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Relevant developments and new insights on Sonoelectrochemistry

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Abstract

Sonoelectrochemistry is undergoing a reemerging activity in the last years with an increasing number of papers appearing in a wide range of peer review journals. Applied studies cover environmental treatments, synthesis and characterization of nanostructures, polymeric materials synthesis, analytical procedures, films preparations, membrane preparations among other interesting applications. Fundamental analyses are also carried out focused on electrochemical processes using unconventional solvents, elucidation of mechanisms and combination with other techniques. The interrelation between Electrochemistry and Acoustics presents mutual benefits for both disciplines, providing interesting information about the bubble dynamics for acoustics physicists and a higher number of possible applications for electrochemists. However, the vast majority of this research has been carried out at laboratory scale with individually designed systems based on ultrasonic horns dipped into traditional glass electrochemistry vessels. It is remarkable that even with this rudimentary experimental set-up many interesting results have been generated. However sonoelectrochemistry has suffered a few drawbacks related to reproducibility, scale-up and design aspects which have slowed its development. Almost certainly the reason for this is the lack of reactors that have been purpose built for sonoelectrochemistry. There have been many attempts to build lab-scale systems e.g. for electroanalysis, nanomaterials synthesis and the electrooxidation of organic pollutants but the results are often contradictory. A few groups have attempted to characterize lab-scale sonochemical reactors adapted as sonoelectrochemical reactors but the true optimization of such reactors requires contributions from many disciplines including physics, fundamental and applied electrochemistry, chemical engineering and material science.

Keywords: cavitation; current distribution; electrocatalysis; electrochemistry; fluid flow; ultrasound.

1. Introduction

It is surprising that there is little information about Sonoelectrochemistry until 1980s. In the 1930s, Morigushi [1] analyzed the effect of ultrasound in the electrolysis of water. In the 1950s, there was considerable interest in

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sonically assisted electroplating [2], especially of nickel and chromium [3]. Analytical applications are found in the 1960s [4]. In the 1980s, sonoelectrochemistry is focused on studies of polymerisation of styrenes [5] and thiophene [6] and also organic sonoelectrosynthesis [7]. However, from 1990s the sonoelectrochemistry started an active development not only on fundamental but also on applied studies. Nowadays, we can find several reviews in general sonoelectrochemistry [8-13] and also in its specific applications such as organic synthesis [14-15], and electroanalysis [16].

2. Fundamental studies

The coupling of an electrical field with an ultrasound field has focused special attention from a fundamental point of view. In 2003, Compton et al [17] reviewed the main results in this field. We can find in literature not only work done under ultrasound on typical electrochemical processes with simple molecules [18] and macromolecules [19], but also analysis about the mechanical effects of the ultrasonic field propagation on the electrode surface such as acoustic streaming and microjetting [20], electrode cleaning [21] and general aspects [22-23] related to the mass transport [24-25]. Cavitation and shock waves have also received attention [26-27].

The influence of the medium in the propagation of the ultrasound field has also been studied. Highly resistive media (mass transport effects) [28], organic solvents [29] and electrochemistry in acoustically emulsified media [30] were also analyzed. This last subject has received a specific review [31]. In spite of the major part of this work has been done at low frequencies (20 kHz), the influence of the high frequencies on the sonoelectrochemical processes in aspects such as mass transport or surface effects have been analyzed [32]. Other specific aspects of the effect of the coupling of the electrical and ultrasonic fields have been analyzed, for example, the sonochemical enhancement of electrochemiluminescence [33], the low-temperature sonoelectrochemical processes [34], high pressure sonoelectrochemistry [35] and also physicochemical aspects with the use of high performance techniques so-called high speed voltammetry [36], adsorption of macromolecules [37] and the electrochemistry at the emitter surfaces [38].

Acoustics also profits from sonoelectrochemical experiments, analyzing the cavitating bubbles [39], [40] and electrochemical characterization of cavitation [41].

