SELF-PRESENTATION

1. Name and surname:

Michał Grdeń

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

1996 – M.Sc. in chemistry, Faculty of Chemistry, University of Warsaw 2001 – Ph.D. in chemistry, Faculty of Chemistry, University of Warsaw, title of the dissertation: "Hydrogen electrosorption in palladium-nickel and palladium-platinum alloys", supervisor Prof. Dr. hab. Andrzej Czerwiński

3. Previous employment in scientific institutions:

February 2002 – March 2013 adjunct in Physical Chemistry Division, 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:

A series of published papers devoted to formation of thin oxidised layers on surfaces of solid electrodes in aqueous electrolytes

b) Publications related to the achievement (habilitation thesis) included in Journal Citation Reports database (star indicates M. Grdeń as corresponding author):

1. M. Grdeń, K. Klimek, A. Czerwiński

"A quartz crystal microbalance study on a metallic nickel electrode"

Journal of Solid State Electrochemistry, **8** (2004) 390-397 (IF(2011) = 2.131)

(my contribution of ca. 55% - the idea, participation in planning and execution of CV, CA and EQCM experiments, participation in data analysis and calculations, preparation of the manuscript)

2. M. Grdeń*, K. Klimek

"EQCM studies on oxidation of metallic nickel electrode in basic solutions" *Journal of Electroanalytical Chemistry*, **581** (2005) 122-131 (IF(2011) = 2.905) (my contribution of ca. 50 % - the idea, plan and participation in execution of CV, EQCM and RDE experiments, participation in data analysis, preparation of the manuscript) 3. M. Alsabet, **M. Grdeń**, G. Jerkiewicz

"Comprehensive Study of the Growth of Ultra-Thin Oxide Layers on Pt Electrodes under Well-Defined Temperature, Potential, and Time Conditions"

Journal of Electroanalytical Chemistry, **589** (2006) 120-127 (IF(2011) = 2.905) With erratum:

G. Jerkiewicz, M. Alsabet, M. Grdeń, H. Varela, G. Tremiliosi-Filho

"Erratum to: "Comprehensive study of the growth of thin oxide layers on Pt electrodes under well-defined temperature, potential, and time conditions" [Journal of Electroanalytical Chemistry, 589 (2006) 120–127]"

Journal of Electroanalytical Chemistry, **625** (2009) 172-174 (IF(2011) = 2.905)

(my total contribution of ca. 10 - 15 % - participation in calculations and data analysis, discussion of the results, participation in preparation of the manuscript)

4. M. Grdeń, A. Czerwiński

"EQCM studies on Pd-Ni alloy oxidation in basic solution"

Journal of Solid State Electrochemistry, **12** (2008) 375-385 (IF(2011) = 2.131)

(my contribution of ca. 90% - the idea, plan and execution of CV, EDX and EQCM experiments, deposition of alloys, data analysis and interpretation, manuscript preparation)

5. M. Grdeń*

"Electrochemical Quartz Crystal Microbalance Studies of a Palladium Electrode Oxidation in a Basic Electrolyte Solution", *Electrochimica Acta*, **54** (2009) 909-920 (IF(2011) = 3.832) (my contribution - 100%)

6. M. Grdeń*, K. Klimek, Z. Rogulski

"A quartz crystal microbalance study on oxidation of a cobalt electrode in an alkaline solution", *Electrochemistry Communications*, **11** (2009) 499-503 (IF(2011) = 4.859) (my contribution of ca. 80% - the idea, Co electrodeposition, plan and execution of CV, EQCM and RRDE experiments, data analysis, manuscript preparation)

7. M. Alsabet, M. Grdeń, G. Jerkiewicz

"Electrochemical Growth of Surface Oxides on Nickel. Part 1: Formation of α -Ni(OH)₂ in Relation to the Polarization Potential, Polarization Time, and Temperature"

Electrocatalysis, **2** (2011) 317-330 (Impact Factor to be announced in 2013) (my contribution of ca. 50 % - participation in CV, CA and EIS measurements, calculations, data analysis, discussion of the results, participation in preparation of the manuscript) 8. **M. Grdeń**, M. Alsabet, G. Jerkiewicz

"Surface science and electrochemical analysis of nickel foams"

ACS Applied Materials and Interfaces, 4(2012) 3012-3021 (IF(2011) = 4.525)

(my contribution of ca. 70% - plan and execution of CV, SEM and XPS experiments, calculations and data analysis, preparation of the electrodes, participation in manuscript preparation)

9. M. Grdeń*, J. Jagiełło

"Oxidation of electrodeposited cobalt electrodes in an alkaline electrolyte" *Journal of Solid State Electrochemistry*, **17** (2013) 145-156 (IF(2011) = 2.131) (my contribution of ca. 50 % - the idea, plan of the experiments, participation in execution of CV, CA, EQCM and RRDE experiments, analysis of XPS data, participation in data analysis and interpretation, preparation of the manuscript)

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

1. Introduction

The main topic of the series of publications that provides a basis for habilitation is related to the electrochemical formation of thin oxidized layers (oxides/hydroxides) at the surface of metallic electrodes in aqueous electrolytes. Term "formation of thin layers" is related to the early stages of surface oxidation processes. These processes start when the electrode surface is metallic (reduced) and lead to formation of oxide/hydroxide layers with the thickness of up to few atomic/molecular layers.

Many electrochemical reactions with a practical importance take place at the surfaces of metallic electrodes covered with oxidized layers with various thicknesses. These reactions include important processes, such as oxygen evolution/reduction, oxidation/reduction of various organic compounds and reactions of carbon oxides. The oxidized layer formed at the electrode surface and present in potential region of above mentioned reactions may act as a barrier, due to e.g. its non-metallic properties [1-3], or may facilitate the process acting as a catalyst [4-7]. In aqueous electrolytes the surface of non-noble metals, with high negative values of standard potentials, is usually oxidized already at potentials close to the onset of hydrogen evolution. Therefore, the properties of the oxidised layer present at the electrode surface may determine electrochemical behavior of such metals in a potential window between onsets of hydrogen and oxygen evolution. From the practical point of view, formation of such oxidized layer could be both destructive for the metal (corrosion) as well as helpful in protection of the material (protecting passive layers) [8-10]. In many cases, such as reactions of carbon oxides or simple organic compounds proceeding at the surface of noble metal electrodes under ambient conditions, the thickness of the oxidized layer present at the surface is small, i.e. about one monolayer or less. Therefore, the knowledge learned from studies on growth of thick, multilayer oxides/hydroxides is not always sufficient for comprehensive description and explanation of the processes studied.

A literature review reveals limited knowledge about the early stages of processes of the surface oxidation taking place on partially oxidized electrodes, i.e. when the significant section of the electrode surface being in contact with the electrolyte is still metallic. A little is known about the mechanism and kinetics of the surface oxidation and physical and chemical properties of the oxidised layers formed during early stages of the surface oxidation. It is common that only a fraction of the surface of the metal is oxidized under ordinary laboratory conditions. Hence, the conclusions drawn from studies on formation of thick, multilayer oxides/hydroxides are not always applicable in analysis of electrochemical processes related to the ultrathin oxidized layers. Determination of processes related to the early stages of the surface oxidation and formation of ultrathin oxidized layers is valuable in terms of comprehensive analysis of above mentioned electrochemical processes taking place on surfaces of electrodes partially oxidized or covered with thin oxides/hydroxides. The series of publications in question focuses on four metals: two noble: platinum and palladium; and two non-noble cobalt and nickel. These metals differ in respect to chemical and electrochemical properties but every one of them plays an important role in industry and in laboratory applications.

Apparently, oxidation studies are less complicated when performed on noble metal electrodes. This is due to several factors:

- High chemical reversibility of processes of oxidation of noble metals. This allows easy reduction of the oxidized layer under conditions not very far from that applied for the oxidation (potential, time). The original, metallic surface can be easily restored while the charge of the oxide/hydroxide reduction can be used for in-situ thickness determination;

- Relatively low solubility of most of the noble metals in various aqueous electrolytes;

- Well defined and widely accepted methods of real surface area determination.

Therefore, noble metal electrodes constitute a good starting point in studies on the mechanism of surface oxidation.

