



# Past and present of electrochemical science in Hungary

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## Abstract

The electrochemistry-related scientific activities in Hungary over the past 3 decades are reviewed. In the first section, we summarize those research areas that are already ceased; in the next section, the ongoing research is discussed; finally, the trends and outlook are highlighted. A special emphasis is put on new experimental methods elaborated in the country.

## Introduction

The history of Hungarian electrochemistry along with its background can perhaps be best shown through the life and activities of the outstanding electrochemist-scientists, just as was recently done by György Inzelt [1]. Out of the many outstanding scientists listed therein, two of them, Tibor Erdey-Grúz (1902–1976) [1], [2, page 352] and Ernő Pungor (1923–2007) [1], [2, page 770] [3], were active in the twentieth century, established two major directions of electrochemistry in Hungary. Their schools: students, followers, and “scientific descendants”—and the ideas they introduced and represented are still noteworthy. One of these directions was electrode kinetics, studied mainly by the school of Erdey-Grúz; the other is certain aspects of electroanalytical chemistry, pursued by people of the school of Pungor.

A previous snapshot of the state of Hungarian electrochemistry can be read in an invitation letter [4] to the 47th International Society of Electrochemistry meeting, held in Veszprém and Balatonfüred, Hungary, in 1996. This letter has been written by the principal local organizer, György Inzelt, now the doyen of the Hungarian electrochemists. It comprises a two-page-long overview of the electrochemical science in Hungary up to the date of the conference.

The present article reviews activities since 1996, starting where [4] has been finished. Since 1996, many of the scientists mentioned in [4] have finished related activities and have retired or are deceased; these activities are summarized in the “[Finished activities](#)” section; the present,

ongoing activities are reviewed in the “[Ongoing activities](#)” section. Finally, the trends and future prospects are listed in the “[Summary and outlook](#)” section.

## Finished activities

Important electrochemical studies and laboratories were at research institutes<sup>1</sup> of the Hungarian Academy of Sciences.

Probably the most efficient and best-known electrochemist of the early years of the twenty-first century was György Horányi (†2006) [1], [2, page 461]. He performed radiotracer experiments to clarify details of adsorption (electrosorption) and electrocatalytic phenomena at electrode surfaces and of ion-exchange processes in polymer-modified electrodes. In particular, important are his studies on sulfate adsorption on many diverse metals and oxide surfaces [5].

Erika Kálmán (†2009) with her group studied the electrochemical corrosion of metals with a focus on corrosion inhibition phenomena, e.g., of phosphonates on steel in neutral aqueous electrolytes, in the context of the structure of the surface layer [6]. The effect of self-assembling on corrosion protection as well as the kinetics of self-assembling have been characterized in [7] and [8], respectively. An analysis of the role of biofilms in corrosion processes is also noteworthy [9]. People of this group recently participated in a long project characterizing carbon nanotube arrays in electrochemical capacitors [10].

Underpotential deposition of metals and fundamental aspects of metal corrosion kinetics have been studied by Sándor Szabó (†2018) [11, 12]. Besides the investigation of electrochemical noise [13], corrosion protection properties

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of various inhibitors [14], and organic coatings [15] were characterized, electrochemical aspects of electrocoagulation have been analyzed [16] by the group of Béla Lengyel.

For understanding relaxation processes within the electrochemical double layer, impedance studies have been performed on noble metals in aqueous solutions and in ionic liquids by Tamás Pajkossy [17].

There have been a number of groups at major Hungarian universities whose electrochemistry-related activities were significant. They are as follows:

Following the long-term traditions in the Department of Analytical Chemistry at the Technical University Budapest,<sup>2</sup> headed for a long time by Ernő Pungor, the group of Klára Tóth (†2013) worked on the theory and utilization of ion-selective electrodes, in particular of ones with bis-crown ether, calixarene, etc., derivatives in polymer membranes. These are selective to  $K^+$ ,  $Na^+$ ,  $Pb^{2+}$ ,  $H^+$ , and  $Zn^{2+}$  ions. Extensive efforts were made to characterize various physicochemical properties of these membranes; characteristic publications are [18–20]. The members of the group made significant contributions to flow analytical methods [21]. Some of the sensor and method developments were supported by the electrochemical instrumentation company, Radelkis (managed by Jenő Havas) and reached industrial production [22].