3. Applied studies

3.1. Sonoelectroanalytical applications

The analytical application has been one of the most active fields in sonoelectrochemistry. The first work found in literature was presented by Allen Bard in 1963 [4], however the Compton group [42-43] have reported a vast work in several applications of sonoelectrochemistry, developing approaches to the coupling of ultrasound with electroanalytical techniques. Metals [44] such as silver [45], nickel [46], copper [47], lead [48], inorganic ions [49] and organic compounds, such as 5-aminosalicylic acid [50], nucleosides and nucleotides [51] have been successfully analyzed with different sonoelectrochemical methods.

3.2. Sonoelectrosynthetic applications

Sonoelectrochemistry has also been used in synthetic routes, especially in organic synthesis. Compton et al [52] have presented an overview of sonoelectrosynthetic work carried out by different research groups between 1985 and 1999, with 20 references. Organic electrochemical reduction has been revised by Durant et al [53]. After that, synthesis has kept going active not only in aqueous media [54] but also in aprotic solvents [55] or liquid ammonia [56] and even with liquid electrodes [57]. Inorganic synthesis has been also developed with sonoelectrochemistry. CuBr has been obtained by sonoelectrochemistry [58], mediated synthesis [59] and also tetramethyladipic acid by a sonoelectrochemical Fenton process [60].

3.3. Coating and surface applications

The influence of ultrasound in electroplating was one of the first topics studied in sonoelectrochemistry. The benefits have been well-established and continuously used. Not only typical metallic deposits are obtained such as silver oxide [61], silver [62], tungsten [63] but also bioactive calcium phosphate coating [64] and CdSe films [65] have been sonoelectrodeposited. Some of these studies have been focused on the scale-up of the process [66]. In addition, other surfaces applications have also been developed by sonoelectrochemical techniques: anodization [67], etching [68], sonoelectropolymerization [69], sonoelectrodeposition [70] and composites electrodes [71].

3.4. Nanomaterials preparation

One of the first works published in this field comes from the Reisse group [72]. Nowadays, nanomaterials preparation is one of the most active fields of research in sonoelectrochemistry. Nanoparticles of magnesium [73], of tungsten [74], of copper [75], of palladium [76], of silver [77], of gold [78], of PbS [79], of copper oxide (I) [80], of Au-Ag alloys [81], of Co-Fe alloys [82] have been synthesized by sonoelectrochemistry. In the same way, nanotubes of CdSe [83], of TiO_2 [84] have also been manufactured and these nanomaterials have been used in water splitting processes [85]. Silver nanowires have also been synthesized by sonoelectrochemical methods [86].

3.5 Environmental applications

The application of sonoelectrochemistry in the protection of the environment is another interesting field of research. The compounds studied are typically Dyes compounds such Procion Blue [87], hydrophilic chloroorganic pollutants [88] and also metals [62].

4. Ultrasonic equipment and sono-electrochemical reactor design

One of the first attempts to analyze the influence of the implementation way to introduce the ultrasonic field in the electrochemical system was reported by E. Namgoong et al [89], analyzing the alternative approach to apply ultrasound directly to the cathode, rather than the electrolyte solution itself, during electroplating experiments. In this way, the idea of a specific design of a sonoelectrochemical reactor has been previously reported in the literature by Reisse el at [90] and different approaches have been reported [91-93]. The necessity for the bipotentiostatic control in some experiments [94] has been pointed out.

