Sonoelectrochemistry: Ultrasound-assisted Organic Electrosynthesis

Higor de Bitencourt Rodrigues[a], Jéssica Oliveira de Brito Lira[a], Natan Padoin[a], Cíntia Soares[a], Ahsanulhaq Qurashi[b], and Nisar Ahmed*[c,d]

[a]Laboratory of Materials and Scientific Computing (LabMAC), Department of Chemical and Food Engineering, Federal University of Santa Catarina, Campus Universitário Reitor João David Ferreira Lima, 88040-900 Florianópolis/SC, Brazil
[b]Department of Chemistry, Khalifa University, Main Campus, Abu Dhabi, 127788, United Arab Emirates
[c]International Centre for Chemical and Biological Sciences, HEJ Research Institute of Chemistry, University of Karachi, Karachi 75270, Pakistan
[d]School of Chemistry, Cardiff University, Main Building Park Place, Cardiff, CF10 3AT, United Kingdom.

***E-mail**: AhmedN14@cardiff.ac.uk

Abstract: The application of ultrasound with electrochemistry in organic chemistry (known as organic sonoelectrochemistry) accelerates the activation process of chemical reactions. This hybrid technology enhances electrical efficiency, modifies and increases the product yield. Moreover, facilitates the mass transfer phenomena, and the processes of cleaning, degassing, and activation of the electrode surfaces, maintains higher current densities for efficient chemical transformations and also works efficiently for mixing of reactants in multi-phases systems. The ultrasound technology has prominent effect in heterogeneous reaction systems especially during solid (electrode)-liquid (electrolytic mixture) interfacial cavitation process. The ultrasound technology gains attention due to its fundamental and positive effect in organic chemistry to makes possible the challenging electrosynthetic processes. Herein, we report the sonoelectrosynthetic methods that will help the researchers to understand and apply this methodology for scale-up of processes in organic synthesis and also in more modern innovative continuous-flow organic electrochemistry. Therefore, this study will provide valuable insight into the effects

caused by ultrasound-assisted electrosynthesis and how this technology revolutionizes organic synthesis. It is believed that the hybrid sonoelectrochemical synthesis serves as a solution to the limitations of the commercialization of synthetic processes and offers a new modern aspect in organic synthesis in a clean, hassle-free, and sustainable approach.

Keywords: Organic sonoelectrosynthesis; Ultrasound; Heterogeneous catalysis; Sustainable chemistry; Synthetic sonoelectrochemistry & organic reaction engineering

INTRODUCTION

Sonoelectrochemistry is a research field that investigates electrochemical processes assisted by ultrasonic radiation [1,2]. This technology has its first application reported in the 1930s with Moriguchi's research [3], which showed an improvement in water hydrolysis by insonation (vibration of electrochemical cells) [4]. Since then, sonoelectrochemistry has become an interesting field due to the advantages that this technology provides in electrochemical systems, such as the generation, growth, collapse of microbubbles within the electrolyte and near the electrode surface [2,5,6]. Thus, the use of ultrasound is classified as one of the process intensification (PI) methods.

PI offers innovative challenges and opportunities for the chemical industries, and it can be seen as a series of tools that seeks to improve process performance in terms of energy efficiency, lower product and system cost, and better sustainability [7,8]. Therefore, the use of ultrasound as an energy source for chemical mechanisms has proven to be a milestone as a process intensification due to the formation of microbubbles (cavities) in liquid reaction media. The synergistic effect of sonoelectrochemical processes has brought new possibilities for active research due to its potential exploration for use wide range of applications. Thus, many studies are found in the literature referring sonoelectrochemistry to environmental remediation as a hybrid technology for the organic compounds degradation present in the aqueous medium [9–11]. Other applications are being used for clean hydrogen production [12], synthesis of nanomaterials [13–17], film production [18] and surface coating [19], lab-on-a-chip analysis [20], and detection of lead [21–23]. Moreover, the application of associating sonoelectrochemistry with organic synthesis has also been highlighted in the literature, mainly because this technology brings interesting aspects because the sonoelectrosynthetic reactions usually present substantial insonation effects, even in less defined acoustic conditions [4,24]. Therefore, among the various relevant

applications to sonoelectrochemistry (showed above), organic sonoelectrosynthesis will be the objective application of the present paper [25].

At the end of the 1980s, Kolbe's electrooxidation of cyclohexane carboxylate in methanol became known as one of the oldest electro-organic synthetic reactions. In the ultrasound-assisted system, there was a conversion of one-electron per molecule of dimer bicyclohexyl into two-electrons per molecule products [26,27]. Since then, sonoelectrochemistry has become a field that has attracted a lot of interest and has been growing due to the series of reports on the topic [28]. Durant et al. [29] applied an ultrasound in the organic electrochemical reduction reaction of benzaldehyde and benzoquinone and evaluated the effects provided to the process. González-García et al. [30] studied power ultrasound-assisted hydrogen peroxide electrosynthesis via oxygen reduction. In another work, González-García et al. [31] reported the hydrogen peroxide production in useful synthetic amounts in a sonoelectrochemical cell through the chemical oxidation of benzonitrile to benzamide. Moreover, Atobe [32] studied the acoustic emulsification in electrochemical reactions (oxidation and reduction) of water-insoluble molecules in aqueous electrolytes. The electrosynthesis reduction could also be performed in aprotic solvents [33] and liquid ammonia [34] in the presence of ultrasound. Other applications of organic sonoelectrosynthesis have been detailed by Marken and Atobe [35].

Moreover, significant benefits are evidenced by the synergy of ultrasound in electrochemical processes [36]. The promotion of chemical reactivity at the solid-liquid interface is the main advantage of organic synthesis by sonoelectrochemistry, this reactivity is done through self-cleaning and degassing, by activating the electrode surface. In this way, the reaction can have an increased yield and the reaction mechanism (i.e., reaction path) changed, along with the change in product disposition [6,37,38]. Improving the electrical efficiency, accelerating the process of activating/driving chemical reactions,

modifying and increasing the yield of the products are also other advantages of ultrasound-assisted organic electrosynthesis [39]. Besides, this technology also has benefits associated with mechanical phenomena, such as cavitation and shock waves [41] and acoustic streaming and microjetting [40]. These phenomena can decrease electrode diffusion layer thickness, improving the overall mass transfer and, consequently, increasing the reaction rates [5,6,36,42].

Therefore, the use of ultrasound for synthetic electrochemical processes stands out as a sustainable strategy that promises to raise the discipline of sonoelectrochemistry, giving innovative dimensions to organic synthesis due to the countless benefits and growing development of green organic synthesis [43,44]. Therefore, the paper aims to provide a brief overview and future prospective of the advances in sonoelectrochemistry for organic synthesis technology. In addition, we hope that this review will open up new possibilities to increase interest in this field, providing additional investigation and boosting this technology to become a viable organic synthetic technique.

SONOELECTROCHEMISTRY

Sonoelectrochemistry is a research field that studies the cavitation process produced by ultrasound in electrochemical systems [41]. This field of work has received attention in recent decades, but its application originated in the mid-1934s [3] when Moriguchi first introduced the use of ultrasound in water electrolysis, where it was tried to reduce the decomposition voltage of water on a platinum working electrode. Following the introduction of sonoelectrochemistry by Moriguchi, in 1960, Nyborg [45] used electrochemical techniques to study acoustic streaming processes using acoustically oscillated electrodes and electrode arrays. Since then, many studies have focused on the use of ultrasonic irradiation in an electrochemical system.

