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Improving education in electrochemistry via a modeling approach and focusing on green chemistry applications

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ABSTRACT

Chemistry is understood by students and practiced by research chemists through models and visualization. Electrochemistry is no exception. This study shows how a modeling approach focusing on green chemistry applications holds great potential in improving electrochemistry education. Green electrosynthesis experiments, for example, allow to largely enhance student's interest and learning in this cross-disciplinary field of chemical knowledge.

1. Introduction

Being the key scientific discipline which originates the Li-ion battery and the hydrogen fuel cell, namely the two key electrochemical technologies for the storage of electricity sourced from highly intermittent renewable energy sources such as sunlight and wind, electrochemistry plays a central role in the unfolding energy transition (Pagliaro and Meneguzzo, 2019). Furthermore, after decades of limited applications chiefly in the production of bulk chemicals, electrochemistry plays an increasingly important role also in the synthesis of valued fine chemicals (Wiebe et al., 2018).

Undergraduate education in electrochemistry needs to be improved in most world's countries. Suggesting avenues to reinvigorate electrochemistry education based on today's inexpensive electronics and open-source software, Kempler and co-workers recently reported that “rigorous training of electrochemists is generally lacking at academic institutions in the United States” (Kempler et al., 2021). Similar findings were reported in 2019 by Tsaparlis in Greece noting that due to complexity and plurality of its concepts electrochemistry was a “hard subject for students” (Tsaparlis, 2019). Indeed, Tsaparlis' study opens with a quotation of Japan's electrochemistry scholar Akiro Yoshino (awarded the Nobel Prize in chemistry in the same year for the development of lithium-ion batteries): “battery technologies are electrochemistry, a complex and difficult interdisciplinary field” (Yoshino cit and Looi, 2018).

Electrochemistry, noted Fergus in 2012 (Fergus, 2012), is not typically identified as a separate academic discipline and formal electrochemistry educational programs were still lacking both in the USA and abroad. Nearly ten years later, talking at the 2019 meeting of the Electrochemical Society, one manager from a reputed electrochemical research instrumentation manufacturer reported how their experience “of repeatedly filling in basic knowledge gaps suggests there may some simple, practical steps that can be taken with those who are beginning electrochemical research” (Spinner et al., 2019).

For decades following the end of World War II, the main applications of electrochemistry research have been electrolysis for the production of various elements, chemical compounds, and organic compounds including aluminum, chlorine and NaOH (Grotheer et

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al., 2006), lead batteries, and electrochemical sensors that find widespread use in environmental chemistry (*i.e.*, electrochemical pH and O_2 concentration measurements) and biomedicine (*i.e.*, sensors for glucose).

The Li-ion battery first (Pagliaro and Meneguzzo, 2019), and new electrochemical processes applied to fine chemical manufacturing (Wiebe et al., 2018), however, have brought electrochemistry to the frontier of today's chemical research. Suddenly, recognizing the poor state of electrochemistry education in many countries, electrochemistry and chemistry education scholars started to publish a number of intervention studies on electrochemistry education (Kempler et al., 2021; Tsaparlis, 2019; Locatelli and Arroio, 2014; Niaz, 2002; Schmidt et al., 2022; Fruehwald et al., 2021).

In brief, evidence from numerous countries suggests that novel and better approaches to teaching and learning electrochemistry are required.

Chemistry is understood and practiced by research chemists by models and visualization (Pagliaro, 2010; Chang, 2022; Locatelli and Arroio, 2014). Electrochemistry is no exception. This study shows how a modeling and visualization approach focusing in particular on green chemistry applications holds great potential in improving electrochemistry education. Green electrosynthesis, we argue in particular, is ideally suited to understand electrochemical phenomena and to foster student's interest and learning in this cross-disciplinary field of chemical knowledge.

2. Modeling approach

Recounting the history of the Li-ion battery joint discovery ("a fruit of collective wisdom") (Yoshino, 2019), Yoshino noted how, looking for a suitable battery cathode, in 1983 he read the 1980 study of Goodenough introducing the lithium cobalt oxide ($LiCoO_2$) cathode enabling a 4.0 V rechargeable battery of unprecedented high energy density (Mizushima et al., 1980).

> "His material provided everything I needed. It conducted well and brought the weight down by a third of the existing lithium–cadmium battery". (Yoshino, 2019)

Yoshino "further developed an aluminium foil collector to draw electricity from the cathode" (Yoshino, 2019), replaced water previously used as an electrolyte in rechargeable batteries with an organic solvent, and achieved over 4.0 V.