2. Platinum electrode

Platinum was one of the first subjects of comprehensive studies on mechanism and kinetics of electrochemical surface oxidation [11-15]. Early oxidation models considered for Pt oxidation were based on an assumption that the surface oxidation begins with formation of stable surface species containing Pt with +1 oxidation state (Pt(I)), usually PtOH [11-13,16-24]. These species are further oxidized to Pt with +2 oxidation state (Pt(II), PtO in acidic electrolytes) in the next stages of the process. Formation of Pt(I) forms was often described as adsorption of oxygen containing species [14,21,25], most likely due to high reversibility of the early stages of the surface oxidation. Many recent works, utilizing e.g.

electrochemical quartz microbalance, EQCM, impedance spectroscopy and ex-situ Auger spectroscopy do not confirm formation of stable Pt(I) species and point out to formation of Pt(II) compounds only [26-28]. In acidic solutions oxidation of Pt surface follows a direct logarithmic law described by a linear relationship between the amount of the oxidized layer formed and the logarithm of the oxidation time [11,28-34]. Various interpretation of direct logarithmic law of Pt oxidation are proposed in the literature: a nucleation and growth model [31]; a reaction proceeding with bond breaking accompanied by migration of metal atom into the oxide with simultaneous transfer of two electrons [34]; and a mechanism called a "field assisted place exchange" [23,30]. For the oxide thicknesses above one monolayer, when the steady state conditions are established, further Pt oxidation in 0.5 M H₂SO₄ was described in [35] as following point defect model [36]. This model, however, is applicable for the thicknesses of at least several monolayers and cannot be applied for the early stages of the surface oxidation, when the first monolayer is formed (submonolayer coverage with the oxidized layer) and the system is not under steady state conditions.

Results of the studies on oxidation of Pt electrodes in 0.5 M H₂SO_{4aq} in potential range 900 – 1500 mV vs. RHE with formation of the oxidized layers with the thickness of up to ca. 2 monolayers are described in [H1]. Term monolayer is defined here as the thickness of the oxidized layer formed by oxidation of a full single layer of Pt atoms from the metallic surface. (The same definition is used also for other metals thorough the text.) It should be bearing in mind, however, that due to differences between molar densities of metal and oxide/hydroxide and due to differences between dimensions of metal and oxygen ions such a layer may be composed with two atomic rows of atoms/ions contributing to the oxide/hydroxide structure. Although apparently misleading, his term is useful and convenient when the amount of the oxidised layer is related to the fully reduced, metallic surface. Further on, very often there is no data concerning composition, structure and physical properties of very thin oxidised layers formed by oxidation of e.g. 1-2 atomic layers of the metal. This is caused by experimental difficulties in application of suitable experimental techniques allowing structure analysis of such thin layers. Thus, the term "monolayer" used in relation to the oxidized layer formed by oxidation of a single layer of atoms of the metal is most convenient in use and most unequivocal for interpretation. Such defined "monolayer" (or "monolayer equivalent") is a term frequently used in electrochemistry.

The results presented in [H1] show that the kinetics and mechanism of Pt oxidation in an acidic electrolyte depend on the oxidized layer thickness. For the thicknesses of up to ca. 1 monolayer a direct logarithmic law is applicable, with a linear relationship between the amount of the oxidized layer formed and the logarithm of the oxidation time. Formation of the layers with thicknesses greater than 1 monolayer can be described by an inverse logarithmic law, i.e. a linear relationship between the logarithm of the oxidation time and reciprocal of the amount of the oxidised layer.

A similar relation between the oxidation mechanism and the oxidized layer thickness was reported also for Pd, Au and Rh [37-39]. Most of the oxidation models discussed in the literature focus on thickening of already existing oxidised layers when the interface between the metal the electrolyte does not exists. These models are not applicable for the early stages of the surface oxidation, when formation of the first monolayer takes place. The model describing early stages of the surface oxidation with formation of the very first monolayer of the oxide/hydroxide was developed by Conway et al. [30]. According to this model, the very first step of the surface oxidation comprises formation of surface dipoles composed with metal and oxygen ions with the latter ion directed towards the electrolyte. This is energetically unfavorable arrangement due to electrostatic repulsion between ions having a charge of the same sign and located in the same layer. Therefore, the next step is rotation of some of the dipoles that leads to formation of a chessboard-like arrangement with alternatively arranged

metal and oxygen ions. The driving force of the dipoles rotation is the electric field established at electrode|electrolyte interface, across the layer of the dipoles. The electric field of the dipoles subjected to the rotation is directed oppositely to the original field at the surface. As a result, the total electric field at the interface, i.e. the driving force o the process, decreases with the progress in the surface oxidation. Assuming that rotation of the surface dipoles is the rate determining step, a linear relation is obtained between the logarithm of the oxidation time and the amount of the oxidized layer formed, the latter expressed as e.g charge of the oxide formation or reduction. The following equation was derived in [30] for a single electron transfer process:

$$q_{ox} = \frac{d_{\rm d} RTF\varepsilon_0}{2N_{\rm A}\beta\mu^2} \ln(t+c_{\rm c}) + C'$$
(1)

where d_d is the size of the rotating dipole, μ is the dipole moment, c_c is the integration constant and β stands for the symmetry factor (typically equal to 0.5). *C*' is a constant and depends on the rate constant, surface potential, potential drop across the dipole layer and on the other parameters from eq. 1. The value of the dipole moment obtained on the basis of eq. 1 for Pt electrodes equals 1.1 ± 0.1 D and is potential independent [H1]. This value is comparable with the values obtained for a PtO molecule in gaseous state with the range from 2.77 to 1.15 D, depending on the energetic state of the molecule [40].

A reversed logarithmic law observed for Pt oxidation above 1 monolayer presented in [H1] can be described by the model proposed by Mott and Cabrera [41-43]. This model assumes formation of a homogenous electric field across the oxidized layer. The existence of additional potential drops at metal/oxidized layer and oxidized layer/electrolyte interfaces is disregarded, the reactions are irreversible and the model describes the system that is not under equilibrium and not under steady state conditions. The rate of the overall process is determined by a transfer of a metal cation at the oxide|metal interface from the metal to the interstitial site in the oxidized layer. The energy barrier to be crossed in this step is higher than for transport of ions through the oxidized layer that is facilitated by the electric field. The electric field drives the metal cation towards electrole|electrolyte interface where the cation combines with oxygen to form next oxide/hydroxide molecule/layer. Thus, the direction of the oxidized layer growth is towards the electrolyte. The following equation describes the relation time, t, [41]:

$$d^{-1} = \frac{k_{\rm B}T}{q_{\rm k}b_{\rm l}V_{\rm ox}}\ln(t) + \frac{k_{\rm B}T}{q_{\rm k}b_{\rm l}V_{\rm ox}}\ln\left(\frac{q_{\rm k}b_{\rm l}V_{\rm ox}u}{k_{\rm B}Td_{\rm L}^2}\right)$$
(2)

where q_k is the charge of the kation, V_{ox} is the potential drop across the oxidised layer, b_1 is the distance between cation site inside the metal and the top of the energy barrier to be crossed and d_L stands for the maximum thickness of the oxidized layer. Parameter u is defined as [41]:

$$u = N_{\rm s} V_{\rm o} v_{\rm m} \exp\left(\frac{-(H_{\rm i} + U)}{k_{\rm B}T}\right)$$
(3)

where N_s is the surface density of atoms in the metal, V_o is the volume of the oxidised layer per one metal cation, v_m is the frequency of vibration of atoms in the metal lattice, H_i is the enthalpy of introduction of the cation into the oxidized layer lattice and U stands for the activation energy for cation transport inside the oxidized layer [41]. The electric field that is established inside the oxidized layer formed on Pt electrodes in the acidic solution is in the range of $(0.15-1.23)\cdot 10^9$ V m⁻¹, and depends on the applied potential and temperature [H1].

3. Palladium electrode

Detailed studies on the kinetics and the mechanism of oxidation of Pd electrodes were performed only in acidic electrolytes [37,44-46], there is a lack of similar comprehensive studies carried out in alkaline electrolytes. In paper [H2] an electrochemical quartz crystal microbalance, EQCM, was employed to study Pd oxidation in 0.1 M KOH and NaOH electrolytes. It has been shown in [H2] that the frequency response of EQCM recorded for Pd electrodes oxidized in an alkaline electrolyte up to the thickness of ca. 2 monolayers is determined by pure mass changes and is not influenced by other factors, such as viscoelastic effects, surface roughness or stress generation due to a mismatch between crystallographic structures of the oxide and the metal. Therefore, for the system Pd/0.1M KOH/NaOH the EQCM can be considered as a pure mass sensor. It was also found [H2] that alkali cations do not participate in Pd oxidation and the effects, such as cations adsorption or incorporation into the oxide/hydroxide lattice, can be neglected.