There are two different sonoelectrochemical configurations based on ultrasound irradiation: ultrasonic bath and immersion of an ultrasonic probe horn (Fig. 1) [9,46]. The ultrasound bath is the simplest configuration and is normally used because of its ability to clean the surfaces of the working electrode [46]. This acoustic wave propagation device differs from the probe-type ultrasound in terms of efficiency and process capability [47]. However, the biggest disadvantage of this configuration is the uncontrolled propagation of the ultrasound waves in the liquid medium with low intensity and non-homogeneous spreading [48]. On the other hand, the ultrasonic probe is the most widely used configuration because the ultrasonic horn can be immersed directly into the electrolyte solution [9,46,49]. This arrangement provides intense effects and more efficient energy control compared to the ultrasonic bath [50]. Furthermore, this configuration allows a homogeneous distribution and a more accessible adjustment of the distance between the horn and the working electrode and the ultrasonic intensity [47,51]. However, this arrangement also has some limitations, such as erosion of the horn and the need for temperature control due to the heating effect of the horn [46,51].

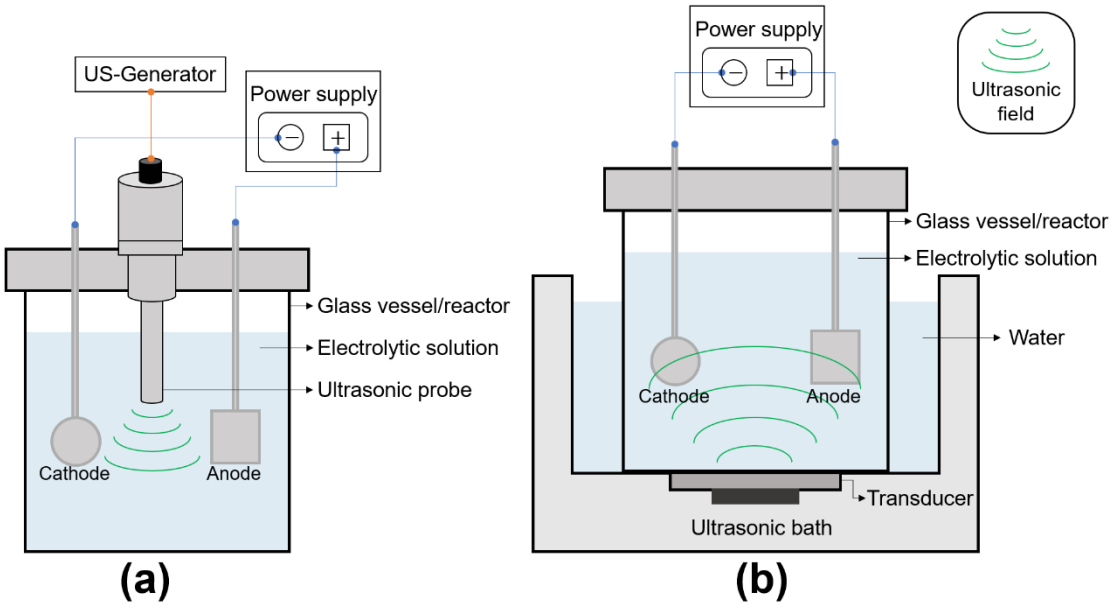


Figure 1: Illustrative diagrams of equipment for sonoelectrochemical processes: (a) ultrasonic bath and (b) immersion of an ultrasonic probe horn. Adapted from ref 49. Copyright 2021 (open access journal), MDPI.

Furthermore, the effects of ultrasonic irradiation in heterogeneous systems, involving the electrode and the electrolyte, and in homogeneous systems, taking place in the bulk solution, have been studied and discussed in the last 30 years [2,12]. Hydrodynamically, the two main phenomena that can be observed when using ultrasonic irradiation are acoustic cavitation and acoustic streaming [52,53] (see Fig. 2). Those physical mechanisms can be induced according to low- and high-frequency ultrasounds. The low frequency of ultrasound generates micro-bubbles cavitation, i.e., it is the process that presents the stages of generation, growth, and collapse of the bubble, induced by the variation of the wave pressure. At the stage where bubbles collapse, local hydrodynamic effects are generated intensely, such as micro-jets, which have very high velocities and shock waves, and therefore, these effects can cause other surrounding bubbles to collapse [54,55]. The most recent interest in sonoelectrochemical processes is due to the variety of these effects linked to the creation, expansion, and collapse of bubbles in the electrolyte or near to the surface of the working-electrode [26]. Cavitation is known as a phenomenon present in the chemical and physical PIs. In cases where high-frequency ultrasound is used, cavitation effects are not normally seen because this type of ultrasound operates at a power below the cavitation limit [56]. However, the acoustic streaming mechanism is evidenced for the application of high-frequency ultrasound. This phenomenon induces recirculating flows, and it is able to enhance mixing. This acoustic flow is the effect of turbulent convection and ultrasound-generated bubble collapse [54,55].

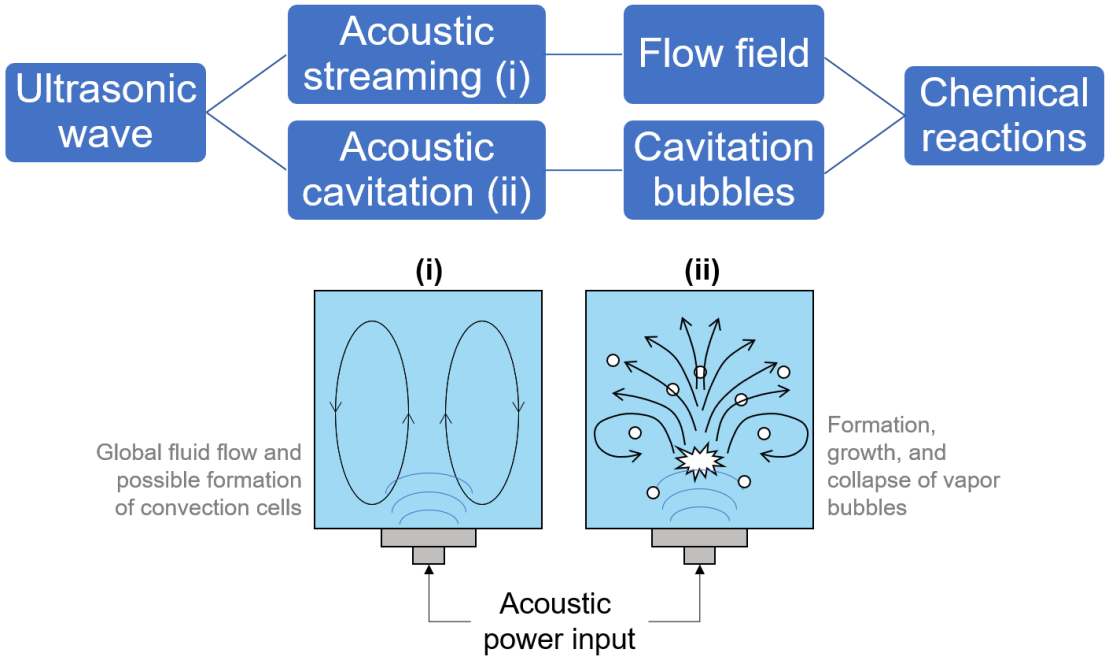


Figure 2: Representation of the main physical mechanisms associated with high- and low-frequency ultrasound: (i) acoustic streaming and (ii) cavitation effect. Adapted from ref 56. Copyright 2020 (open access journal), MDPI.