György Farsang (†2009), professor of analytical chemistry at the Eötvös Loránd University, Budapest, studied anodic reactions (electrochemical oxidation and dimerization) of various halogenated aniline compounds in organic solutions using microelectrodes and high scan rate cyclic voltammetry [23].

Pál Joó (†2022), professor of colloid chemistry at the University of Debrecen up till his retirement in 2007, studied mostly adsorption onto and transport processes through colloidal systems by electrochemical and radiochemical methods [24].

Nonlinear dynamics of oscillatory and chaotic metal electrodisolution were studied both experimentally and theoretically by Vilmos Gáspár and István Z. Kiss at the University of Debrecen [25, 26]. Nonequilibrium phase diagrams were constructed, and their dependence on various experimental parameters, such as mass transport conditions and solution resistance, etc., were determined. Control and synchronization algorithms, including neural networks, were developed for the chaotic dynamics of single and multielectrode systems.

Kálmán Varga (†2009) at the University of Veszprém<sup>3</sup> has studied corrosion and radioactive contamination processes

on metal surfaces typical in primary circuits of nuclear power plants [27]. Apart from these, other combined radiochemical and electrochemical electrosorption experiments have been performed with noble metal surfaces like the one in [28].

Csaba Visy at the University of Szeged has studied electrically conductive polymers, charge transport, and structural transformations therein using in situ combined spectroelectrochemical methods since the late 1980s [29, 30]; since the millennium, the main research direction has been the production, characterization, and application possibilities of composite materials based on conducting polymers [31]. Such composites with special magnetic, photocatalytic, thermoelectric, and biocatalytic properties have been successfully produced; a characteristic example is in [32].

László Dobos (†2008), electrical engineer at the University of Szeged, manufactured a number of potentiostats for Hungarian universities and research institutions, according to their specific needs. In the 1990s, his affordable computer-controlled potentiostats were practically on the same technical level as the commercial potentiostats made in Western countries.

## Ongoing activities

### Electrode kinetics

Following many decades of “Erdey-Grúz tradition” in the physical chemistry department of the Eötvös Loránd University, Budapest, most of the research activities are related to electrode kinetics. At present, there are three main research directions in that department, mostly associated with three senior electrochemists:

György Inzelt has been studying the deposition, properties, and transformations of various electroactive layers (conducting polymers, various metal phthalocyanines, lead sulfate, etc.) on inert electrodes within an electrochemical quartz crystal microbalance (EQCM). Important summarizing publications on conducting polymers are [33] and [34].

Győző G. Láng and coworkers have been studying polymer film electrodes, mostly poly(3,4-ethylenedioxythiophene) (PEDOT), in particular, it is aging as well as overoxidation and its consequences [35]. From an environmental protection point of view important are their studies on perchlorate reduction on various metals [36].

Soma Vesztergom constructed a mathematical model of hydrogen evolution from a mildly acidic, unbuffered, stirred aqueous solution when the hydrogen gas stems both from  $H^+$  ions and from water. This model's predictions are in accord with hydrogen evolution experiments carried out with RDE [37], also even if hydrogen evolution proceeds parallel to a metal deposition [38].

<sup>2</sup> At present, the name of TU Budapest is Budapest University of Technology and Economics.

<sup>3</sup> At present, its name is University of Pannonia.

The research activities of Janáky and his group at the University of Szeged have been focusing on semiconductor photoelectrochemistry. New photoelectrode assemblies were designed for energetics-related processes (e.g., water oxidation and CO<sub>2</sub> reduction) [39, 40], or to drive value-adding processes yielding chemicals or fuels. A new instrument—an in situ ultrafast spectroelectrochemical tool—was constructed for tracking light-induced processes at the fs-ps timescale under electrochemical control [41] and was used for the study of optically active perovskites.