A relation between potential or thickness dependent composition of the oxidized layer formed on a Pd electrode in an alkaline electrolyte and reversibility of the oxidation process has been found [H2]. The reversibility is defined here as a distance between oxidation potential and potential of the voltammetric peak due to reduction of the oxidised layer. The oxidized layer containing mainly Pd(OH)₂ or PdO·H₂O is easier to reduce as compared to PdO. This observation suggests existence of a relation between oxidation mechanism and composition of the oxidized layer formed [H2]. A similar effect was reported for Co electrodes in [H3]. Thus, formation of PdO-type species could be related to formation of a new phase, e.g. due to rotation of surface dipoles, with a structure that is significantly different from the crystallographic structure of the host metal. This difference between structures of the oxidized layer and the metal can be more pronounced for PdO than for Pd(OH)₂/PdO·H₂O with the surface coverage below unity. As a result, reduction of PdO accompanied by a pronounced phase transition can be strongly impeded as compared to Pd(OH)₂/PdO·H₂O. A similar relation between composition of palladium containing oxidized layer and reversibility of Pd oxidation has been found also for electrodeposited Pd-Ni alloys [H4]. Formation of PdO-type species is accompanied by an increase in irreversibility of the oxidation process also for PdNi alloys. Therefore, the electrochemical behaviour of Pd atoms constituting the surface of a PdNi alloy is the same as in case of pure palladium.

A comparison of the processes participating in surface oxidation of Pd and subsequent reduction of the oxidized layer in an alkaline electrolyte is presented in [H2]. It was found that reduction of the oxidized layer with the thickness of up to ca. 2 monolayers contains the same elementary steps as the oxidation process but with reversed sequence. Therefore, the same processes and the same compounds take part both in formation of the oxidized layer and its subsequent reduction.

4. Nickel electrode

Nickel is a good example of a non-noble electrode with relatively well developed potential regions [47,48] that could be linked to the regions defined in corrosion science as active, passive and transpassive ones. It is expected that composition of the oxidized layer and the mechanism and kinetics of the oxidation process are different for each of these potential regions.

Papers [H5,H6] describe measurements carried out mainly in an aqueous 0.1 M KOH electrolyte by means of EQCM for electrodeposited Ni electrodes. Three distinctive, well separated regions of current changes on cyclic voltammetry curve, and mass changes on EQCM curves, can be connected with the following potential ranges:

- reversible oxidation region where formation of ca. 1 monolayer of the oxidized layer containing Ni with +2 oxidation state takes place (room temperature, ca. -950 - -500 mV vs.

SCE for electrodeposited Ni, ca. -50 - 500 mV vs. RHE for bulk, massive Ni [H7,H8]. For the latter electrode up to ca. 2 monolayers are formed at elevated temperatures);

- irreversible oxidation region that can be linked to the passive region in corrosion science terminology (ca. -500 - 400 mV vs. SCE, nickel oxidation to +2 oxidation state takes place in this potential region);

- potential range of nickel oxidation to oxidation state higher than +2 (potentials more positive than ca. 400 mV vs. SCE, an equivalent to transpassive region in corrosion science).

Ni(III) (oxidation state of +3) is the most often reported product of Ni(II) oxidation in the latter potential region [48]. EQCM measurements with electrodeposited Ni electrodes performed in KOH and NaOH solutions [H5] show that oxidation of Ni(II) is accompanied by incorporation/removal of alkali metal cations into/from the oxidized layer, the effect similar to that reported for nickel hydroxide electrodes [49]. Direction of mass changes accompanying Ni(II) oxidation is different for electrodeposited Ni [H5] and for Pd-Ni alloys [H4]. Despite differences in mass transport accompanying the process, thermodynamics of Ni(II) oxidation is generally the same for metallic Ni and for PdNi alloys as follows from the alloy composition independent potentials of voltammetric peaks related to Ni(II)/Ni(III) redox couple [H4].

It has been discussed in [H4] that stabilized gravimetric profiles recorded for Ni(II) oxidation on electrodeposited Ni electrodes point to direction of mass changes opposite to what is observed for Ni(OH)₂ electrodes after stabilization of EQCM profiles [49]. Therefore, application of the scheme of reactions of Ni(II)-Ni(III) red-ox couple that was developed for Ni(OH)₂ electrodes [49] is not always justified for metallic, electrodeposited nickel. EQCM profiles presented in [H4] suggest that crystallographic form of the hydroxide unstable for Ni(OH)₂ electrodes, called α Ni(OH)₂, could be significantly more stable when formed on the surface of metallic, electrodeposited nickel electrodes.

Paper [H6] presents results of EQCM studies on Ni oxidation in 0.1 M KOH at potentials up to -500 mV vs. SCE. Possible contribution from stresses generated during surface oxidation to the measured frequency response of the microbalance is discussed in [H6]. Soluble Ni species and alkali metal cations do not participate in the surface oxidation process. Adsorbed H_2O/OH^2 play a role in the oxidation process, their contribution depends on applied potential and/or thickness of the oxidized layer. The oxidized layer formed at potentials of up to -500 mV vs. SCE contains significant amounts of NiO whose contribution depends on the electrode potential.

Hydrogen absorption in nickel electrodes was studied in [H6] by means of EQCM. Bulk hydrogen absorption during polarization at potentials in hydrogen evolution region was detected for both metallic Ni, with fully reduced surface, and for Ni electrodes subjected to the irreversible oxidation. Paper [H6] shows that the process of bulk hydrogen sorption in nickel electrodes in alkaline electrolytes takes place in the absence of promoters, such as thiourea or arsenic or selenium oxides [50-52] and is facilitated by presence of the oxidized layer at the electrode surface. The oxidized layer may increase the amount of absorbed hydrogen or may speed up the rate of the absorption process. EQCM results [H6] also show that hydrogen absorption in nickel is accompanied by formation of stresses inside the absorbing material, similarly like for other hydrogen absorbing metals and alloys [53-56].

The methods of real surface area determination of Ni based electrodes: foams with open pores, and massive, bulk Ni rods, are discussed in paper [H7]. This topic is related to practical utilization of measurements of currents due to surface oxidation/reduction. The discussed methods of the surface area determination were applied in studies of Ni oxidation in 0.5 M KOH [H8]. Selection of a proper method of the real surface determination requires knowledge about the chemical state of the electrode surface. In the case of a non noble metal, such as Ni, presence of a native oxide that can be easily formed when the metal is brought

into contact with moisture [57,58], is a very important factor that may affect electrochemical properties of the electrode. Therefore, the chemical properties of Ni surface are expected to depend strongly on the surface preparation and treatment.

The methods of real surface area determination which are based on measurements of currents due to Faradaic reactions, such as surface oxidation/reduction, deliver information about only a fraction of the surface available for the reaction (electrochemically active area). Existence of a non-reducible oxidised layer on a fraction of the surface may lead to their inactivity towards the specified faradaic processes. On the other hand, the double layer capacitance measurements give information about the whole surface being in contact with the electrolyte but this method requires the knowledge about surface distribution of the specific interfacial capacitance, i.e. the capacitance expressed per unit area. This is related to the fact that Ni compounds, such as NiO and Ni(OH)₂, possess non-metallic, semiconducting or dielectric properties [59-61]. Under such conditions, the overall interfacial capacitance contains contributions from the double layer on the electrolyte side and from the capacitance of the oxidized layer (solid state capacitance). It is clear that application of a specified method of the surface area determination of Ni electrodes requires suitable pretreatment of the electrode surface and evaluation of the degree of the surface oxidation [H7]. It should be noted that the surface pretreatment may affect the electrochemical performance of Ni electrodes in respect to various electrochemical processes as well (e.g. [62]). Apart from bulk, massive Ni electrodes, treated here as a reference material, the experiments described in [H7] were carried out also for a Ni foam, Incofoam [63,64], with open pores with diameter of ca. 500 µm (prevailing ones) and ca. 50 µm. There is a growing interest in application of such kind of Ni-based materials in various fields of electrochemistry, including electrocatalysis, supercapacitors, batteries and cells [65-70].

Two methods of pretreatment of the surface of Ni foam were applied: chemical etching and chemical reduction with gaseous H₂ [H7]. Apart from the cyclic voltammetry also surface science techniques, namely x-ray photoelectron spectroscopy, XPS, and scanning electron microscopy, SEM, were used in [H7] to characterize the surface of the foam. The real surface area was determined according to two methods: the method proposed in [71] and based on the surface oxidation/reduction charge measurements (correlation factor of 514 μ C cm⁻²), and on the basis of the double layer capacitance measurements [72,73] at potentials in hydrogen evolution region (-200 - -250 mV), where no surface oxidation takes place (correlation factor of 20 μ F cm⁻² [62,74-76]). The impedance spectra were fitted into the equivalent circuit containing an ohmic resistance, a constant phase element, CPE, depicting the capacitance of the double layer, and a charge transfer resistance connected in parallel with CPE. It was not possible to determine faradaic pseudocapacitance related to adsorbed hydrogen, most likely due to establishment of potential independent surface coverage with hydrogen [62,77]. The CPE was recalculated into the capacitance units according to the method described in [78] with assumed surface distribution of time constants.