The two acoustic phenomena (cavitation and streaming) cause an increase in heat transfer in liquids. Thus, heat transfer can be improved with the acoustic field appearance despite the heat transfer mode [57]. If cavitation occurs close to the electrode surface, there will be the formation of high-speed micro-jets of liquid that collide toward the surface [6, 26]. Furthermore, other phenomena are also associated with the collapse of the bubble, such as shock waves [41] and microextraction [40], when the ultrasound intensity is greater than the threshold intensity. All of these effects bring benefits to the sonoelectrochemical process, and they can decrease the thickness of the electrode's diffusion layer [58] and enhance the overall mass transport. In addition, these effects can also increase reaction rates and can also improve electrode surface cleaning and degassing [28]. Also, chemical effects associated with radical generation from solvent sonolysis can also be observed

[59,60]. Acoustic cavitation and acoustic streaming are phenomena qualified as they use low-frequency ultrasound, defined between 20 and near 100 kHz. Indeed, high-power levels (a few tens of Watts) of ultrasound are generally used due to its ability to propagate and generate powerful macroscopic effects capable of improving the heat and the mass transfer rates [61]. However, a thorough and methodical approach must be employed to the ultrasound as irradiation treatments using either an ultrasonic bath or an ultrasonic probe can have adverse effects depending on the frequency, power, and time of ultrasonic irradiation. Pollet [62,63] found that ultrasonic treatment was useful for dispersing the catalyst in fuel cells, improving the catalytic activity (operating at 1.82 W and 3.03 W). However, a more aggressive treatment, e.g., 12.23 W (ultrasound probe), resulted in a detrimental effect causing agglomeration and dissolution of the catalyst particles.

Some mechanical phenomena such as microjetting and acoustic streaming, cavitation, and shock waves are related to the propagation of the ultrasonic field on the surface of the working electrode. Furthermore, general aspects such as the mass transport enhancement and the electrode surface cleaning can also be associated with the propagation of the ultrasonic field. All these aspects at low and high frequencies have been reviewed in 2003 by Compton et al. [5,42]. More recently, several advances in the field of sonoelectrochemistry have allowed this technology to be increasingly explored for a wide array of applications. These applications of sonoelectrochemistry processes were reported by Pollet [25], some of the important applications of the combined power ultrasound and electrochemical technology are: sonoelectrodeposition, sonoelectroanalysis, environmental remediation, and sonoelectrosynthesis [11].

SONOELECTROCHEMICAL APPLICATIONS

The application of sonoelectrochemistry was studied with the aim of generating highly reactive species [11]. Thus, significant improvements have been achieved due to the synergistic combination of ultrasound and electrochemical systems. The applications of ultrasound in electrochemical systems as described in detail by Pollet [25] include sonoelectrodeposition, sonoelectroanalysis, environmental remediation, and sonoelectrosynthesis (see Fig. 3). The last application will be discussed further as case studies for organic synthesis.

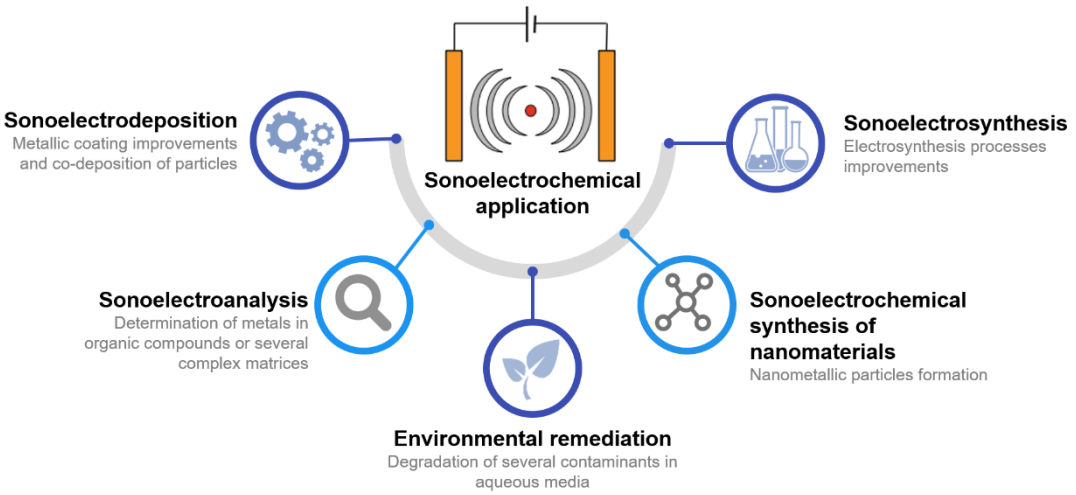


Figure 3: Sonoelectrochemical applications.

Sonoelectrodeposition

The combination of ultrasound-assisted electrodeposition processes is well established, and it has been used extensively in the literature due to its numerous benefits [47,64,65]. Better deposition rates and efficiency and improvement of deposit quality, increasing the thickness, hardness, and brightness of the deposited films, in addition to ensuring a greater adhesion on the electrode, are the most frequent benefits mentioned for the application of ultrasound in metal electrodeposition [25]. Sonochemical assisted approach was employed to develop secondary and tertiary layers of hematite and silver on nanotube surface to make highly effective visible photoanode [66]. Furthermore, several works were also

published to apply ultrasound in electrochemical processes for deposits and metallic coatings, nanocomposites, films, and nanosheets [67–73].

Sonoelectroanalysis

Another sonoelectrochemistry research field that is currently emerging is focused on analytical applications as a powerful electroanalytical tool [74,75]. Several studies have reported the use of ultrasound for electroanalytical determination of metals [76,77] such as silver, nickel, copper, lead, inorganic ions in organic compounds using stripping techniques on different types of electrodes and in several complex matrices [25].

Environmental remediation

The application of sonoelectrochemistry in environmental remediation is an emerging field of research that has received increasing interest recently, as it allows the degradation of several contaminants in aqueous media [9,11,42,78]. The compounds are typically dyes [79–82], pharmaceutical effluents [83–86], and metals [87]. Furthermore, the application of ultrasound can improve advanced oxidative processes (AOPs) due to the oxidative species generation [10]. Therefore, this technology can be combined with other advanced oxidation processes, including biological treatment [88], ozonation process [89], and Fenton process [90], to achieve synergistic effects in reducing organic and energy-efficient matter.

Sonoelectrochemical synthesis of nanomaterials

One of the most active areas in sonoelectrochemistry technology involves the production of metallic nanomaterials in a simple and controlled way [91–93]. Reisse et al. [94] introduced sonoelectrochemistry for the metallic nanoparticles synthesis, and the copper was synthesized using this technique. After that, significant efforts were made and, currently,

different metallic nanoparticles were produced by sonoelectrochemical methods [25,36]. Islam et al. [36] presented the most recent advances for the synthesis of nanomaterials via sonoelectrochemical processes. For example, it can be mentioned: nanoparticles of copper, gold, silver, magnesium, palladium, among others.

Sonoelectrosynthesis

The development of new synthetic routes, focusing on green chemistry, is also another application for sonoelectrochemistry, mainly in organic synthesis. Although this area is not as active as the previous ones, it presents a wide range of possibilities. The benefits of using ultrasound for organic electrosynthesis include activation of the electrode surface, increased yield, and the possibility of alteration in the reaction path, along with the altered product distribution [4,38]. Compton et al. [76] reviewed the works available in the literature on sonoelectrosynthesis between 1985 and 1995. González-García et al. [42] summarized some works that involve the application in sonoelectrosynthesis. Also, more recently, some insightful book chapters have been published on organic sonoelectrosynthesis [25,95]. Thus, this topic will be more detailed in the next section, where works on organic sonoelectrosynthesis will be approached with more emphasis.

