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Shang Jiang, Evelyne Kafui Yawa Late, François Jerome, Prince Nana Amaniampong, Samir Mushrif. Ultrasound-Assisted Catalysis: A Pathway to Novel and Selective Chemical Transformations in Condensed Phase. *ChemistrySelect*, 2024, 9 (35), pp.e202401953. <10.1002/slct.202401953>. <hal-04785461>

**HAL Id: hal-04785461**

**<https://hal.science/hal-04785461v1>**

Submitted on 15 Nov 2024

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# Ultrasound-Assisted Catalysis: A Pathway to Novel and Selective Transformation of chemical Substrates in Condensed Phase

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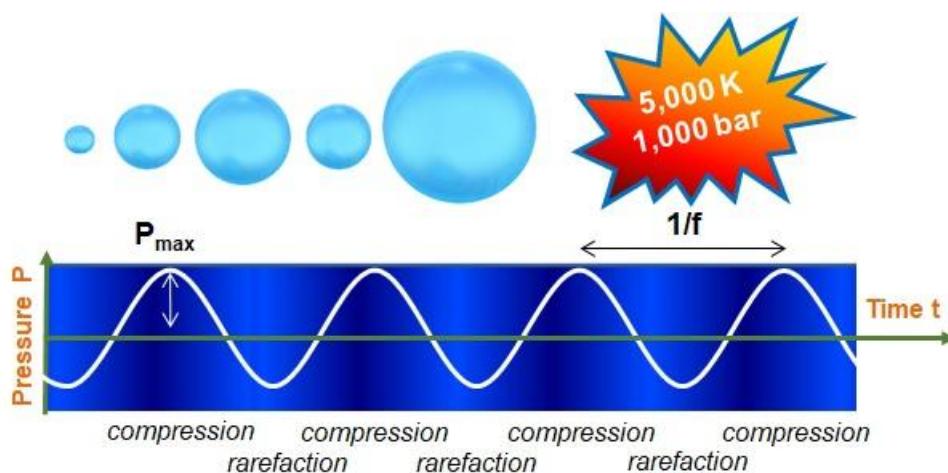
Keywords: Sono-catalysis. Ultrasound, Heterogeneous Catalysis, Catalysis, Radicals

## **Abstract:**

In this paper, we introduce the concept of synergistically using ultrasonic sonochemistry in combination with heterogeneous catalysis, to selectively perform condensed phase chemical transformations. Through specific examples, we highlight that ultrasound induced cavitation bubbles not only provide local high temperature and pressure conditions for initiating chemical reactions but also improves heat, mass and charge transfer, crucial for enhancing productivity of chemical reactions. These examples also show that sonocatalysis can alter reaction mechanisms and open new activation and reaction pathways that are not intuitive to thermocatalysis. Computational chemistry tools are introduced and combined with sonocatalysis to demonstrate that, in-situ generated radicals propelled into the bulk liquid solution upon bubbles implosion can be stabilized by catalyst surfaces, while creating active sites for unique reactions during heterogeneous sonocatalytic chemistry. We also report recent examples of condensed phase sonocatalysis applications, including pollutant degradation, chemical and pharmaceutical synthesis and biomass upgrading, and provide mechanistic insights into the process.

## 1. Introduction

Ultrasonic sonochemistry has emerged over the last decade as a promising disruptive and unconventional technology in the field of sustainable chemical synthesis and catalysis. Its emergence is fueled by the ever-mounting concern for environmental impacts and the quest for alternative technologies for assisted catalysis. Of course, this transformative journey owes much to the pioneering works of Loomis and associates,<sup>1</sup> who unearthed the profound physical and chemical effects triggered by ultrasound frequencies. Initially restricted to a specialized domain, ultrasonic sonochemistry is now used in multidisciplinary fields, for instance in the area of nanotechnology for material synthesis, drug delivery, chemical synthesis and environmental remediation, among others. The heart of sonochemistry lies in the irradiation of liquids with ultrasound, a process that locally induces pressure fluctuations.<sup>2,3</sup> When a liquid experiences a localized pressure decrease, it plunges below its vapor pressure threshold, thus generating a cavitation bubble. This bubble, composed of gas and vaporized liquid, expands in size (*Figure 1*) before eventually collapsing violently. As it absorbs energy from the surrounding sound waves, it creates extreme yet confined conditions within its imploded cavity; conditions characterized by extremely high temperatures and pressures.<sup>4-6</sup> Simultaneously, this dramatic collapse is accompanied by shock waves, high-speed jets, and reactive radicals. These extreme conditions serve as primary catalysts for the chemical and physical transformations brought about by acoustic cavitation in liquids.<sup>1,6,7</sup>