Both methods of the real surface area determination require the degree of surface oxidation to be determined. This was accomplished on the basis of Ni2p3/2XPS spectra deconvoluted into Ni(0) and Ni(II) signals. An analysis of SEM images shows that surface roughening and dissolution during chemical etching is a local process. XPS spectra reveal higher degree of surface reduction for the chemically etched foam as deduced from a comparison of intensities of Ni(0) and N(II) 2p3/2 signals [H7]. XPS is not surface-only analysis and the depth of the analysis may reach few atomic layers, depending on the material [79]. However, the surface pretreatment induced changes in XPS spectra correlate well with the evolution of the shape of cyclic voltammetry curves revealing higher oxidation/reduction currents for chemically etched foam [H7]. Thus, chemical etching of the foam leads to an increase in the area of reduced (metallic) section of the surface being in contact with the

electrolyte. Therefore, it is likely that the surface of the foam contains fully reduced, metallic sections and sections oxidised, covered with the native oxides with the thickness that could be greater than one monolayer. The latter areas could be related to e.g. specific crystallographic orientation or local surface roughness, more susceptible to deep oxidation than the other sections of the surface. This idea is partially supported by conclusions drawn in [H8] where different rates of the surface oxidation of various sections of polycrystalline Ni surface were proposed. SEM images of Ni foam presented in [H7] shows that the increase in the surface roughness due to chemical etching is local and, most likely, is connected to the sections of the surface with specific structure. This idea is in line with the conclusions presented above.

Obtained in [H7] average real surface area was 241 ± 80 cm² g⁻¹ and 160 ± 23 cm² g⁻¹ reduced chemically etched and with respectively. for the foam H₂, The gaseous H₂ constitutes less aggressive environment as compared to the strongly acidic etching bath used in [H7] even at elevated temperature and under elevated pressure. Therefore, one can assume that the surface roughness of H₂ reduced foam is very close to that of untreated material and the value of ca. 160 cm² g⁻¹ can be adopted for calculation of the real surface area of Incofoam Ni foam with pore diameter of ca. 500 µm.

Detailed studies on the mechanism and kinetics of Ni oxidation were carried out in 0.5 M KOH_{aq} for a smooth, bulk Ni electrode in potential range 300 - 500 mV vs. RHE. The results are presented in paper [H8]. Earlier literature devoted to the studies of Ni oxidation in this potential range is often unambiguous in respect to the mechanism of the process. Dissolution of Ni single crystals was detected with STM in alkaline electrolytes [80,81]. It is not clear, however, if the dissolution is a step in the overall process of formation of the oxidized layer or a side reaction only. Ni oxidation according to nucleation and growth scheme was proposed in [80-86], in [87] a logarithmic grow of the oxidized layer was reported.

Oxidation of Ni electrode below 500 mV vs. RHE is a reversible process as follows from a comparison of the values of the interfacial capacitance measured before the oxidation and after reduction of the oxidized layer [H8]. The reversibility is also confirmed by an analysis of EQCM results obtained for electrodeposited Ni electrodes [H6]. An analysis of peak current-potential scan rate plots and chronoamperometric curves reveals that oxidation of Ni surface below 500 mV vs. RHE cannot be treated as a simple surface adsorption process without formation of a new phase, and shows that the nucleation and grow model with significant participation of soluble Ni species is not applicable as well.

The thickness of the oxidized layer formed on a Ni electrode was determined on the basis of the surface reduction charges recorded in voltammetric experiments. It has been found that the thickness increases linearly with the logarithm of the oxidation time up to ca. 1 – 1.9 monolayers, depending on applied potential and temperature [H8]. Both the thickness and the slope of the thickness vs. logarithm of time plots increase with increase in potential and temperature [H8].

The mechanism of Ni oxidation at 300 - 500 mV vs. RHE was identified in [H8] as "field assisted place exchange", the same as used in [H1] for description of Pt oxidation. The overall process becomes more complicated than formation of PtO discussed in [H1] when Ni(OH)₂ is considered as the oxidation product. This is related to two factors: identity of the species rotating in the electric field, and small probability of simultaneous transfer of more than one electron [88]. Therefore, the following steps of formation of Ni(OH)₂ can be proposed: 1) fast transfer of the first electron to form surface dipoles of Ni(OH); 2) rate determining step of rotation of Ni(OH) dipoles in the electric field; 3) fast transfer of the second electron and formation of Ni(OH)₂ type structures. The dipole moment of rotating Ni(OH) species calculated from eq. 1 is in the range of 1.17 – 1.56 D [H8] and is comparable

with the values obtained by means of the same method for noble metal oxides Pt (1.1D) [H1] and Au (1.5D) [37].

Potential influence on the slopes of the thickness vs. logarithm of time plots is reported and discussed in [H8]. It has been suggested [H8] that this effect can be attributed to potential dependent properties of the oxidized layer, e.g. chemical composition. This idea is partially supported by the results presented in [89] and showing a relation between crystallographic orientation of the surface and the shape of the surface oxidation/reduction currents for Ni single crystals in an alkaline electrolyte.

5. Cobalt electrode

Despite numerous practical applications of cobalt electrodes only few papers have been devoted to the studies on the early stages of Co oxidation in alkaline electrolytes at potentials negative of ca. -700 mV vs. Hg|HgO. Co oxidation according to a nucleation and growth mechanism was proposed for this potential range in 1 M NaOH in [90] while in [91] a solid state pathway of the oxidation was assumed for 0.1 and 1 M NaOH with a marginal role of cobalt dissolution as a side reaction. Cobalt oxidation without participation of soluble Co species was also confirmed in [92].

Papers [H3,H9] describe results of studies on oxidation of electrodeposited cobalt in aqueous solutions of 0.1 M and 1 M KOH. The demand for application of other than current/charge measurements techniques, such as EQCM and rotating ring disk electrode, RRDE, leads to use of electrodeposited Co electrodes as a subject of the studies instead of massive, bulk materials. The matrix for the electrodeposition was a gold wire (CV experiments), gold foil (XPS), EQCM quartz crystals and the disk of RRDE. Strong irreversibility of the oxidation processes and lack of widely accepted methods of the real surface area determination make the studies on cobalt oxidation much more complicated than in the case of nickel. A method similar to the one used for Ni electrodes [71] and utilizing charges of the surface oxidation-reduction was proposed for the real surface area determination of Co electrodes in alkaline environment [93]. Application of such procedure is, however, strongly complicated by strong irreversibility of Co surface oxidation, observed already for the early stages of the surface oxidation. This irreversibility manifests itself as a decrease of surface oxidation currents with potential cycling progress in cyclic voltammetry experiments [H9,93,94]. Cobalt dissolution, detected in alkaline electrolytes by means of e.g. RRDE [91,92], was suggested to be responsible for this irreversibility [93-95]. However, the EQCM measurements presented in [H3,H9] revealed that the irreversible changes of voltammetric curves due to Co surface oxidation in 0.1 M and 1 M KOH at potentials range in question are attributed to formation of a hardly reducible oxidised layer at the electrode surface. The oxidation of a bare metallic surface is accompanied by a mass increase which is not equalized by mass decrease recorded during subsequent reduction [H9]. A long term polarisation at -1000 - -1100 mV vs. Ag|AgCl leads to reduction of only a small fraction of such formed oxidized layer. Co dissolution was confirmed for 1 M and 3 M KOH by means of RRDE experiments in [H3,H9] but was not detected in 0.1 M KOH at potentials negative of ca. -880 mV regardless the fact that similar irreversibility of the surface oxidation is observed for all these concentrations [H3,H9]. This observation confirms the conclusion that the dissolution cannot be the main reason of irreversibility of Co oxidation in alkaline electrolytes.

The electrochemical impedance spectroscopy was employed in real surface area measurements [H3]. The measurements were carried out at potentials of hydrogen evolution region where the surface oxidation processes do not take place. Recorded spectra were fitted to a simple equivalent circuit containing constant phase element, CPE, representing the

double layer capacitance, and connected in parallel with a resistor related to the charge transfer resistance [H3]. The same type of the equivalent circuit was used also for Ni electrodes in [H7]. It was not possible to determine the faradic pseudocapacitance related to adsorbed hydrogen. Thus, the surface coverage with adsorbed hydrogen is constant in potential region of interest or is negligibly small. The CPE was recalculated into the capacitance units according to [78], the correlation factor of 20 μ F cm⁻² was assumed [96] for the real surface area determination.