CASE STUDIES OF ORGANIC SONOELECTROSYNTHESIS

As seen earlier, there are several applications of ultrasound-assisted electrochemistry. However, in recent years, the old field of electrochemistry is receiving considerable attention in organic synthesis with a focus on the production of fine chemicals and pharmaceuticals. The anodic oxidation and cathodic reduction reactions require the electron transfer reaction (SET), which, according to the rules of true sonochemistry, can be enhanced by ultrasonic irradiation [95]. It can be said that the results of the use of

ultrasonic fields in electrosynthesis reactions, in most reports, are found in the substantial modification of the products, high yield and selectivity rates, or the process parameters [4]. However, the effects caused by the ultrasonic irradiation presence must be attributed to the large increase in hydrodynamics and improvement in the mass transfer between the bulk of the electrolyte solution and the working electrode [96].

The group by Nonaka and Atobe et al. showed consistency in research and development in the field of sonoelectrosynthesis. A series of articles focusing on the ultrasonic effect in electro-organic processes reached part 27 in 2006 [97]. For example (see **Fig. 4**), Atobe et al. studied sonication effect at the suspended electrode of lead particles during the electro-reduction of acrylonitrile. The production of adiponitrile **1** was observed more selective as compared to propionitrile **2** in the presence of lead particles, and, under ultrasound, an increased yield with high selectivity was achieved. They attributed that the ultrasonication process increased the effective surface area of lead particles to help mass transfer to the feeder cathode.

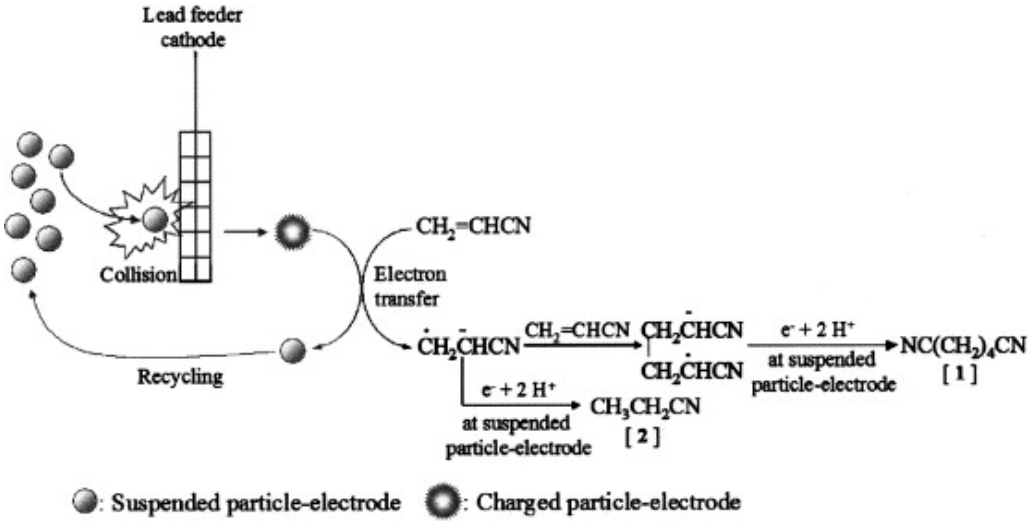


Figure 4: Illustrative schematic of the electroreduction reaction of acrylonitrile using a suspended lead particle-working-electrode. Reproduced with permission from ref 97, copyright 2006, Elsevier

In the first studies, in the mid-1990s, they first examined a series of electroreduction of carbonyl compounds. Ultrasound induced a marked change in product distribution that offered different products for one- or two-electron mechanisms per molecule [98–100].

Following the same line of electroreduction, González-García et al. [30,31] studied the ultrasound effects on the oxygen reduction reaction for hydrogen peroxide electrosynthesis, and the reaction was also evaluated under silent conditions. For the study, the authors used as a cathode, an electrochemical flow reactor with reticulated vitreous carbon (RVC). Firstly [31], the authors concluded that mass transport enhanced with the use of ultrasound increased the efficiency of H_2O_2 formation. Later, in another study by the same research group [30], it was found that both the cumulative hydrogen peroxide concentration and the process current efficiency were increased due to the application of high-power ultrasound. In other words, the ultrasound-assisted process achieved an efficiency of 60%, while under silent conditions, the efficiencies were 20% or less. The authors also justified this improvement due to a boost in mass transport by ultrasound.

Similarly, Walton et al. [101] used a frequency of 850 kHz through ultrasound (high-frequency) during cyclohexanoate electrooxidation. Using this frequency, the authors observed that ultrasound had positive effects, such as the development of a cleaner reaction and higher product yields. The reaction yield using the high frequency of 850 kHz was double and one and a half times greater when compared to the silent system and low-frequency insonation (38 kHz), respectively (see Table 1).

Current density/ $mA \cdot cm^{-2}$	%Partially neutralised	Silent	38 kHz	850 kHz
150	25	0.8	1.2	1.6
200	50	0.7	1.0	2.1

Table 1: Total quantities (g) of ether-soluble materials from the cyclohexanoate electrooxidation reaction for silent system, 38 kHz, and 850 kHz insonation. Reproduced with permission from ref 101, Copyright 2000, Elsevier.

Valcarel et al. [102] analyzed the ultrasound effects on the thiophene *S*-oxides electrooxidation. Sonovoltamograms with 40 and 850 kHz bath insonation showed the expected current increases for tetraphenylthiophene *S*-oxide and tetracyclone. Thus, the authors inferred that ultrasound promoted a beneficial effect on the balance of products in synthetic electro-organic reactions, such as electroreduction of carbonyl compounds and Kolbe electrooxidation of carboxylates. However, interestingly, the ultrasound did not attenuate the fouling phenomena of the electrode, which had to be manually cleaned several times during the electrolysis. The justification for this phenomenon was the formation of a polymeric species in the electrode.

In another synthetic electrooxidation study, Lindermeir et al. [103] evaluated the effects of ultrasound application on the electrooxidation reaction of substituted toluenes. Higher product yield and current efficiency were found for the ultrasound-assisted reaction. This result is shown in **Fig. 5** (i), which shows the relationship between different current densities with the maximum product yield for the ultrasound configurations. Then, the yield of the products highly decreased with increasing current density. The authors

justified this behavior by the fact that the predominant formation of oligomeric and polymeric by-products. Moreover, in the same work, the authors assessed the influence of ultrasound on potentiostatic measurements. **Fig. 5** (ii) shows the dependence of the distance between the anode and the sound transducer concerning the current densities received for the electrooxidation of *p*-methoxytoluene. Lindermeir et al. noted that both ultrasound configurations showed an increase in current density, but the effect was higher with the forced energy entry. Furthermore, the FEM-model simulations of the sound field in the test cell were performed and verified with the experimental results (see **Fig. 5** (iii)). The authors found that at a distance less than 15 mm, the electrode operates directly in the sound pressure range; however, at a distance more than 15 mm, the bubble protector detaches from the working electrode.