Tamás Pajkossy has recently reconsidered the theories of electrode charge transfer in cases of diffusional and coverage hindrances, for conditions of both voltammetry and impedance spectroscopy. His theories yield simple, exact equations [42].

Theories of double-layer structure play a big role in understanding electrode kinetics. Dezső Boda and coworkers at the University of Veszprém<sup>3</sup> have been doing Monte Carlo simulations for the properties of various structures having electrochemical relevance: electrical double layers [43], ion channels [44], and ion atmospheres [45]. In their models, the ions of the electrolyte are charged hard spheres distributed in an implicit continuum solvent. With this—apparently simple—modeling even complex phenomena can be explained, provided that the important degrees of freedom are included in the model [46].

## Electroanalysis

The development of chemical sensors and biosensors based on electrochemical transducers and “nanoelectrochemistry” is the main research interest of Róbert Gyurcsányi’s group at the TU Budapest<sup>2</sup>. Novel sensing concepts, materials, and investigation methods have been introduced to support the application of ion sensors for ultra-trace analysis, bioanalysis, and field deployment. Perhaps the most important innovation was the development of the selective solid-state ion channels based on chemically modified nanopores [47], which also enabled the use of hydrophilic complexing agents of biological origin for the potentiometric measurement of ions and polyions [48]. Through novel nanoscale electrochemical sensing and synthetic concepts, they made possible the detection of single entities/particles such as viruses [49] as well as the generation of affinity ligands for selective protein recognition based on surface-imprinted electropolymerized nanofilms [50].

The group of Géza Nagy at the University of Pécs contributed to the advancement of scanning electrochemical microscopy by employing multi-barrel measuring tips containing ion-selective microelectrodes [51] in potentiostatic mode to studies related to corrosion and life sciences. They have been also elaborating and using various amperometric and potentiometric methods for many and diverse analytical

purposes; a characteristic example is concentration determination in tortuous media [52].

## Applied electrochemistry

The fundamental research on electrode kinetics made at Eötvös Loránd University had also practical outcomes. The development of hydrogen-based fuel cells and their application were also a field of activity of György Inzelt’s group [53]. Győző G. Láng and coworkers elaborated a procedure for the electrochemical generation of ferrate salts [54, 55], which can eliminate even the chlorinated organic compounds, e.g., from polluted groundwater.

The staff of the electrochemistry laboratory of the Institute of Solid State Physics<sup>4</sup> in Budapest has long been dealing with various aspects of electrodeposition of metals. Imre Bakonyi initiated the studies of the electrodeposition of nanocrystalline metals in the early 1990s [56] as well as that of ferromagnetic/nonmagnetic multilayers exhibiting giant magnetoresistance. The latter was elaborated with László Péter, leading to an important review of the topic [57]. László Péter continued the metal electroplating studies with composition depth profile analysis of electrodeposited alloys and multilayers [58], and fine structural studies of various metals and alloys [59].

A group of the KFKI Atomic Energy Research Institute<sup>5</sup> in Budapest has been studying corrosion-related problems appearing in primary circuits of the pressurized water nuclear power plants. Their representative publication is [60], and remarkable technical details are in [61].

Magda Lakatos-Varsányi (Bay Institute, Budapest) made various metal layers by pulse plating (e.g., [62]) and characterized metals from the corrosion point of view (e.g., [63]).

The Corrosion Research Group of the University of Veszprém<sup>3</sup> has been focusing its activities on industrial corrosion prevention. The group regularly performs industrial corrosion root-cause analyses. Recent academic research includes corrosion testing of structural materials of the oil industry [64] and improvements in evaluation methods for corrosion-related electrochemical techniques [65].

Balázs Endrődi and Csaba Janáky, with their group at the University of Szeged, perform studies of industrial electrochemical processes, mainly water splitting and CO<sub>2</sub> reduction, under industrially relevant conditions. New electrolyzer cell and stack architectures [66], membrane electrode assemblies, and operational protocols [67] have been developed.