The results presented in [H3] for a Co electrode oxidized in 0.1 M KOH reveal a relation between composition of the oxidized layer and the mechanism of its formation. EQCM and XPS experiments indicate that the prevailing components of the oxidized layer formed in potential range -1000 - -880 mV are CoO or Co(OH)₂, depending on the potential value [H3]. The oxidized layer composition can be connected with reversibility and kinetics of the oxidation process. It was found [H3] that under the same conditions (reduction potential and time) the degree of CoO layer reduction is significantly smaller as compared to Co(OH)₂. This observation points out to a strong irreversibility of processes of formation of CoO structures. It should be noted that EQCM experiment did not reveal any effects which could be attributed to hydrogen absorption in the bulk of a Co electrode during polarization in hydrogen evolution region [H9]. Further on, no influence of identity of alkali cation present in the solution was observed on discussed Co oxidation process [H3].

Three different techniques, including two independent: cyclic voltammetry (CV, reduction charge measurements), chronoamperometry (CA, oxidation charge/current measurements) and EQCM (mass changes) [H3] reveal that below -880 mV vs. Hg|HgO the oxidation of Co electrodes in 0.1 M KOH can be described by a direct logarithmic law, in the same way as in the case of Pt in 0.5 M H₂SO₄ [H1] and Ni in 0.5 M KOH [H8]. At potentials negative of -980 mV the slope of the thickness vs. logarithm of time plots is potential independent for all three techniques. At potentials more positive than ca.-980 mV the slope increases with the potential increase and the extent of the increase may reach even 300 - 900 % per 100 mV [H3]. This is a much stronger increase as compared to Ni oxidation in 0.5 M KOH [H8].

Potential independent slope of thickness vs. logarithm of time plots is predicted by the "field assisted place exchange model", derived by Conway et al. [30]. The same model describes also oxidation of Pt in $0.5 \text{ M H}_2\text{SO}_4$ [H1] and Ni in 0.1 M KOH [H8]. Therefore, the general scheme of Co(OH)₂ formation process can be considered the same as in the case of formation of Ni(OH)₂ on a Ni electrode [H8]. Thus, the first step is a fast transfer of the first electron that leads to formation of surface dipoles of Co(OH). Next, in the rate determining step, the surface dipoles rotate in the electric field established at the interface. Finally, a fast transfer of the second electron occurs, which is expected to be not impeded by relatively thin layer of the surface dipoles. The latter step leads to formation of Co(OH)₂ structures at the electrode surface [H3].

A strong potential influence on the slope of the thickness vs. logarithm of time plots observed for Co electrodes at potentials positive to -980 mV cannot be explained in the framework of the "field assisted place exchange" theory. The model based on the "nucleation and grow" mechanism [31] that predicts a linear relationship between the oxide thickness and logarithm of the oxidation time also cannot be applied to Co oxidation above -980 mV. This model predicts a linear increase in the slope with increasing potential, the effect roughly observed for CV data in [H3] while for CA and EQCM a strong non-linearity is observed.

It has been suggested in [H3] that the strong potential influence on kinetics of formation of CoO-type structures can be related to the reaction of decomposition of H₂O/OH⁻ into O²⁻. O²⁻ is the oxygen form that exists in the oxide and reaction of its formation should be included in the overall scheme of CoO generation. In contrast, the process of Co(OH)₂

formation does not require H_2O/OH^- decomposition into O^{2-} . The charged species participate in the process of H_2O/OH^- decomposition into O^{2-} and the rate of this reaction may depend on the potential despite the fact that there is no charge transfer accompanying the reaction. Therefore, the rate of processes taking part in O^{2-} formation, such as e.g. surface adsorption of charged species, can be potential influenced. Typical analysis of the surface oxidation reactions at potentials of thermodynamic stability of water usually does not include the separate step of H_2O/OH^- decomposition into O^{2-} . Hence, it has been suggested in [H3] that the contribution from the process of O^{2-} formation should be evaluated in comprehensive analysis of kinetics of formation of surface oxides.

Summary

The most important achievements related to the papers that provide a basis for the habilitation can be summarized as follows:

evaluation of the mechanism of early stages of the surface oxidation, leading to formation of the first monolayer of the oxide/hydroxide/ for Co (0.1 M KOH); Ni (0.5 M KOH) and Pt (0.5 M (H₂SO₄) electrodes, for the latter two metals a wide range of temperatures was investigated;
identification of the processes responsible for irreversible changes of the oxidation currents observed for Co electrodes in alkaline electrolytes during early stages of the surface oxidation; critical discussion of the method of the real surface area determination of Co electrodes based on measurements of the oxidation charges;

- identification of a relation between composition of the oxidised layer formed in alkaline electrolytes on Pd, Co and PdNi alloy electrodes and some aspects of the surface oxidation process, such as oxidation kinetics and reversibility;

- evaluation of possibility of bulk hydrogen absorption in electrodeposited Ni and Co electrodes from an alkaline electrolyte in the absence of species acting as absorption promoters;

- an analysis of the method of real surface area determination of Ni foam electrodes with open-pore structure; evaluation of influence of the surface pretreatment methods on the surface properties;

- a comparison of processes taking place during formation of up to ca. 2 monolayers of the oxidised layer on Pd and PdNi alloy electrodes in an alkaline electrolyte and during subsequent reduction of the oxides/hydroxides; evaluation of the sequence of reactions participating in the surface oxidation and reduction of Pd;

- evaluation of mass changes accompanying oxidation of Ni(II) compounds on electrodeposited metallic Ni electrodes, evaluation of alkali cations role in the process.

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H4. M. Grdeń, A. Czerwiński, J. Solid State Electrochem., 12 (2008) 375

H5. M. Grdeń, K. Klimek, A. Czerwiński, J. Solid State Electrochem., 8 (2004) 390

H6. M. Grdeń, K. Klimek, J. Electroanal. Chem., 581 (2005) 122

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5. Other scientific achievements

a) Full list of publications in peer reviewed journals from Journal Citation Reports list, including publications from habilitation thesis

Before Ph.D. title (total 9 items)

1. A. Czerwiński, M. Dmochowska, M. Grdeń, M. Kopczyk, G. Wójcik, G. Młynarek, J. Kołata, J.M. Skowroński

"Electrochemical Behavior of Nickel Deposited on Reticulated Vitreous Carbon (RVC)"

Journal of Power Sources, **77** (1999) 28-33 (IF(2011) = 4.951)

(my contribution -10%, SEM analysis, discussion of the results, participation in preparation of the manuscript)

2. M. Grdeń, A. Czerwiński, J. Golimowski, E. Bulska, B. Krasnodębska-Ostręga, R. Marassi, S. Zamponi

"Hydrogen electrosorption in Ni-Pd Alloys"

Journal of Electroanalytical Chemistry, **460** (1999) 30-37 (IF(2011) = 2.905)

(my contribution – 58%, preparation of the alloys; measurements of hydrogen absorption,

calculations and data analysis, participation in preparation of the manuscript)

3. M. Grdeń, J. Kotowski, A. Czerwiński

"The Study of Electrochemical Palladium Behavior by Quartz Crystal Microbalance. I. Acidic Solution" *Journal of Solid State Electrochemistry*, **3** (1999) 348-351 (IF(2011) = 2.131) (my contribution – 81%, Pd electrodeposition, CV and EQCM measurements, data analysis, calculations, preparation of the manuscript)

4. A. Czerwiński, I. Kiersztyn, M. Grdeń, J. Czapla

"The Study of Hydrogen Sorption in Palladium Limited Volume Electrodes (Pd-LVE) I. Acidic Solutions"

Journal of Electroanalytical Chemistry, **471** (1999) 190-195 (IF(2011) = 2.905)

(my contribution -8%, CV analysis, discussion of the results, participation in preparation of the manuscript)

5. M. Grdeń, J. Kotowski, A. Czerwiński

"The Study of Electrochemical Palladium Behavior by Quartz Crystal Microbalance. II. Basic Solution"

Journal of Solid State Electrochemistry, **4** (2000) 273-278 (IF(2011) = 2.131)

(my contribution - 81%, CV and EQCM measurements, Pd electrodeposition, data analysis, calculations, preparation of the manuscript)

6. A. Czerwiński, M. Żelazowska-Zakrent, **M. Grdeń**, K. Kuc, J.D. Milewski, A. Nowacki, G. Wójcik, M. Kopczyk

"Electrochemical behavior of lead in sulfuric acid"

Journal of Power Sources, **85** (2000) 49-56 (IF(2011) = 4.951)

(my contribution -8%, participation in data analysis and discussion of the results,

participation in preparation of the manuscript)

7. A. Czerwiński, I. Kiersztyn, M. Grdeń

"The Study of Hydrogen Sorption in Palladium Limited Volume Electrodes (LVE). II. Basic Solutions"

Journal of Electroanalytical Chemistry, **492** (2000) 128-136 (IF(2011) = 2.905)