**Figure 1.** Phenomenon of cavitation bubbles compression and rarefaction under ultrasound

A cavitation bubble can be viewed as a micro-reactor from a chemical synthesis perspective. These microscopic bubbles create an environment where extreme conditions of pressure (reaching up to 1,000 bar) and temperature (5000 K) give rise to unique chemical reactions.<sup>1</sup> For instance, when neat water is subjected to irradiation at high-frequency ultrasound (>100 kHz), tiny cavitation bubbles are generated that serve as micro-reactors. Inside these bubbles, water molecules undergo dissociation to form •OH and •H radicals.<sup>4,7</sup> Upon the bubble's implosion, these radicals are propelled into the surrounding bulk liquid, initiating chemical reactions. Although of significant interest, a major challenge in utilizing ultrasound for chemical synthesis in a catalyst-free environment is the control of product selectivity. This is because of the high reactivity of radicals. As a result, current sonochemistry approaches are used for total oxidation reactions, particularly for environmental applications. To solve these issues related to controlling reaction selectivity, the use of catalytic materials in sonochemistry applications, often coined as sonocatalysis, has emerged. The concept of sonocatalysis has been studied for decades, primarily using low frequency ultrasound (<80 kHz). However, under this condition, the formation of radicals is limited, and mainly physical effects (mass transfer, turbulent flow, shockwaves, etc) occur. Sonocatalysis utilizes turbulent flow and shock waves from cavitation bubble implosion to enhance catalyst/reactant dispersion, mass and heat transfer, thereby improving reaction rates. Ultrasonication can make chemical reactions feasible by providing the necessary kinetic energy to reacting species, resulting in a more effective collision with the catalyst surface. This phenomenon has been observed in various reactions such as oxidation of carbohydrates, vanillin, and oxidative depolymerization of lignin. In intriguing scenarios, a synergistic interaction between ultrasound and catalysts results in reactions typically unattainable without ultrasound. This synergy, particularly noticeable with solid catalysts, opens avenues for otherwise impossible chemical reactions. This paper presents the concepts of sonocatalysis, the interactions between ultrasound, catalysts and solvents under sonocatalytic reaction conditions through selected examples, and some recent major applications of sonocatalysis.

## **2. Concepts of Sonocatalysis**

Catalyst-free cavitation shows promise for enhancing chemical reactions and changing reaction mechanisms, but it often requires high pressure and acoustic power to initiate bubble formation.

In contrast, in heterogeneous solutions, cavitation bubbles form mainly on solid particle surfaces due to heterogeneous nucleation. This phenomenon is vital in sonochemistry, allowing better transfer of radicals to solid catalyst surfaces and maximizing the localized thermal effect produced during bubble implosion on catalyst surfaces, leading to improved substrate conversion and reaction selectivity. However, in sonocatalysis, the enhancement of reaction rates benefits from enhanced interactions between cavitation bubbles and catalytic materials. Currently, there is limited understanding and link between catalysis and sonochemistry.<sup>8-11</sup> As such, the use and control of catalysis in ultrasound applications remains almost a black box. In this context, engineering catalytic materials with surface properties critical for enhancing particle-bubble interactions are crucial.

Figure 2 highlights a few key advantages of utilizing ultrasound during catalytic conversion. The catalyst does not only provide an active reaction site but also is able to help stabilize some key radicals dissociated from solvent via ultrasound to skew the reaction towards a desired selective path. For example, studies have shown promising results for biomass upgrading to desired products in sonocatalytic systems,<sup>12-15</sup> under specific ultrasound conditions and with an appropriate choice of catalysts. Previous works have demonstrated that the combination of high-frequency ultrasound and metal oxide catalysts can lead to unique and selective transformations such as glucose to glucuronic acid<sup>16</sup> (oxidation) and benzyl alcohol to phenol (demethylenation) (Figure 3), to name a few.<sup>17</sup> Sonochemistry faces two predominant challenges: targeting selective chemical transformation with specific frequency of ultrasound and achieving higher selectivity/conversion/yield at a lower energy expense than the traditional thermo-catalytic process. The selection of a heterogeneous catalyst also is critical for process selectivity, necessitating designs that can endure the mechanical and thermal stresses from ultrasound-induced cavitation. Catalysts in sonocatalytic systems often risk deactivation due to these high-energy conditions, and catalyst leaching can lead to product contamination and material loss.<sup>18, 19</sup> Loning et al.<sup>20</sup> highlighted that energy transformation in sonochemistry is significantly influenced by the properties of the irradiated medium, with high volatility and viscosity reducing energy transfer. High volatility leads to the formation of vapor-filled cavities that absorb less energy, while high viscosity impedes the propagation of ultrasonic waves, both resulting in reduced energy transfer efficiency. Moreover, with the assistance of ultrasound, solvents can also play a role in chemical transformation by directly participating in the chemistry. Amaniampong