The example introduces the opportunity to adopt the modeling "bottom-up" approach to materials typical of nanochemistry (Ozin and Arsenault, 2005), and its advanced education (Pagliaro, 2015), to teaching and learning electrochemistry based on the chemical methodology through which chemists synthesize and create new substances based on mental visualization and association of atoms, atom groups and synthetic "building blocks" using various synthetic methodologies (Pagliaro, 2010, 2015; Ozin and Arsenault, 2005).

The relevance of nanoscience to electrochemistry education was recognized by Magnussen as early as of 2006. "Electrochemistry", he wrote "is not exclusively the domain of physical and analytical chemists anymore, but also becomes relevant to condensed matter physicists and material scientists" (Magnussen, 2006).

The main consequence of this shift is that electrochemical education should no longer be confined to introductory courses on physical or analytical chemistry but rather included "in programs on nanoscience and nanotechnology, at present emerging in most science departments" (Magnussen, 2006).

For example, two of us in Iran (M.G. and F.V.) learned the core concepts of electrochemistry during undergraduate courses on analytical chemistry. These courses covered the fundamental concepts of electrochemistry including electron transfer, electrode and electrolyte structure, thermodynamic of the cell, kinetics of electrode reactions, faradaic and non-faradaic current, as well as the introduction to the most common electroanalytical methods and techniques such as potentiometry, voltammetry, polarography, coulometry, and chronoamperometry.

The relevance of the modeling approach in understanding today's electrochemistry is unveiled by what kept electro-organic synthesis limited for over a century to the synthesis of a few bulk chemicals, namely the limited choice of electro-active materials, and the low current efficiency due to the use of bare (unmodified) electrodes as electron transferring agents (Wiebe et al., 2018).

Enabling the production of modified, highly active and durable electrodes, nanochemistry opens the route to the effective utilization of electrons in the oxidation and reduction of organic compounds, without the use of any environmentally undesirable oxidants or reductants, thereby generating no harmful effluents.

For example, in 2015 Karimi's team conceived an electrode suitable for the electro-oxidation of alcohols to carbonyl compounds comprised of a hybrid silica functionalized with stable nitroxyl radicals deposited as thin film over glassy carbon or graphite electrodes (Karimi et al., 2015).

Due to electro-assisted self-assembly (EASA) synthetic route employed (via the application of a cathodic potential to an electrode immersed in a sol solution containing the tetraethylorthosilicate silica precursor and cetyltrimethylammonium bromide surfactant as structure-directing agent), the periodic mesoporous channels are perpendicular to the electrode carbon substrate (Fig. 1).

The uniquely high accessibility of the aminoxyl radicals in the electrode functionalized with the silica thin film due to the highly organized (periodic) nanoarchitecture made of highly porous well-defined 2D hexagonal mesostructures perpendicular to the electrode surface originates its exceptional activity in the waste-free electro-oxidation of alcohols (turnover frequencies of up to 3070 h^{-1}, far superior to all the reported nitroxyl radicals under chemical or aerobic oxidation conditions) (Karimi et al., 2015).

Once more, this single example shows how, in practice, research chemists conceived and synthesized the "modified" electrodes with the desired nanoarchitecture via the simple and highly reproducible EASA material nanochemistry synthetic route.

Hence, by combining the uniquely high activity of the latter electrode with the catalytically highly stable (Palmisano et al., 2006) but structurally disordered (Palmisano et al., 2007) organically modified silica electrodes functionalized with aminoxyl radicals, a