(my contribution -8%, CV analysis, discussion of the results, participation in preparation of the manuscript)

8. M.Grdeń, A. Paruszewska A.Czerwiński

"Carbon Dioxide Adsorption on Pt-Pd Alloys"

Journal of Electroanalytical Chemistry, **502** (2001) 91-101 (IF(2011) = 2.905) (my contribution – 50%, participation in preparation of the electrodes, CV and CO₂ adsorption experiments, EDX measurements, data analysis, calculations, preparation of the manuscript)

9. M.Yu. Rusanowa, **M. Grdeń**, A. Czerwiński, G.A. Tsirlina, O.A. Petrii, T.Ya. Safonowa

"Isotope effects in α -PdH(D) as an instrument for diagnosis bulk defects" Journal of Solid State Electrochemistry, **5** (2001) 212-220 (IF(2011) = 2.131) (my contribution – 8%, participation in the measurements, and data analysis)

After Ph.D. title (total 24 items including 9 included in habilitation thesis)

10. M. Grdeń, A. Piaścik, Z. Koczorowski, A. Czerwiński "Hydrogen Electrosorption in Pd-Pt Alloys" *Journal of Electroanalytical Chemistry*, **532** (2002) 35-42 (IF(2011) = 2.905) (my contribution -51 %, participation in CV and CA experiments and deposition of the alloys, EDX analysis, data analysis, calculations, preparation of the manuscript) 11. M. Grdeń, K. Kuśmierczyk, A. Czerwiński "The Study of the Hydrogen Electrosorption in Pd-Ni Alloys by Quartz Crystal Microbalance" *Journal of Solid State Electrochemistry*, **7** (2002) 43-48 (IF(2011) = 2.131) (my contribution - 81%, preparation of the electrodes, CV, EQCM and EDX measurements, data analysis, calculations, preparation of the manuscript) 12. A. Czerwiński, I. Kiersztyn, M. Grdeń "Temperature influence on hydrogen sorption in palladium limited-volume electrodes (Pd-LVE)" Journal of Solid State Electrochemistry, 7 (2003) 321-326 (IF(2011) = 2.131) (my contribution -8%, CV analysis, discussion of the results, participation in preparation of the manuscript) 13. M. Łukaszewski, M. Grdeń, A. Czerwiński "Hydrogen insertion into Pd-Pt-Rh alloy limited volume electrodes (LVEs)" *Journal of Physics and Chemistry of Solids*, **65** (2004) 523-528 (IF(2011) = 1.632) (my contribution -8%, participation in data analysis and discussion of the results, participation in preparation of the manuscript) 14. A. Czerwiński, M. Grdeń, M. Łukaszewski "Dual mechanism of hydrogen desorption from palladium alloys postulated on the basis of cyclic voltammetric studies" *Journal of Solid State Electrochemistry*, **8** (2004) 411-415 (IF(2011) = 2.131) (my contribution -15%, participation in experiments with PdPt alloys, participation in data analysis and discussion, participation in preparation of the manuscript) 15. M. Łukaszewski, M. Grdeń, A. Czerwiński "Hydrogen electrosorption in Pd-Pt-Rh alloys in the presence of adsorbed CO" Analytical Letters, **37** (2004) 967-978 (IF(2011) = 1.016) (my contribution -8%, participation in data analysis and discussion, participation in preparation of the manuscript) 16. M. Łukaszewski, M. Grdeń, A. Czerwiński "Influence of adsorbed carbon dioxide on hydrogen electrosorption in palladium-platinumrhodium alloys"

Electrochimica Acta, **49** (2004) 3161-3167 (IF(2011) = 3.832)

(my contribution -15%, participation in experiments with PdPt alloys, participation in data analysis and discussion, participation in preparation of the manuscript)

17. M. Łukaszewski, M. Grdeń, A. Czerwiński

"Hydrogen electrosorption in Pd-Pt-Rh alloys"

Journal of Electroanalytical Chemistry, **573** (2004) 87-98 (IF(2011) = 2.905)

(my contribution -15%, participation in experiments with PdPt alloys, participation in data analysis and discussion, participation in preparation of the manuscript)

18. M. Grdeń, K. Klimek, A. Czerwiński

"A quartz crystal microbalance study on a metallic nickel electrode"

Journal of Solid State Electrochemistry, **8** (2004) 390-397 (IF(2011) = 2.131)

(included in habilitation thesis)

19. A. Czerwiński, M. Łukaszewski, **M. Grdeń**, H. Siwek, "The effect of carbon oxides on the absorption of hydrogen in palladium alloys"

Przemysł Chemiczny, **83** (2004) 508-512 (IF(2011) = 0.414)

(my contribution -7%, participation in data analysis and discussion, participation in preparation of the manuscript)

20. M. Grdeń*, K. Klimek

"EQCM studies on oxidation of metallic nickel electrode in basic solutions" Journal of Electroanalytical Chemistry, **581** (2005) 122-131 (IF(2011) = 2.905)

(included in habilitation thesis)

21. M. Łukaszewski, M. Grdeń, A. Czerwiński

"Cyclic voltammetric behavior of Pd-Pt-Rh ternary alloys"

Journal of Solid State Electrochemistry, 9 (2005) 1-9 (IF(2011) = 2.131)

(my contribution -8%, participation in data analysis and discussion, participation in preparation of the manuscript)

22. M. Alsabet, M. Grdeń, G. Jerkiewicz

"Comprehensive Study of the Growth of Ultra-Thin Oxide Layers on Pt Electrodes under Well-Defined Temperature, Potential, and Time Conditions"

Journal of Electroanalytical Chemistry, **589** (2006) 120-127 (IF(2011) = 2.905) Including erratum:

G. Jerkiewicz, M. Alsabet, M. Grdeń, H. Varela, G. Tremiliosi-Filho

"Erratum to: "Comprehensive study of the growth of thin oxide layers on Pt electrodes under well-defined temperature, potential, and time conditions" [Journal of Electroanalytical Chemistry, 589 (2006) 120–127]"

Journal of Electroanalytical Chemistry, **625** (2009) 172-174 (IF(2011) = 2.905) *(included in habilitation thesis)*

23. M. Łukaszewski, M. Grdeń, A. Czerwiński

"Comparative study on hydrogen electrosorption in palladium and palladium-noble metal alloys"

Journal of New Materials for Electrochemical Systems, **9** (2006) 409-417 (IF(2011) = 0.53) (my contribution -19%, experiments with PdPt alloys, participation in data analysis and discussion, participation in preparation of the manuscript)

24. M. Grdeń, K. Klimek, A. Czerwiński

"Quartz Crystal Microbalance studies on electrochemical behavior of electrodeposited Pd-Ni alloys"

Electrochimica Acta, **51** (2006) 2221–2229 (IF(2011) = 3.832)

(my contribution – 51%, EQCM, CV, EDX measurements, data analysis, calculations, preparation of the manuscript)

25. M. Łukaszewski, A. Żurowski, M. Grdeń, A. Czerwiński

"Correlations between hydrogen electrosorption properties and composition of Pd-noble metal alloys"

Electrochemistry Communications, **9** (2007) 671-676 (IF(2011) = 4.859)

(my contribution -17%, experiments with PdPt, participation in data analysis and discussion, participation in preparation of the manuscript)

26. M. Łukaszewski, M. Grdeń, A. Czerwiński

"Electrochemical studies on hydrogen absorption in palladium and its alloys with other noble metals"

Przemysł Chemiczny, **86** (2007) 137-142 (IF(2011) = 0.414)

(my contribution -12%, participation in experiments with PdPt alloys, participation in data analysis and discussion, participation in preparation of the manuscript)

27. M. Grdeń, A. Czerwiński

"EQCM studies on Pd-Ni alloy oxidation in basic solution"

Journal of Solid State Electrochemistry, **12** (2008) 375-385 (IF(2011) = 2.131)

(included in habilitation thesis)

28. M. Grdeń, M. Łukaszewski, G. Jerkiewicz, A. Czerwiński

"Electrochemical behaviour of palladium electrode: Oxidation, electrodissolution and ionic adsorption"

Electrochimica Acta, **53** (2008) 7583-7598 (IF(2011) = 3.832)

(review)

(my contribution -57%, selection and analysis of the literature, preparation of the manuscript)

29. M. Grdeń*

"Electrochemical Quartz Crystal Microbalance Studies of a Palladium Electrode Oxidation in a Basic Electrolyte Solution", *Electrochimica Acta*, **54** (2009) 909-920 (IF(2011) = 3.832) *(included in habilitation thesis)*

30. M. Grdeń*, K. Klimek, Z. Rogulski

"A quartz crystal microbalance study on oxidation of a cobalt electrode in an alkaline solution", *Electrochemistry Communications*, **11** (2009) 499-503 (IF(2011) = 4.859)