et al.<sup>21</sup> reported that H<sub>2</sub>O<sub>2</sub> can be dissociated into OH radicals under high-frequency ultrasound, and OH are stabilized on the CuO catalyst surface (Figure 3, b). This surface stabilized OH groups are essential for glycerol's initial activation, subsequent C–H activation, and finally incorporate themselves into glyceric acid selectively. Moreover, this also prevents the reduction of metal oxide via conventional Mars-Van Krevelen mechanism, thus enhancing the stability of the catalyst. In another case (Figure 3, c), ultrasound-induced, CuO catalyst stabilized OH groups are also vital to initiate the conversion of benzyl alcohol to phenol.<sup>17</sup> Similarly, high-frequency ultrasound in this case also aids in dissociating solvent water to form adsorbed OH groups, which lowers the activation barrier of key activation steps and incorporate themselves into final products. On the other hand, selecting a suitable frequency of ultrasound for a given reaction remains a challenge. Bößl et al.<sup>22</sup> explored the effects of ultrasonic frequency and power on piezocatalytic and sonochemical dye degradation. They find that lower frequencies (<100 kHz) enhance piezocatalysis through acoustic cavitation, while higher frequencies (>100 kHz) favor sonochemical degradation, potentially eliminating the need for a piezocatalyst. Therefore, selecting a suitable frequency of ultrasound, catalysts, and solvents is vital in sonochemistry, contributing to a process characterized by enhanced stability, activity, and selectivity.

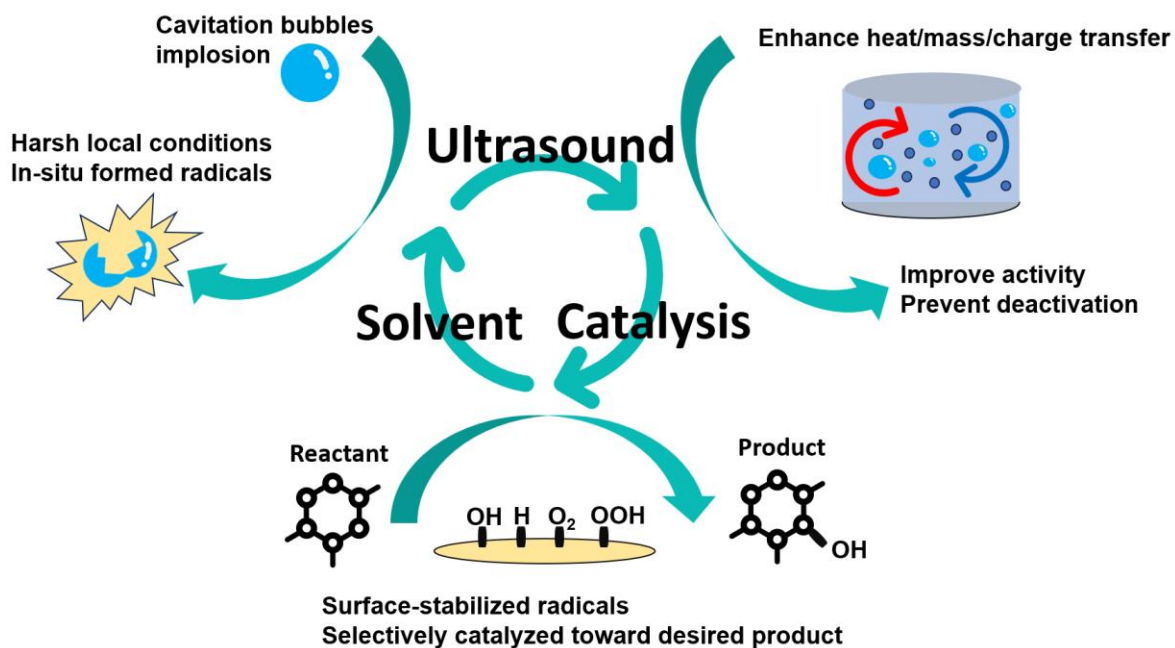


Figure 2. A schematic overview for the synergy between ultrasound/solvent and catalysis to create local harsh conditions, enhanced heat/mass/charge transfer and surface-stabilized, in-situ formed radicals.

