW, (finished in 2013), 293 000 PLN
2. ONET-Group, France, Provisions of services contract, 62 000 EUR, finished in 2015
3. A flow system method for preparing substantially pure nanoparticles.  
National Centre for Research and Development, (finished in 2015), 510 317,78 PLN
4. Method of preparation of selective catalyst for oxygen electroreduction  
National Centre for Research and Development, (finished in 2015), 536 681 PLN
5. Grants of University of Warsaw:  
501/68-BW-182101, (2008-2010 r.) /70 000 PLN/

**c) Scientific practices**

2015 (1 week)	Electrochemistry in anhydrous hydrofluoric acid. Jozef Stefan Institute in Ljubljana, Slovenia
2004-2007 (18 months)	Postdoctoral Fellow Laboratory of Advanced Electrochemistry, Prof. Andrzej Lasia Sherbrooke University, Quebec CANADA
2000/2001 (7-months)	Visiting Fellow Laboratory of Advanced Electrochemistry, Prof. Andrzej Lasia Sherbrooke University, Quebec CANADA