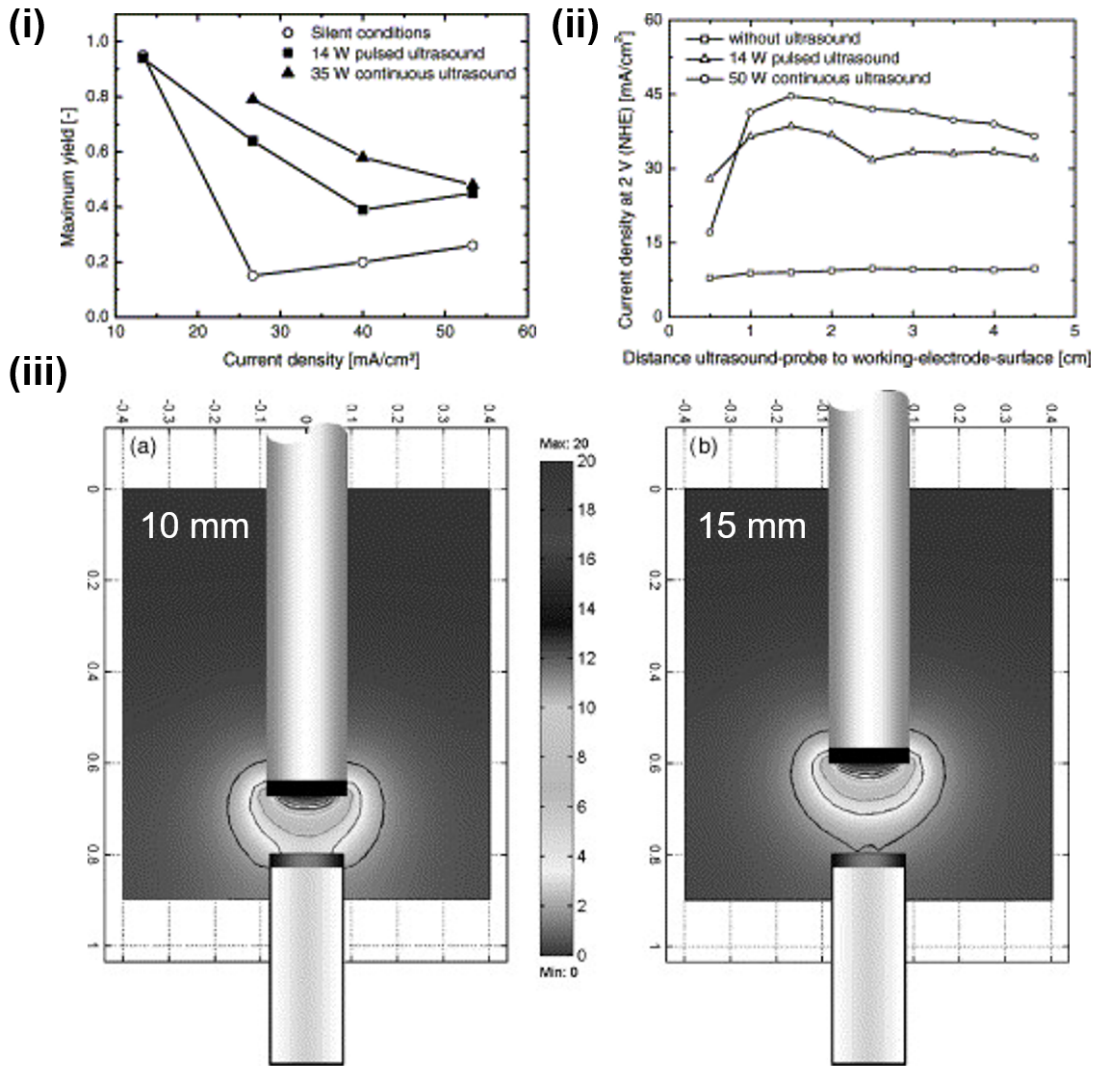


Figure 5: (i) Maximum *p*-methoxybenzaldehyde-dimethylacetal yield under silent and sonificated conditions, (ii) Average current densities as a function of the ultrasound-probe distance to the working-electrode-surface for potentiostatic oxidation, and (iii) Sound pressure field for (a) 10 mm and (b) 15 mm distance between the ultrasound-probe to the working-electrode-surface. Reproduced with permission from ref 103, Copyright 2003, Elsevier.

Sunaga et al. [104] applied the sonoelectrochemical reaction using Et_3N-3HF ionic liquid to a type of Pummerer-type fluorination of organosulfur compounds. The authors showed that the use of ultrasound was very effective when compared to mechanical stirring in

terms of yield, current efficiency, and also in reaction selectivity. It can be regarded in Fig. 6 (ii) that the oxidation current of the compound 1 substantially increased in the process under ultrasonication compared to mechanical stirring, which was not greatly increased. The authors justified it due to the viscosity of the ionic liquid fluoride salt, as this parameter is higher than that of common molecular solvents. Thus, sonication facilitated the mass transport of compound 1 to the working-electrode-surface from the ionic liquid bulk. Moreover, it is evident in Fig. 6 (iii) that the compound 2 e 3 yields increased to 85% and 65%, respectively, under ultrasonication.

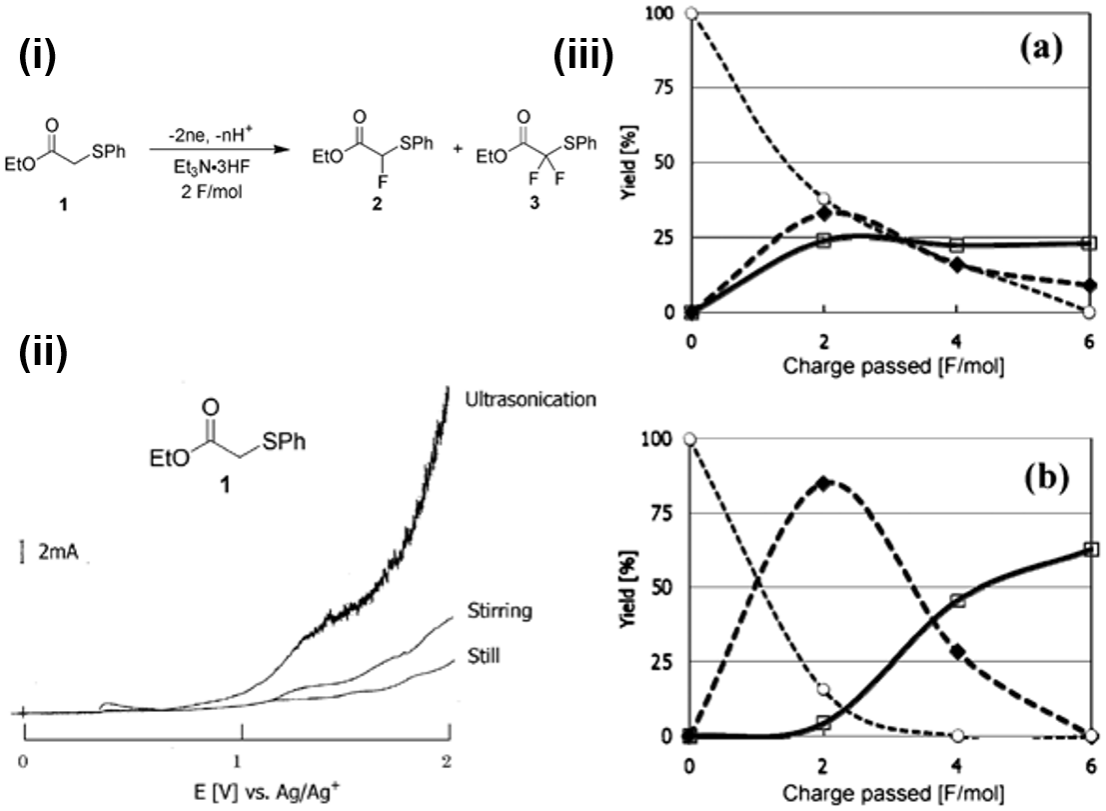


Figure 6: (i) Ultrasound-assisted electrochemical fluorination reaction of organosulfur compounds in $Et_3N{\cdot}3HF$ ionic liquid, (ii) Voltammograms of ethyl α-(phenylthio)acetate in $Et_3N{\cdot}3HF$ ionic liquid, and (iii) Product yields and reagent recovery considering (a) mechanical stirring and (b) ultrasonication: (◆) **2**, (□) **3**, (○) **1**. Reproduced with permission from ref 104, Copyright 2009, The Royal Society of Chemistry.