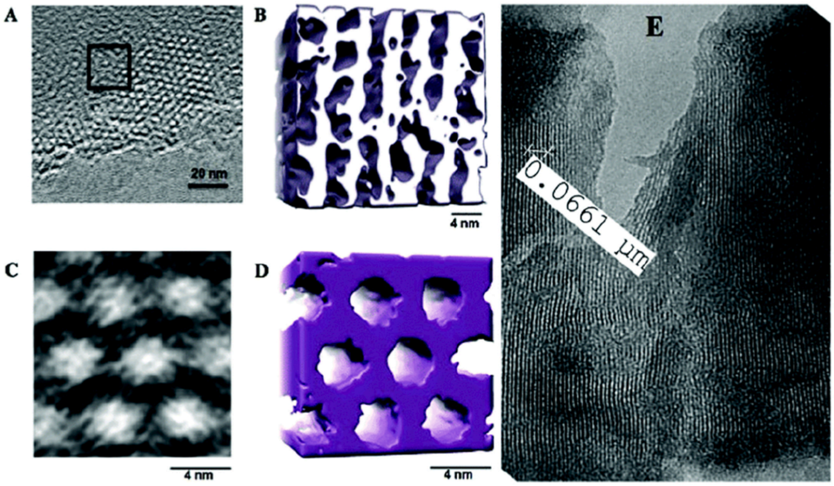

Fig. 1. TEM images (A,C and E) and reconstructed images (B,D) of TEMPO-functionalized ordered mesoporous silica electrode. (Reproduced from Karimi et al., 2015. With kind permission).

stable *and* highly active zero-emission oxidation electrode could be obtained (Karimi et al., 2018). Being stable and reproducibly obtained at low cost, the electrode is suitable for industrial uptake to replace the conventional catalytic oxidation of alcohols with NaOCl mediated by aminoxyl radicals with a waste-free process affording highly valued carbonyl or carboxylic compounds using an electric current only (Ciriminna et al., 2021).

The relevance of modeling to the effective teaching of electrochemistry concerns also the non-structural aspects of the discipline. One of us (M.G.) during undergraduate chemistry studies at the University of Tabriz was taught the basic electrochemical concepts by Professor Golabi. The scholar taught the concepts by systematically drawing schemes that greatly helped to explain complex electrochemical phenomena through imaging. Besides, in the lab classes, he tried to explain the application of electrochemical instruments.

Said “imaging” through “the drawing of schemes” is the visualization process assisted by drawing central to effective teaching and learning of scientific disciplines (Katz, 2017). The more abstract and intrinsically complex is said discipline, as it is the case of electrochemistry, the more such approach becomes essential to its effective teaching and learning.

As underlined by Osman and co-workers, however, in electrochemistry “the dimension of the examined objects (movement of particles) is too small and the parameters of the experiment are not directly available to the observer's senses in which the changes of the process are at the microscopic level” (Tien Tien and Osman, 2014). This fact suggests the development and use of animation to help students to visualize and learn electrochemistry, for example by showing the movement of ions and electrons during the electrolysis process to help students to visualize the oxidation and reduction processes occurring at both electrodes (Tien Tien and Osman, 2014).

Similarly, Arroio and Locatelli have shown how Brazilian students presented with a metavisual representation of the electrochemical phenomenon became able “to better visualize the interaction between the particles” reaching the scientific concept based on their ability to visualize and model the electrochemical phenomena (the number of correct answers before and after using the modeling approach nearly tripled from 32% to 88%) (Locatelli and Arroio, 2014).

Chemists and chemistry students are of course well acquainted to draw chemical structures (Peragovics and Biró, 2021), not only to visualize the invisible, namely atoms and molecules, but also to “predict the visible”, namely chemical reactions and chemical phenomena (Cooper et al., 2017; Niaz, 2002).

In this learning and predictive process, sketching is used as a tool to support model-based reasoning (Cooper et al., 2017), after which laboratory work becomes essential to support both reasoning and learning.

3. Focusing on green chemistry applications

Likewise to other chemistry fields in which recent research achievements can be purposefully used to improve chemistry teaching aiming to foster student creativity (Pagliaro, 2019), recent electrochemistry research is ideally suited to enhance learning of electrochemistry.

Familiar with the key concept of green chemistry, namely pollution prevention by proper design of the chemical synthesis (Linthorst, 2010), students become fascinated by the fact that industrially relevant reactions can now be carried out using electrons as reactants, generating no by-products and thus no pollution.

Electrocatalysis teaching (and learning) until a few years ago, suffered from the same problems of photocatalysis, namely the limited number of practical applications. Today, likewise to what happened for photocatalysis (Ciriminna et al., 2016), electrocatalysis is actually used by chemical companies to make highly valued fine chemicals (Perry et al., 2020).

Students not familiar with electrocatalysis and its manifold applications, will rapidly become fascinated by electrochemistry. They discover that in today's electrosynthesis the electrode does not merely act as an electron donor or acceptor, but as functional material deliberately designed to work as a selective mediator (catalyst) of a waste-free and highly selective chemical synthesis.

Learning efforts then will no longer focus on learning equations or reaction schemes (Locatelli and Arroio, 2014), but rather to creatively understand how research chemists using modeling actually developed a wide range of materials to be used as active, stable, inexpensive and industrially scalable electrocatalysts.