(included in habilitation thesis)

31. M. Alsabet, M. Grdeń, G. Jerkiewicz

"Electrochemical Growth of Surface Oxides on Nickel. Part 1: Formation of α -Ni(OH)₂ in Relation to the Polarization Potential, Polarization Time, and Temperature"

Electrocatalysis, 2 (2011) 317-330 (Impact Factor ma być przyznany w 2013 roku)

(included in habilitation thesis)

32. M. Grdeń, M. Alsabet, G. Jerkiewicz

"Surface science and electrochemical analysis of nickel foams"

ACS Applied Materials and Interfaces, **4** (2012) 3012-3021 (IF(2011) = 4.525)

(included in habilitation thesis)

33. M. Grdeń*, J. Jagiełło

"Oxidation of electrodeposited cobalt electrodes in an alkaline electrolyte" Journal of Solid State Electrochemistry, **17** (2013) 145-156 (IF(2011) = 2.131) (included in habilitation thesis)

b) Other publications (not included in JCR database)

 G. Wójcik, G. Młynarek, J. Kołata, M. Kopczyk, M. Dmochowska, M. Grdeń, M. Żelazowska-Zakrent, A. Czerwiński, J.M. Skowroński
 "Comparative Study of Electrochemical Behaviour of Nickel Carriers for Nickel Hydroxide Positive Electrodes in Alkaline Batteries" Polish Journal of Applied Chemistry, 44 (2000) 7-17

(my contribution – 8%, SEM analysis, participation in data analysis,

participation in preparation of the manuscript)

2. A. Czerwiński, I. Kiersztyn, M. Łukaszewski, M. Grdeń

"Hydrogen Sorption Behavior of Nanodeposits of Palladium and Its Alloys"

Electrocatalysis: Proceedings of the International Symposium (207th Meeting of The Electrocatalysis) *PNV 2005*, 11 (2005) *AC 54*

Electrochemical Society, Quebec City, 2005), PV 2005-11 (2005) 46-54

(my contribution -15%, participation in experiments with PdPt alloys, participation in SEM analysis, participation in data nalysis and discussion, participation in preparation of the manuscript)

3. M. Chotkowski, M. Grdeń, Z. Rogulski, A. Siporska

"Should we be afraid of ionising radiation - safe application of nuclear energy"

Chemistry in School (in polish), 5 (2010) 61-65

(my contribution -16%, preparation and execution of an experiment contributing to the series of laboratory courses, participation in data analysis)

c) Bibliographic data:

Total impact factor 2011 = 88.029; total impact factor according to the year of publication = 59.03

Number of citations without self citations of all authors = 354 (Web of Science, 24.05.2013) Number of all citations = 737 (Web of Science, 24.05.2013) h index without self citation of all authors = 10 (24.05.2013) h index with self citations = 17 (24.05.2013)

d. Conferences and meetings

oral presentations given by the author of self-presentation

1. **M. Grdeń**, K. Klimek, A. Czerwiński, "Electrochemical processes on nickel metal electrode studied by quartz crystal microbalance", 54th Annual Meeting of ISE, São Pedro, Brasil, 2003

2. **M. Grdeń**, K. Klimek, A. Czerwiński, "Quartz Crystal Microbalance Study on Hydrogen Absorption in Metallic and Alloy Electrodes", 55th Annual Meeting of ISE, Thesaloniki, Greece, 2004

3. **M. Grdeń**, "Electrochemical Quartz Crystal Microbalance Studies of a Palladium Electrode Oxidation in a Basic Electrolyte Solution", Young Author's Symposium of Canadian Section of The Electrochemical society, Kingston, Canada, 2008

4. M. Alsabet, **M. Grdeń**, G. Jerkiewicz, "Electrochemical Growth of β -Ni(OH)₂ and NiOOH on Ni Electrodes at Well-defined Polarization Potential, Polarization Time, and Temperature Conditions", 55th International Conference on Analytical Sciences and Spectroscopy, Kingston, Canada, 2009

presentations given by other co-authors and posters

1. A. Czerwiński, I. Kiersztyn, **M. Grdeń**, "Behavior of palladium electrodes deposited on gold", 47th Annual Meeting of ISE, Budapest, Hungary, 1996

2. I. Kiersztyn, **M. Grdeń**, "Behaviour of Palladium Electrode", International Conference of PhD Students, Miskolc, Hungary, 1997

3. **M. Grdeń**, I. Kiersztyn, M. Dmochowska, A. Czerwiński, "Hydrogen Electrosorption in Pd-Ni Alloys", XL Meeting of Polish Chemical Society, Gdańsk, Poland, 1997

4. **M. Grdeń**, I. Kiersztyn, J. Kotowski, A. Czerwiński, "Hydrogen Sorption in Pd-Ni electrodes", International meeting Electrochemistry at the turn of XX century:Industry, Bioand Surface Science, Cracow, Poland, 1998

5. A. Czerwiński , **M. Grdeń**, I. Kiersztyn, J. Kotowski, M. Żelazowska-Zakrent, "Hydrogen Sorption in Pd, Ni and Pd-Ni Electrodes with Limited Volume (LVE)", 49th Annual Meeting of ISE, Kitakyushu, Japan, 1998

6. G. Wójcik, M. Kopczyk, J.M. Skowroński . A. Czerwiński, M. Dmochowska, **M. Grdeń**, "Electrochemical Behavior of Nickel deposited on Reticulated Vitreous Carbon (RVC)", 21th Conference on Chemical Current Sources, Brno, Czech Republic, 1998

7. G. Wójcik, M. Kopczyk, J.M. Skowroński, A. Czerwiński, M. Dmochowska, **M. Grdeń**, "Electrochemical Behavior of Nickel deposited on Reticulated Vitreous Carbon (RVC)", 8th International Ni-Cd Conference, Prague, Czech Republic, 1998

8. G. Wójcik, M. Kopczyk, J.M. Skowroński A. Czerwiński, M. Dmochowska, **M. Grdeń**, "Electrochemical Behavior of Nickel deposited on Reticulated Vitreous Carbon (RVC)", International Interbat Conference, Poznań, Poland, 1998

 A. Czerwiński, I. Kiersztyn, M. Grdeń, J. Czapla, "The study of hydrogen sorption in palladium electrodes with limited volume (LVE)", 14th International Symposium of Electrochemical Section of Polish Chemical Society, Poraj near Częstochowa, Poland, 1999
 A. Czerwiński, M. Żelazowska-Zakrent, M. Grdeń, K. Kuc, J.D. Milewski, A. Nowacki, G. Wójcik, M. Kopczyk, "Electrochemical behavior of lead in various sulfuric acid solutions", 14th International Symposium of Electrochemical Section of Polish Chemical Society, Poraj near Częstochowa, Poland, 1999

11. A. Czerwiński, M. Żelazowska, **M. Grdeń**, K. Kuc, J.D. Milewski, A. Nowacki, G. Wójcik, M. Kopczyk, "Electrochemical Behavior of Lead in Different Concentrations of Sulfuric Acid", International Conference on Lead-Acid Batteries LABAT ' 99, Sofia, Bulgaria, 1999

12. A. Czerwiński, M. Żelazowska, **M. Grdeń**, K. Kuc, J.D. Milewski, A. Nowacki, G. Wójcik, M. Kopczyk, "Electrochemical Behavior of Lead in Different Concentrations of Sulfuric Acid", 50th Annual Meeting of ISE, Pavia, Italy, 1999

 M. Grdeń, J. Kotowski, A. Czerwiński, "The Study of Hydrogen Electrosorption in Palladium by Quartz Crystal Microbalance", 50th Annual Meeting of ISE, Pavia, Italy, 1999
 M. Grdeń, A, Paruszewska, A. Czerwiński, "Carbon dioxide Adsorption on Pd-Pt Alloys", 51th Annual Meeting of ISE, Warsaw, Poland, 2000

15. I. Kiersztyn, **M. Grdeń**, A. Czerwiński, "Influence of Temperature on Hydrogen Sorption in Palladium Electrodes with Limited Volume (Pd-LVE)" 51th Annual Meeting of ISE, Warsaw, Poland, 2000