Paddon et al. [105] presented the electrochemical oxidation reaction of anthracene in acetonitrile at different temperatures under ultrasonic and silent conditions. Overall, the formation of anthraquinone and its reduced form is improved at low temperatures, while the product yield was strongly increased with the addition of ultrasound at low temperatures. The authors justified this behavior due to the superiority of Faradaic currents concerning currents induced by the high mass transfer rates created by ultrasound in electrochemical processes. Another study by the same research group, Paddon et al. [33] studied the sonoelectrosynthetic reduction of an organic iodide in aprotic solvents. The authors showed that the insonation effect on the process monitoring mechanism favored the formation of a monomer.

Moreover, other works focusing on sonoelectrosynthesis can be cited. Et Taouil et al. [106,107] performed the electropolymerization of pyrrole and thiophene derivatives under high-frequency ultrasound irradiation (500 kHz). Yu et al. [108] developed the ultrasound-promoted parallel synthesis of trifluoroatrolactamide derivatives via an one-pot Passerini/hydrolysis reaction sequence. Atobe et al. [109] studied the sonication effects on the emulsification reaction through the allylation reaction performed in an ionic liquid (1-ethyl-3-methylimidazolium BF_4) (**Fig. 7**). This concept was applied first for the anodic substitution reaction of *N*-(methoxy-carbonyl)pyrrolidine **1** with allyltrimethylsilane **2** as a model reaction (**Scheme 1**) with 70% yield and further successfully studied substrates scope.

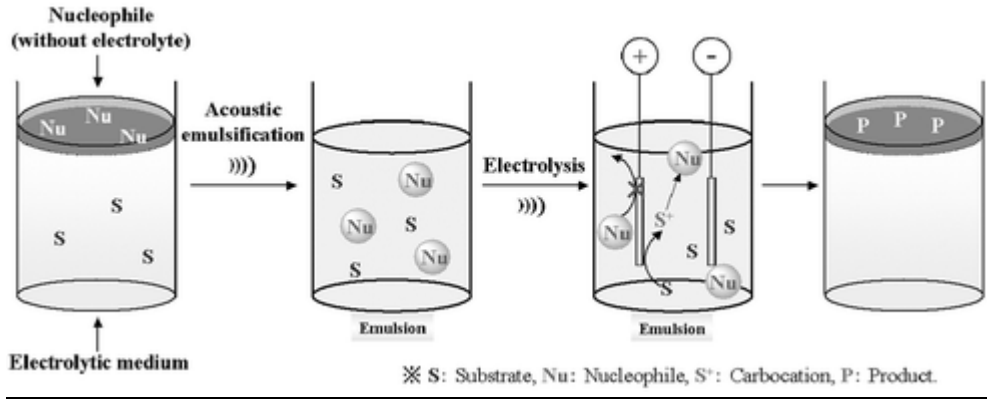
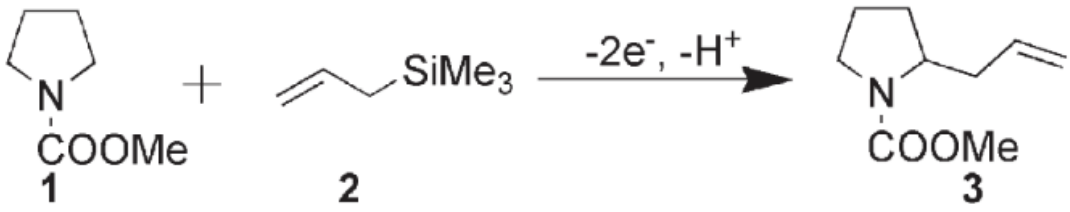


Figure 7: Schematic representation of the anodic substitution reaction system assisted by acoustic emulsification. Reproduced with permission from ref 109, Copyright 2007, The Royal Society of Chemistry.



Scheme 1. Anodic substitution reaction of *N*-(methoxycarbonyl)pyrrolidine 1 with allyltrimethylsilane 2. Reproduced with permission from ref 109, Copyright 2007, The Royal Society of Chemistry.

Wadhawan et al. [110] developed an aqueous protocol for Kolbe's reaction of water-immiscible aliphatic acids by ultrasound and emulsion formation. The authors observed that the product yields obtained from this two-phase system are independent of the material of the electrode used, unlike homogeneous systems.

Therefore, it is evident from the studies reported above that organic sonoelectrosynthesis is still a challenging issue that involves many advantages, limitations, being considered a hotspot mainly for organic synthesis. However, this research field still has a lot to be investigated to establish a reliable and scalable technology.

ADVANTAGES, LIMITATIONS AND THE FUTURE FOR INTENSIFIED ORGANIC SONOELECTROSYNTHESIS

Ultrasound-assisted electrochemical systems offer numerous advantageous outcomes that make this technology interesting for molecular transformations. Unlike other energy sources, the application of ultrasound for chemical synthesis is not connected with the direct interaction of molecules [111]. However, the energy from ultrasound irradiation is channeled into the generation, growth, and collapse of bubbles in the liquid medium, a phenomenon known as acoustic cavitation [112,113]. The bubble collapse effect brings benefits to the system, as this phenomenon is capable of activating chemical transformations without significantly altering the liquid medium. It occurs because the collapse of the microbubbles generates hot spots of temperature and pressure inside them, i.e., high temperatures > 5000 K and high pressures > 1000 atm [111,113,114]. Thus, the ultrasonically generated bubbles play an important role in cleaning the electrode surfaces in electrochemical systems assisted by ultrasound. Consequently, the jets of liquid and shock waves emitted at different times during the acoustic cavitation are the principal phenomena that promote changes in the working-electrode-surface, causing their cleaning and activation. Fig. 8 shows the interaction between individual bubbles as they collapse with each other and against nearby surfaces.

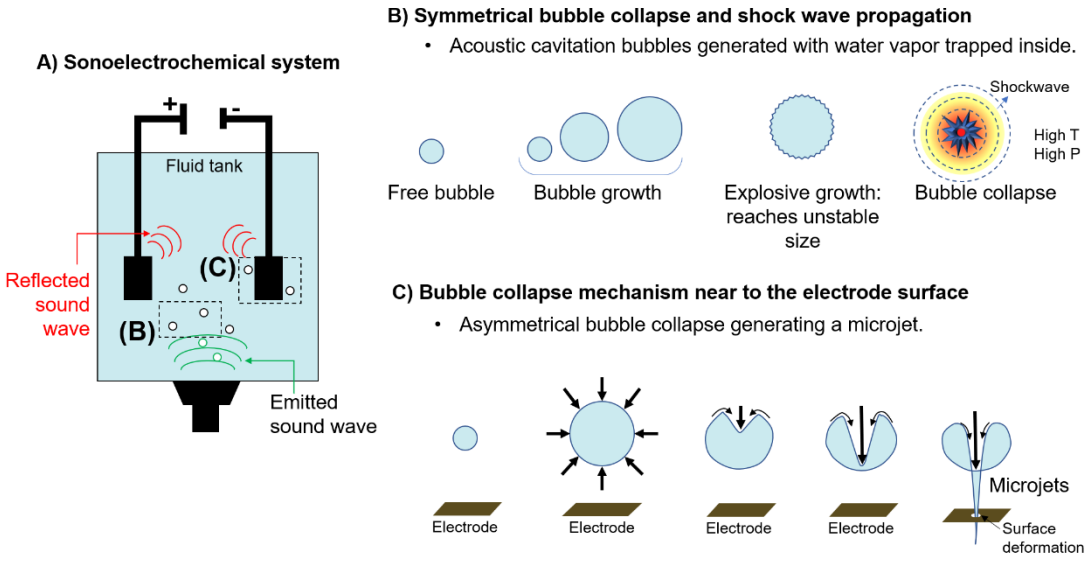


Figure 8: (A) Schematic representation of the sonoelectrochemical system. (B) Mechanism of the collapse of free symmetrical bubbles and (C) Mechanism of the collapse of a bubble near the working-electrode-surface. Adapted with permission from ref 143, Copyright 2019, John Wiley and Sons.