The same modeling-based learning approach can be applied to the development of the Li-ion battery cathodes. It is enough to study the evolution of their chemistry from the early $LiCoO_2$ oxide developed by Goodenough (Mizushima et al., 1980), through today's batteries in the $LiFePO_4$ cathodes are creatively arranged in large stacks based on cell-to-pack technology to optimize the structure and composition of the cell (Chen et al., 2022), to realize that chemical creativity here manifests itself in devising both new crystals structure that enable the flow of Li^+ ions within the cathode material as well as in devising completely new battery cell assembly.

The need to expand and improve the electrochemistry laboratory education is common to many countries. Recognizing the "lack of exposure and appropriate basic training of synthetic chemists and engineers in electrochemistry and electrochemical engineering" Russia's and Canada's scholars developed a single laboratory experiment, for undergraduate students to learn the basic electrochemical principles (Medvedev et al., 2022).

In detail, experimenting with the electrochemical reduction of CO_2 to CO using low-cost and readily available materials, students learn how to assemble a divided electrochemical cell, measure the rate of CO production under different electrolysis conditions, and understand the effects of operating parameters (applied potential, electrolyte concentration, and nature of the electrode) on the outcome of the reaction.

It is therefore relevant that today's low cost 3D printing technology, allows to replace expensive commercial electrodes such as the Ag/AgCl reference electrode, the platinum counter electrode, and carbon paste working electrode with low cost 3D printed electrodes rapidly produced at a fraction of the cost of commercial electrodes (Schmidt et al., 2022).

Similarly, experimenting with the rotating ring disk electrode technique, students learn how to assess the activity of the catalyst mediating the oxygen reduction reaction taking place at the cathode of H_2 fuel cells (Fruehwald et al., 2021), thereby acquiring "an extended set of electrochemistry skills that are highly demanded by modern employers" (Fruehwald et al., 2021).

Driven by the rapidly growing energy storage industry (the Li-ion battery industry and also the H_2 fuel cell and water electrolyser) (Pagliaro and Meneguzzo, 2019) and by the new applications of electro-organic syntheses in the chemical industry (Wiebe et al., 2018), research in electrochemistry is flourishing in both economically developed and developing countries.

Table 1 shows the top ten countries in electrochemistry research based on a scientometric assessment of research articles, reviews and letters published in indexed scientific journals between 2017 and 2021 (Scopus, 2022).

China alone contributed 17,708 studies out of 38,871 scientific articles, a share close to half of the total output (45.5%). The subsequent five countries are Canada (993 papers), Iran (974), Brazil (856), Italy (824) and Taiwan (767). Research is growing at fast pace. The number of scientific articles went from 6510 in 2017 to 8738 in 2021 (+34.2%). These articles are no longer mostly published in specialized journals such as *Electrochimica Acta* and *ChemElectroChem*.

The top 10 scientific journals publishing papers in electrochemistry indeed now include (Table 2) the three main general chemistry journals (*Chemical Communications*, *Angewandte Chemie International Edition*, *Journal of the American Chemical Society*).

Table 1
Top ten countries in electrochemistry research, 2017–2021 (Source: Scopus, 2022).

Ranking	Country	Number of papers
1	China	17,708
2	United States of America	6049
3	Germany	2487
4	India	2360
5	South Korea	1822
6	United Kingdom	1618
7	Japan	1524
8	Australia	1484
9	France	1309
10	Spain	1148

Table 2
Top 10 journals publishing in electrochemistry research, 2017–2021 (Source: Scopus, 2022).

Ranking	Journal	Number of papers
1	Electrochimica Acta	1050
2	ACS Applied Materials and Interfaces	901
3	Angewandte Chemie International Edition	817
4	Chemical Communications	774
5	ChemElectroChem	700
6	Journal of the American Chemical Society	674
7	Journal of Colloid and Interface Science	668
8	New Journal of Chemistry	661
9	Biosensors and Bioelectronics	632
10	ChemSusChem	628

In full agreement with the aforementioned advances concerning the application of electrochemistry to synthetic organic chemistry (Wiebe et al., 2018), the relevance of new electroorganic syntheses to green chemistry is also revealed by the presence in the ranking of a primary green chemistry journal such as *ChemSusChem*.