 M. Grdeń, A. Czerwiński, "Hydrogen Electrosorption in Binary Palladium Aloys", Electrochem 2000, ISE Sponsored Scientific Meetings, Dublin, Republic of Ireland, 2000
 M. Grdeń, J. Kotowski, K. Kuśmierczyk, A. Czerwiński, "Electrochemical Behaviour of Binary Palladium Alloys", 52nd Annual Meeting of ISE, San Francisco, USA, 2001
 M. Grdeń, A. Czerwiński, K. Kuśmierczyk, "Oxidation of CO2 electrosorption products on Pd-Pt alloy electrodes", 53rd Annual Meeting of ISE, Düsseldorf, Germany, 2002
 M. Grdeń, K. Klimek, A. Czerwiński, "Quartz crystal microbalance study on nickel metal electrode", 3rd International Baltic Conference on Electrochemistry, Gdańsk, Poland, 2003
 A. Czerwiński, M. Grdeń, M. Łukaszewski, "Limited Volume Electrodes (LVEs). Study of hydrogen sorption in palladium alloys", 203rd Annual meeting of The Electrochemical Society, Paris, France, 2003

21. A. Czerwiński, **M. Grdeń**, M. Łukaszewski, "Electrochemical behavior of palladium alloys limited volume electrodes (LVE)", 54th Annual Meeting of ISE, São Pedro, Brasil, 2003

22. M. Łukaszewski, **M. Grdeń**, A. Czerwiński, "Carbon oxides adsorption as diagnostic tools in studies on hydrogen electrosorption in Pd-Pt-Rh alloys", 54th Annual Meeting of ISE, São Pedro, Brasil, 2003

23. R. Pruszkowska-Drachal, H. Siwek, I. Paleska, J. Kotowski, **M. Grdeń**, A. Czerwiński, "Investigation of antimony-lead alloys electrodes in sulfuric and phosphoric acids", 54th Annual Meeting of ISE, São Pedro, Brasil, 2003

24. Z. Rogulski, **M. Grdeń**, D. Żochowska, A. Czerwiński, "Electrochemical oxidation of Mn²⁺ ions on Pt, Au and RVC® electrodes", 54th Annual Meeting of ISE, São Pedro, Brasil, 2003

25. Z. Rogulski, **M. Grdeń**, A. Czerwiński, "Reticulated Vitreous Carbon (RVC®) new current collector in the zinc-manganese dioxide cells", 54th Annual Meeting of ISE, São Pedro, Brasil, 2003

26. A. Czerwiński, K. Klimek, **M. Grdeń**, "Electrochemical Quartz Crystal Microbalance Study on Behavior of Pd and Ni Electrodes", International Conference on Electrode Processes, Szczyrk, Poland, 2004

27. M. Łukaszewski, A. Czerwiński, **M. Grdeń**, "EQCM Studies on Hydrogen Absorption in Metals and its Alloys", International Symposium on Metal-Hydrogen Systems; Fundamentals & Applications, Cracow, Poland, 2004

28. A. Czerwiński, M. Łukaszewski, **M. Grdeń**, "Application of Limited Volume Electrodes (LVEs) for Examination of Hydrogen Absorption in Palladium and its Alloys", International Symposium on Metal-Hydrogen Systems; Fundamentals & Applications, Cracow, Poland, 2004

29. M. Łukaszewski, **M. Grdeń**, A. Czerwiński, "Electrosorption of Hydrogen into Palladium-Platinum-Rhodium Alloys", 55th Annual Meeting of ISE, Thesaloniki, Greece, 2004

30. A. Czerwinski, I. Kiersztyn, M. Łukaszewski, M. Grdeń, "Hydrogen Sorption Behavior of Nanodeposits of Palladium and Its Alloys", 207th ECS Meeting, Quebec, Canada, 2005
31. M. Grdeń, G. Jerkiewicz, "Electrochemical Behavior of Micro-Structured Nickel Materials", Nanoforum Canada – 2nd Canadian Nanoscience & Nanotechnology Forum,

Montreal, Canada, 2005

32. G. Jerkiewicz, **M. Grdeń**, M. Alsabet, E. Sourty, "3D Electron Tomography Characterization of Nickel Foams and Their Electrocatalytic Activity", 57th Annual Meeting of ISE, Edinburgh, UK, 2006

33. G. Jerkiewicz, **M. Grdeń**, M. Alsabet, E. Sourty, "Kinetics and Mechanisms of the Growth of Thin Oxides on Pt-Group Metals: Combination of Experiments and Data Modeling", 57th Annual Meeting of ISE, Edinburgh, UK, 2006

34. M. Alsabet, **M. Grdeń**, G. Jerkiewicz, "Growth of Monolayer Oxides on Ni Electrodes in Aqueous KOH at Well-Defined Potential, Time and Temperature Conditions", 58th Annual Meeting of ISE, Banff, Canada, 2007

35. J. Lessard, **M. Grdeń**, G. Jerkiewicz, M. Obradovic, "Under-Potential Deposition of Hydrogen on C_6H_{6ads} -Modified Pt(111) in Aqueous HClO₄", 58th Annual Meeting of ISE, Banff, Canada, 2007

36. M. Alsabet, **M. Grdeń**, G. Jerkiewicz, "Comprehensive Study of the Growth of Monolayer Oxides on Pt Electrodes in Aqueous H_2SO_4 at Well-Defined Potential, Time and Temperature Conditions ", 58th Annual Meeting of ISE, Banff, Canada, 2007

37. G. Jerkiewicz, M. Grdeń, "Materials Science and Electrochemical Characterization of Microstructured Nickel Foam", 58th Annual Meeting of ISE, Banff, Canada, 2007
38. G. Jerkiewicz, M. Grdeń, M. Alsabet, E. Sourty, "Electron Tomography Analysis and Electrochemical Characterization of Dual Porosity Nickel Foams", Nanoforum Canada – 3rd

Canadian Nanoscience & Nanotechnology Forum, Waterloo, Canada, 2007

39. **M. Grdeń**, "Electrochemical Characterization of Microstructured Nickel Foam", Young Author's Symposium of Canadian Section of The Electrochemical society, Kingston, Canada, 2008

40. **M. Grdeń**, G. Jerkiewicz, "Electrochemical and Surface Science Characterization of Nickel Foams", 7th Spring Meeting of ISE, Szczyrk, Poland, 2009

41. **M. Grdeń**, "The Interfacial Capacitance of a Palladium Electrode in an Alkaline Electrolyte", 7th Spring Meeting of ISE, Szczyrk, Poland, 2009

42. J. van Drunen, G. Jerkiewicz, **M. Grdeń**, "Characterization of Nickel-containing Foams Using Focused Ion Beam, Electron Microscopy and Electrochemical Techniques", 55th International Conference on Analytical Sciences and Spectroscopy, Kingston, Canada, 2009 43. **M. Grdeń**, M. Alsabet, G. Jerkiewicz, "Surface and Materials Science, and

Electrochemical Analysis of Nickel Foams", 2nd Ertl Symposium, Stuttgart, Germany, 2012

e) Patents

1. A. Czerwiński, M. Dmochowska, **M. Grdeń**, G. Wójcik, G. Młynarek, M. Kopczyk, J.M. Skowroński, "Nickel electrode for electrochemical power sources", Patent of RP Nr185542 (1998)

2. A. Czerwiński, M. Grdeń, "Hydrogen storage material", Patent of RP Nr 184549 (2002)

6. Other information

a) Teaching:

Physical chemistry – laboratory, basic level (Physical Chemistry I, undergraduate studies) and advanced level (Physical Chemistry II, undergraduate studies)

Nuclear Chemistry - laboratory (undergraduate studies)

Participation in preparation of lectures and laboratories for Nuclear Energy and Chemistry degree course organized by Faculties of Chemistry and Physics of University of Warsaw M.Sc. thesis supervisor and/or adviser of 7 completed M.Sc. thesis in chemistry (1998-2011)

b) Research grants:

1. Research project MNiSW N204 125037 "Components of the interfacial capacitance at the oxide/hydroxide-aqueous electrolyte interface" (2009-2013) (grant leader/head of the project and principal investigator)

2. Research project KBN 3-T09A-003-19 "Hydrogen electrosorption in multicomponent palladium-noble metals alloys" (2000-2002) (principal investigator 2)

3. Research Project of 6 Framework NMP3-CT-2006-032517 "HydroNanoPol - Advancement in storage capability and Hydrogen kinetics of hydride storage alloys through nanocoating with multifunctional hybrid polymer" (2007-2009) (investigator)

c) Scientific practices

1. 11.2004-10.2005; 02.2006 – 02.2007; 07-09.2008; 07-09.2009 (total ca. 2,5 years) – postdoctoral research and scientific trainings in laboratory of Prof. G. Jerkiewicz, Department of Chemistry, Queen's University, Canada

2. 02. 1999 – one week scientific practice in laboratory of Prof. H. Baumgärtel, Institute of Chemistry and Biochemistry - Physical and Theoretical Chemistry, Freie Universität, Berlin, Germany

3. 02-03.1998 – scientific training (few weeks) in laboratory of Prof. R. Marassi, Department of Chemical Sciences, University of Camerino, Italy

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