Additionally, for electrochemical synthesis reactions, the application of ultrasound also results in other advantageous effects based on improving the mass transport of ions through the double layer and influencing the reaction path. A product is observed in a reaction under classical conditions, i.e., heating and/or mechanical agitation, then, under ultrasound irradiation, a different product can be formed (different reaction pathway). This phenomenon is named sonochemical switching [115]. Furthermore, the convective flow towards the electrode is boosted due to the ultrasound application, favoring the activation of the electrode surface and, consequently, better cleaning and suppression in the encrustation [116]. An improvement in yield and selectivity is provided as a result of combining these combined effects [25].

However, the boundaries between physical and chemical phenomena for organic sonoelectrosynthesis remain blurred, which contributes to the extended use in the form of

a black box. The reason for this is due to its limitations, mainly due to the complex dependence on physical-chemical phenomena resulting from the effects of cavitation such as jets of liquid, flow, shock waves, and the production of chemical radical molecules [113]. These effects have a complex interrelationship that are difficult to resolve in space and time, impairing the reproducibility of the process. Thus, one of the main limitations of organic sonoelectrosynthesis reactions is the lack of reproducibility. This is due to the fact that ultrasonic bubbles are normally created from impurities randomly distributed within the reactor. Therefore, the cavitation phenomenon is known to be difficult to reproduce [113]. Moreover, the lack of reactors built especially for organic sonoelectrochemistry is another limiting factor for this technology to reach new scientific levels.

Other limitations of these systems are the heterogeneity of the electric field, thermal loss due to heating, and the use of support electrolytes; however, this is associated with sonoelectrochemical batch methods [117]. To work around these limitations, new concepts of process intensification have stood out in science, and they are highly linked to modern chemical engineering because they aim at a more sustainable and efficient way of synthesizing pharmaceuticals and fine chemicals [118,119]. This is achieved by the association of the development of new equipment and techniques to intensify the process, aiming at a significant improvement in the efficiency of the process and the quality of the product, further reducing the waste stream [120]. Thus, miniaturized chemical reaction devices (lab-on-a-chip), known as microreactors, can be used for process intensification and applied in areas such as chemical synthesis [121,122].

This field of flow chemistry (in microreactors) has been gaining interest, proving to be a hot topic for science and engineering [123,124]. Thus, the number of researches has been increasing so that miniaturized devices become commercially available from a small-

scale for use in laboratories as well as for scaling up for the industrial application, mainly to obtain information on new methods of organic synthesis [125–127]. The microreactor technology advantages are the mass and heat transfer enhancement, high surface-to-volume ratio, laminar flow operation, precise residence time control, and short molecular diffusion distances [128,129]. Also, this technology results in better yields with higher selectivity due to regular flow settings [130]. These advantages can be applied to a variety of organic synthesis processes, highlighting the integration of organic electrosynthesis with microreactor technology (flow chemistry) and electrodes for reaction performance (electrosynthesis) [131–135], and the integration of ultrasound with small-scale flow reactors for organic synthesis [56,136–138].

However, the association of organic sonoelectrosynthesis and microstructured devices is still a new area, but it is constantly growing. Acoustic energy, through the application of ultrasound, plays a fundamental role because it has been shown to be effective in reducing the size of agglomerate particles (solids created during a given reaction that accumulate along the walls of a flow reactor), avoiding the clogging of the microreactor channels [139–142]. However, few examples are available in the literature on organic sonoelectrosynthesis using microfluidic devices. Overall, much remains to be studied, but ultrasound-assisted organic electrosynthesis remains an emerging research field in order to explore all of its potential effects. It is believed that this technology serves as a solution to the limitations of the commercialization of synthetic processes and offers a new aspect in industrial synthesis.

CONCLUSION & FUTURE PERSPECTIVE

Technological advancement has transformed the conventional organic synthesis into various swift easy and valuable methods such as mechanochemistry, electrochemical

synthesis and more recently sonoelectrochemical synthesis. The latter is based on combination of electrochemical parameters and physical effects such as high intensity ultrasound through rigorous shock waves stimulated by cavitation in sonoelectrochemical cells. Conventionally these ultrasound waves are often used for emulsions preparation, making uniform dispersions, modification of surfaces and liquid exfoliation of multilayered materials and nanomaterials design. To advance the conventional electrochemical synthesis of organic molecules, the three-electrode system is coupled with sonochemical horn in sonoelectrochemical reaction cell which can enhance the reaction rate, product selectivity and efficiency. This can be applied in both types of redox organic reactions simultaneously and ameliorate mass transport, reducing diffusion layer around working electrode which affects electron transfer and high current densities can be accomplished. Depending upon the reaction nature, different configurations of sonoelectrochemical cells can be designed in-house to improve product selectivity and faradaic efficiency. The major advantage of sonoelectrochemical organic synthesis is that it promotes chemical reactivity in liquid-solid interface through self-cleaning and degassing. Due to several above-mentioned advantages of sonoelectrochemical synthesis, there are various improvements which can be extended to small molecule organic synthesis. Furthermore, due to tremendous efforts for the development of new nobel-metal free electrocatalysts for oxidation and reduction, there are wide opportunities to use recently developed electrode materials as working electrode. Therefore, advanced high surface area nanomaterials such as layer double hydroxides (LDH) nanostructures based on iron, nickel, and cobalt, can certainly replace conventional nobel metal catalyst for oxidation and similarly Pt can be replaced by metal phosphide catalysts. Likewise, for sonoelectrochemical organic reduction reactions 2D materials such as molybdenum sulphide MoS_2 and Mxene can be explored. Screen printing and inkjet printing are

advanced techniques that can be employed for the development of high-performance cost-effective electrodes.

Sonoelectrochemical synthesis can be extended to various other promising reactions such as benzylic C-H bonds to generate arene radical cations which are often hindered by large overpotential. Sonoelectrochemical could possibly drive such reactions at lower overpotential due to ultrasound waves. This can also stimulate hydrogen-atom-transfer (HAT) from benzylic C-H bond to Fe-oxo species, expanded to iodination and various oxidative coupling reactions. Sonoelectrochemical synthesis can also be potentially coupled with photochemistry to drive organic reactions with diverse functional groups. Sonoelectrochemical synthesis can be applied to pharmaceutical intermediate synthesis and biomass conversion reactions. Also, possible mechanism how it promotes chemical reactivity in solid-liquid interface for oxidation and reduction reactions can be suggested for diverse redox organic reactions.