4. Conclusions

Whether employed in the development of economically viable electrocatalysts for synthesizing valued fine chemicals, or to make new cathodes for Li-ion batteries, the modeling approach of nanochemistry allows educators to shape new courses in which electrochemistry "is connected strongly to imaging and visualization" (Unwin, 2022).

Furthermore, the commonly observed need to reinforce electrochemistry laboratory education can be met by developing new experiments in green organic electrosynthesis through which students may for instance learn how new generation "modified" electrodes are ideally suited for employment in continuous-flow electro-reactors making the synthesis of valued chemicals efficient and waste-free.

Tailoring the molecular and nanostructure of the electrode surface, in brief, allows to carry out highly selective electrooorganic reactions under continuous flow that are both technically and economically viable due to the enhanced mass transfer, large catalyst surface:volume ratio, and better control of the reaction temperature due to the more efficient heat exchange in flow reactors (Tanbouza et al., 2020; Wills et al., 2021).

From continuous flow process for the electrochemical oxidation of primary alcohols bearing nitrogen-containing heterocycles in a continuous single-pass flow reactor (Jud et al., 2022) through the synthesis of 2-phenyl-1*H*-benzimidazole through bulk electrolysis of *o*-phenylene diamine and benzaldehyde over anchored ferrocene (Thadathil et al., 2022), examples of commercially scalable processes are currently reported on a monthly basis.

Remarkably, nearly all said works include a sustainability assessment of the new electrochemical process to evaluate the applicability of scaling the methodology. For example, the ultralow leaching (22.3 ppb) of nickel in solution from the electrooxidation of alcohols at the surface of the NiOOH modified electrode and the stable catalytic activity, led Kappe and co-workers to conclude that the process is viable for continuous manufacturing of carboxylic acids at lower capital and operational cost (Jud et al., 2022).

Similarly, Gosh and co-workers used the EcoScale (Van Aken et al., 2006) tool to evaluate the environmental sustainability and economic sustainability of the synthesis of 2-phenyl-1*H*-benzimidazole via the conventional homogeneous synthesis in batch (66/100) and the heterogeneously catalyzed electrochemical flow process (72/100) (Thadathil et al., 2022). Besides identical high atom economy of 90.7%, in nearly every aspect (greener solvent, greener purification via crystallization, lack of catalyst loss) the new method was significantly better than the conventional synthesis. The team concluded that the new electrocatalytic process under flow opens the route to the green and low cost commercial synthesis of phenyl benzimidazoles (Thadathil et al., 2022).

In conclusion, three main guidelines originating from the present analysis can be provided to chemistry educators wishing to enhance electrochemistry education.

First, aware that the modeling and visualization approach significantly improves electrochemistry teaching and learning (Locatelli and Arroio, 2014; Ozin and Arsenault, 2005; Pagliaro, 2015; Nakiboğlu and Nakiboğlu, 2021), educators need to improve their own competence for visualizing and presenting electrochemical phenomena. In practice, such teaching skill requires educators to develop suitable graphic organizers by which "to enable students to construct the electrochemistry concepts and subjects meaningfully in their minds" (Nakiboğlu and Nakiboğlu, 2021).

Second, introducing new experiments in green organic electrosynthesis in electrochemistry laboratory education is particularly well suited to enhance student interest in learning concepts and ideas that otherwise remain often abstract and obscure.

Third, aware that new tools from today's inexpensive electronics and open-source software (Kempler et al., 2021), through magnetic resonance imaging for probing electrochemical processes in a model electrochemical cell are available (Britton et al., 2013), educators will review and select some of said new tools to expand and enrich their instructional material both for the classroom and for the laboratory education.

Meaningful learning of students of similarly renewed courses in electrochemistry enabling profound comprehension of the electrochemistry concepts and tools will also foster student motivation and creativity (Torres et al., 2014; Nakiboğlu and Nakiboğlu, 2021). Creativity of (electro)chemists, after all, is what mankind needs to further advance the electrochemical electricity storage technologies to store and release energy sourced from renewable energy sources to end global reliance on today's economically unsustainable petroleum (Meneguzzo et al., 2016).

Author statement

Rosaria Ciriminna: Methodology, Analysis, Validation, and Writing original draft. Mina Ghahremani: Analysis and Writing-Reviewing and Editing. Fahimeh Varmaghan: Analysis and Writing-Reviewing and Editing. Babak Karimi: Conceptualization, Supervision, Validation, Writing-Reviewing and Editing. Mario Pagliaro: Conceptualization, Supervision, Validation, Writing-Reviewing and Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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