<sup>4</sup> At present, it is a part of the Wigner Research Centre of the Eötvös Loránd Research Network.

<sup>5</sup> At present, it is a part of the Centre for Energy Research of the Eötvös Loránd Research Network.

Development of catalysts for the PEM fuel cell anodes is done at the Institute of Materials and Environmental Chemistry<sup>1</sup>, Budapest, in the group of András Tompos. As it can be demonstrated, the Pt catalyst becomes more stable and CO tolerant if the support is a mixed Ti–W oxide [68] or Ti–Mo oxide [69] on activated carbon rather than the usual pure activated carbon.

A form of local corrosion in electronic circuits is caused by electromigration leading to dendritic shortcuts between metal stripes, which finally result in the failure of the circuit. Electromigration phenomena are studied at the Department of Electronic Technology of the TU Budapest<sup>2</sup> [70].

## Instrument development

In general, in most laboratories, factory-built instruments are used. Nevertheless, a couple of special devices/setups have been assembled—or are under development. These are as follows:

A special setup for controlling an RRDE has been assembled by Soma Vesztergom. The potential of the two working electrodes can be controlled individually; e.g., voltammetry with different potential programs can be measured on them [71]. A characteristic example of its use is the study of metal dissolution kinetics [72].

Surface charge density can be measured through the changes in surface stress that lead to small deformations [73]. A suitable optical arrangement called a “bending beam” has been assembled by Győző G. Láng recently; the method has been demonstrated on various systems [74].

With an optically transparent (e.g., ITO-based working electrode) of an EQCM, simultaneous measurement of surface mass changes, optical absorbance spectra, and voltammetry is possible. Such a combination has been implemented for the study of deposition and electrochemical transformations of polyaniline films by György Inzelt and coworkers [75]. Another electrode construction has been developed by Csaba Visy and coworkers by which simultaneous measurement of optical absorbance spectra, voltammetry, and conductivity of an electrochemically generated polymer film can be performed [76].

A bipotentiostat with a high dynamic range (i.e., with logarithmic current-to-voltage converters of 0.1 pA to 100 mA ranges) has been developed by Gábor Mészáros (RCNS IMEC) for various scanning microprobe measurements [77] and also for molecular conductance measurements [78]. About a dozen of these are in operation in Central-European research laboratories.

A dEIS (dynamic electrochemical impedance spectroscopy) measurement setup and the program has been developed by which audio-frequency impedance spectra can be taken repetitively at a fast rate which enables the taking

of a number of impedance spectra during the run of cyclic voltammograms [79].

An electrochemical hydrogen permeation method, based on the measurement of the temporal evolution of the hydrogen breakthrough rate, has been elaborated by László Péter in the physics institute<sup>4</sup> for characterizing enamel-grade steel plate products. This method was implemented in 2003 at DUNAFERR, the largest steel company in Hungary at that time.

## Monographs and handbooks

A few electrochemistry textbooks, handbooks, and monographs have been recently written by Hungarian authors. Perhaps the best known is the Electrochemistry Dictionary [2], one of the editors of which was György Inzelt. This book has had already two editions (in 2008 and an extended version in 2012); it is widely accepted and has received excellent reviews. This book, in fact, is an invaluable resource of knowledge of concepts, terms most used in electrochemistry, and the biography of scientists who greatly contributed to electrochemistry. It may serve as a reference for the whole electrochemist community.

Four monographs were written by Hungarian authors for a less broad professional field of interest. They are as follows, in the temporal order of their appearance:

The first, “Laser Techniques for the Study of Electrode Processes” [73], was co-authored by Győző G. Láng. Described therein are *in situ* techniques for the investigation of interfacial processes for those cases when there is surface geometry change due to the change of interfacial tension or there appears a refractive index gradient at the vicinity of a solid–liquid interface.

The second monograph, Handbook of Reference Electrodes [80], co-edited by György Inzelt, filled a half-century-old gap: since 1961 (when the last book on this subject appeared), many novel electrochemical systems appeared in the laboratory, which need appropriate reference electrodes.