The combination of sonochemistry and electrochemical flow has the advantage of resolving the limitations concerning the handling of solids formed inside the channels. This approach will tackle the main drawback that has limited the scale up and exploitation of this methodology. Applications of microreactors in academia and industry are frequently limited by issues of coping with the formation of solids, either as a desired product or an undesired by-product. Other potential drawbacks that endanger the lifetime of microfluidic reactors are the emergence and handling of solid products, which can cause clogging and encrustation of their channels. Managing solid particles in microchannels is often a difficult task for many research groups, even for a small laboratory-scale synthesis. Therefore, to further explore microreactors, some important challenges must be faced concerning solids handling, so more research are needed on how to avoid clogging and how to control the formation of particles. One strategy to prevent

clogging could be the application of a continuous ultrasound field in the electrochemical flow setup at low frequencies which would successfully breakup particulate agglomerates in the bulk flow, together with wall modifications to provide prevent or limit adhesion and more efficient and increase yields would be expected.

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Conflict of interest

The authors declare no conflict of interest.

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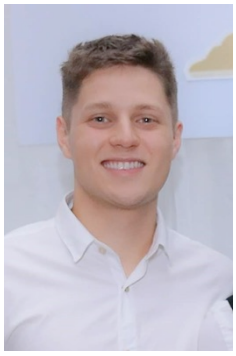
Author's Profiles:

Nisar Ahmed obtained his PhD in organic chemistry (2012) under Brain Korea BK21 fellowship working in the group of Prof. Kwang S. Kim (POSTECH, Korea). Then, he moved to the University of Zurich, Switzerland (2013) for a postdoctoral stay with a Novartis Fellowship. Subsequently (2015), he joined the University of Bristol as a research associate. He started his research career in 2017 at School of Chemistry, Cardiff University, United Kingdom with Marie Curie COFUND fellowship as Sêr Cymru Fellow. Subsequently, he was appointed EPSRC Investigator (UKRI) at Cardiff School of Chemistry, Cardiff University. He is also an Adjunct faculty at HEJ Research Institute of Chemistry, ICCBS since 2018. His fields of research include value-added chemicals synthesis using modern synthetic tools (electro-, photo-, sono-, flow-, digital-, automated) and molecular recognition of biomolecules using fluorescent probes (supramolecular chemistry)



Higor de Bitencourt Rodrigues received his bachelor's degree in Chemical Engineering (2015-2019) from Franciscana University, Brazil. He is currently a Master's student at the Federal University of Santa Catarina (UFSC), Brazil, under the guidance of Dr. C. Soares and Dr. N. Padoin. His research focuses on the contributions of the microreactor technology to the development of green and sustainable synthesis in chemical and biotechnological processes. He has experience in Chemical Engineering with a focus on the following areas: organic chemistry, chemical kinetics, and analytical chemistry.

Jéssica Oliveira de Brito Lira is currently a Ph.D. student in Chemical Engineering at the Federal University of Santa Catarina (UFSC), Brazil, under the supervision of Dr. C. Soares and Dr. N. Padoin. Her Ph.D. studies are supported by a CAPES Scholarship. She received her B.Sc. (2016) and M.Sc. (2019) in Chemical Engineering from Federal University of Paraiba and Federal University of Santa Catarina, respectively. Her current research interests in Dr. C. Soares' group are focused on CFD-based numerical simulation of photo-microreactors.

Natan Padoin studied Chemical Engineering at the Federal University of Santa Catarina (UFSC), Brazil, completing his Master's and Ph.D. degrees in Chemical Engineering at the same institution in the field of mathematical modeling and CFD simulation of multiphase flow, involving heat and mass transfer, and microfluidic reactors, under supervision of Dr. C. Soares. Currently, he is an adjunct professor at the Department of Chemical and Food Engineering of the Federal University of Santa Catarina, carrying out his research as a permanent professor at UFSC's Chemical Engineering Graduate Program, within the Laboratory of Materials and Scientific Computing (LabMAC), in the main fields of process systems engineering (PSE) and chemical reactors, with a focus on computational fluid dynamics (CFD), multiphase reactive flow (chemical reactor design, heat and mass transfer, etc.), microfluidics and nanotechnology.

Cíntia Soares studied Chemical Engineering at the University of Blumenau (FURB), Brazil. She received her master's and Ph.D. degrees in Chemical Engineering from the University of Campinas (UNICAMP), Brazil, working on computational fluid dynamics (CFD) applied to multiphase flows. Dr. Soares is currently an associate professor at the Department of Chemical and Food Engineering at the Federal University of Santa Catarina (UFSC), Brazil. She carries out her research as a permanent professor at UFSC's Chemical Engineering Graduate Program, within the Laboratory of Materials and Scientific Computing (LabMAC). Dr. Soares research interests are focused on computational fluid dynamics (CFD), multiphase reactive flow (chemical reactor design, heat and mass transfer, etc.), microfluidics and nanotechnology.

Ahsanulhaq Qurashi received his PhD in 2008 Chonbuk national University, South Korea, and completed postdoctoral fellowship from Toyama University, Japan (2008-2010). Currently he is Associate Professor at department of Chemistry Khalifa University. His research is focused on the development of advanced functional nanomaterials for Clean Energy, Catalysis and Chemical Sensor applications. He is editor of book "Metal Chalcogenide Nanostructures for Renewable Energy Applications", Subject Assistant Editor to International Journal of Hydrogen Energy, and editorial board member of Scientific Report, and MRB. He has published more than 90 papers in internationally reputed peer review (ISI) journals and presented over 55 papers in various international conferences.

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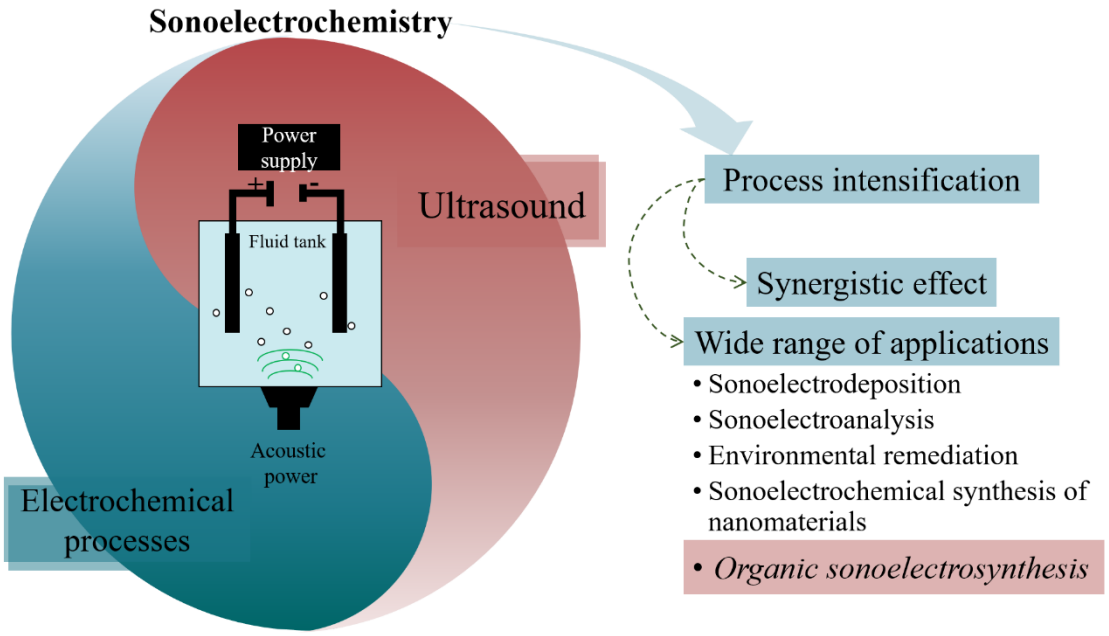

Ultrasound-assisted electrochemical processes stand out as a sustainable strategy giving innovative dimensions to organic synthesis due to its synergistic effect