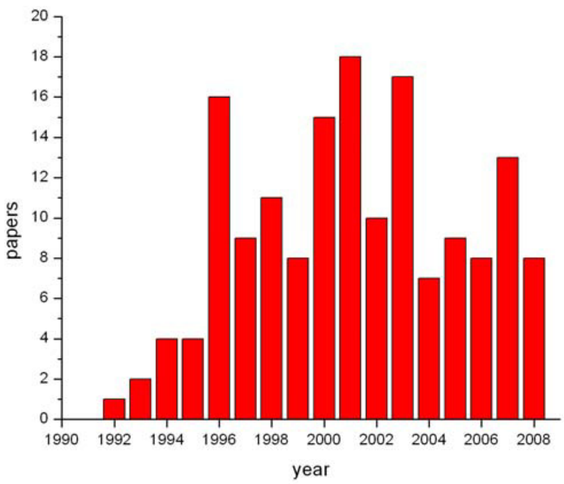

Fig.1 Temporal evolution of papers in sonoelectrochemistry (Source: Scifinder Scholar. Keyword: Sonoelectrochemistry).

The characterization of the cavitating cells is being subject of study [95] by electrochemical [96-97], visual [98] and numerical methods [99] in order to design an optimized sonoelectrochemical reactor. Finally, new attempts to develop hybrid techniques have been recently reported in the literature, such as the combination of EQCM (Electrochemical quartz crystal microbalance) plus ultrasound [100] or special processes at low temperatures [101].

Fig. 1 and 2 present the recent evolution of the sonoelectrochemical activity. As it can be seen, two main subjects have received special attention, in the 1990s the physicochemical aspects and now in the 2000s the nanostructures synthesis and characterization. The sonoelectrohemistry is consolidating as an active field of research.

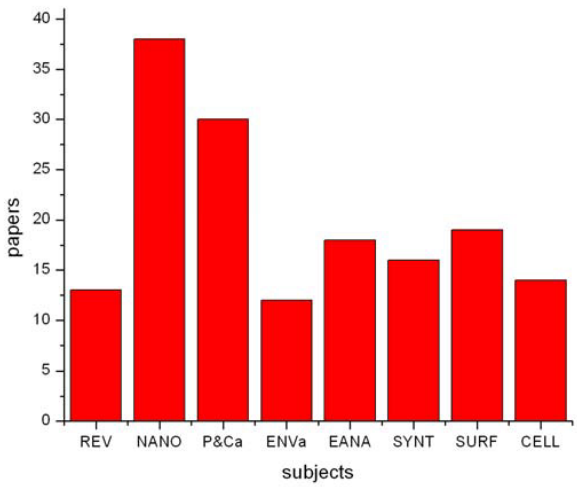

Fig.2 Number of papers published in sonoelectrochemistry by subject (Source: Scifinder. Keyword: Sonoelectrochemistry). REV: reviews and general aspects; NANO: nanoestructures, P&Ca: physicochemical aspects, ENVa: environmental applications, EANA: application in electroanalysis; SYNT: application in electrosynthesis, SURF: coating and films preparation, CELL: sonoelectrochemical reactor design.

5. Conclusions

From all the references analyzed, the following items should address attention:

a) To solve the problem of coupling between the electrical field for electrochemical reactions and the excitation field for an ultrasonic transducer in a sonoelectrochemical reactor.

b) To design and build a flexible sonoelectrochemical multi-purpose reactor.

c) To study the frequency dependence of sonoelectrochemical phenomena.

d) To study the intensity of acoustic field dependence in some sonoelectrochemical reactions in controlled way.

e) To measure in high accuracy way the acoustic field inside the sonoelectrochemical reactor.

f) To establish the best electrical field to improve sonoelectrochemical reactions.

g) To assess the influence of an electrical field in the tip of the sonoelectrochemical reactor.

h) To study the dependence from transient cavitation threshold and the gas solved in the cavitating liquid.

i) To assess the participation of cavitation in the generation of new compounds in sonoelectrochemical reactions.

j) Advance in the knowledge of electrode material behaviour in the sonoelectrochemical reactions.

k) To explore new and exciting applications of sonoelectrochemical reactions in the nanomaterials production.

l) To produce the engineering knowledge for the design of sonoelectrochemical reactors.

m) To advance in the design of equipment to continuous process.

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