The third book, “*In situ* Combined Electrochemical Techniques for Conducting Polymers” [81], is authored by Csaba Visy. This book surveys the advantages of combinations of *in situ* electrochemical techniques for analyzing the properties of conducting polymers.

The fourth monograph, “Electrochemical Methods of Nanostructure Preparation” [82], written by László Péter, summarizes the electrochemical routes of synthesizing nanostructures, including electrodeposition, anodization, carbon nanotube preparation, and other methods.

Two electrochemistry textbooks, in Hungarian, appeared in the past 2 decades [83, 84]. They both are used in universities for physical chemistry courses.

## Summary and outlook

In the previous sections, a number of scientists have been mentioned by name. These are the colleagues who have (or had) smaller or larger groups, have done or have been doing electrochemical research typically for decades, and published enough to be “visible” when scientometric figures are considered. The name of their colleagues, in most cases, appears as co-authors in the publication list. In addition, there are many colleagues, not mentioned by name, who contributed to electrochemistry temporarily, worked, e.g., on industrial problems leading to no publication, or carried out the tiresome task of educating students to obtain basic laboratory skills. Also, these colleagues played an important role in maintaining the professional culture of electrochemistry.

The above list of topics reveals the continuity of research traditions in Hungary. On the one hand, the good news is that there are many researchers who deeply understand the structure of metal–electrolyte interfaces and the processes therein, along with their practical implications. The study of ion-selective electrodes was and still is important in electroanalysis. There are still inventive researchers who put together novel instruments. In three major universities, there are three young, ambitious, gifted electrochemist colleagues (Róbert E. Gyurcsányi, Soma Veszteg, and Csaba Janáky, each holding a prestigious “Momentum” fellowship; Janáky has also an ERC grant); they can (and do) build up research groups with bright students. In addition, these three people are the ones who have strong international connections for cooperation, mostly in the USA, Switzerland, and elsewhere in Europe. The electroanalytical chemistry community is stable enough to run a conference series (the Mátrafüred International Conference on Electrochemical Sensors) since 1972.

On the other hand, unfortunately, the bad news is that, just as in the past, certain research areas are missing or are of low intensity, like the development of novel batteries and electrochemical technologies. Since the publication of [1] almost 3 decades ago, the number of electrochemical groups and scientists in Hungarian research sites has approximately halved. Funding has also decreased; brain drain from Western countries is strong; less and less students choose electrochemistry as a specialization. There are universities where, as there are no electrochemist faculty members, only the most necessary basic textbook knowledge is taught, and the students do not get the “hands-on” laboratory experience. Only a few students choose a Ph.D. topic related to electrochemistry. The lack of funding leads to the lack of appropriate instruments, in particular the facilities for the appropriate surface analyses.

As it is often claimed, the flourishing electrochemistry in Eastern Europe can be rationalized by the statement that

electrochemistry is the “physical chemistry of the poor”; i.e., conducting electrochemical experiments is often the cheapest/simplest way to demonstrate physicochemical laws. Though Hungary is still not rich, electrochemistry apparently, is gradually losing its importance because of various reasons: One of the leading branches of electrochemistry, electroanalysis, including the various polarographic techniques, has been relegated to the background because better, more practical analytical methods (ICP and X-ray fluorescence) appeared. Several large-scale industries that require electrochemical background knowledge (e.g., aluminum electrolysis, light bulb production) have ceased to exist; other factory laboratories have also become unnecessary and hence were closed. Therefore, as need ceases, the money spent on laboratory education also decreases; instead, it is spent on teaching other “less expensive” skills, like computer simulations. At the same time, the general prestige of that particular discipline also decreases—even the electrochemist colleagues become prone to identify themselves as chemists dealing with nanoscience (or similar).

Nevertheless, the overall situation, though not very bright, is far from disappointing. Some industries occasionally may require electrochemical research to solve ad-hoc problems. During the application of new technologies, new corrosion problems always appear. Electrochemists might be needed to train the staff of the large battery factories planned for Hungary. These indicate that there will be a need for electrochemical research, knowledge, and education also in the future.

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