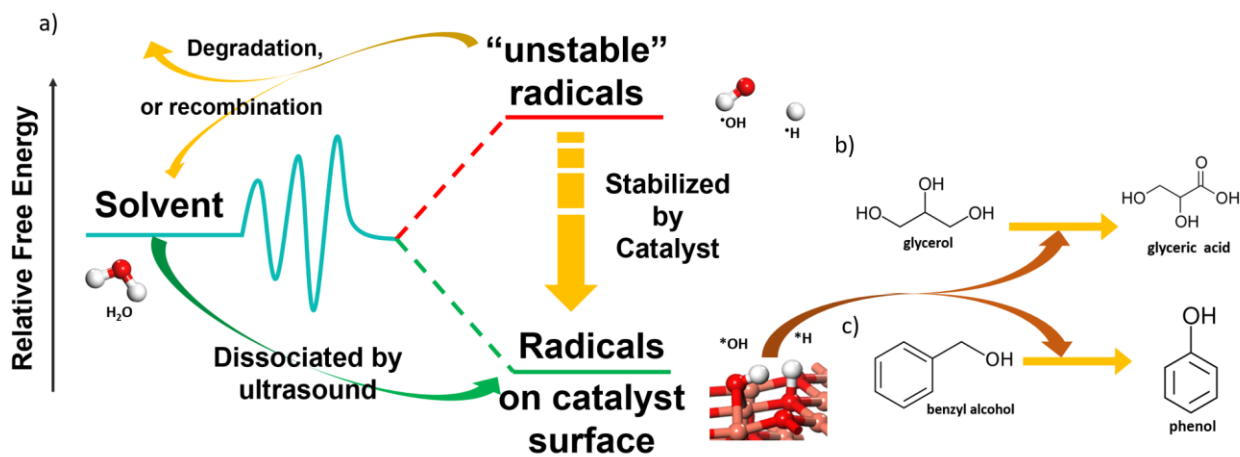


Figure 3. a) Solvent dissociated by ultrasound and stabilized by catalyst surface and ultrasound-assisted unique and selective transformations for (b) benzyl alcohol demethylenation to phenol<sup>17</sup> and (c) glycerol oxidation to glyceric acid<sup>21</sup>

In the context of tandem catalysis and sonochemistry, or sonocatalysis, the liquid-solid interface is also crucial. Similar to conventional heterogeneous catalysis, active sites on catalysts aid in adsorbing reactants, forming intermediates, desorbing products, and lowering activation energies. In sonocatalysis, in-situ formed radicals induced by ultrasound from solvent, can be stabilized on the catalyst surface, acting as key reactive intermediates. For instance, CuO can inhibit rapid  $\bullet\text{OH}$  recombination into  $\text{H}_2\text{O}_2$ , and utilize these radicals dissociated from water due to ultrasound.<sup>16, 17</sup> Additionally, for metal oxide catalysts such as CuO, they are less likely to be reduced during the sonocatalysis reaction. These in-situ formed, oxygen-containing radicals (such as  $\bullet\text{O}$  and  $\bullet\text{OH}$ ) can replenish reduced oxide catalysts sites, or in some cases, these surface-stabilized radicals can directly participate into the reaction as preferred reaction pathways instead of using surface lattice oxygens. Other metal oxide catalysts such as titanium dioxide, zinc oxide, and copper oxide<sup>23-27</sup> are frequently selected for their reactivity and stability. Similarly, metals like palladium and platinum<sup>28, 29</sup> are popular for their exceptional catalytic properties and good

stability under local harsh conditions. These catalysts are often utilized in the degradation process of dye and pharmaceutical pollutants in wastewater and they have shown excellent resistance to mechanical stress under ultrasound and collapse of ultrasound-induced cavitation bubbles.

Additionally, ultrasound also enhances mass, heat, and charge transfer at the liquid-solid interface (Figure 4).<sup>19, 30</sup> Dehane et al.<sup>31</sup> conducted numerical simulations that elucidate how ultrasound improves mass and heat transfer by enhancing the internal energy transport within acoustic bubbles, which leads to higher production rates of chemical species such as  $\bullet\text{OH}$  and  $\bullet\text{H}$ . This increase in transfer efficiency is particularly notable at higher acoustic amplitudes and specific frequencies. Regarding charge transfer, Zhang et al.<sup>32</sup> recently reported that in the sonocatalytic degradation of tetracycline hydrochloride, ultrasound accelerates charge transfer and the separation of electron-hole pairs. This is achieved through the S-scheme heterojunctional interface between the  $\text{CoFe}_2\text{O}_4$  and  $\text{g-C}_3\text{N}_4$  catalysts. When the semiconductor catalyst,  $\text{CoFe}_2\text{O}_4$ , absorbs energy from ultrasound, it excites an electron from the valence band to the conduction band. This leaves behind a "hole" in the valence band. The electron and the hole together constitute an electron-hole pair. This phenomenon was supported by strong evidence of interaction between the components, as confirmed by photoluminescence and photocurrent analyses. These findings explain the benefits of ultrasound in enhancing mass, heat, and charge transfer processes, thereby significantly improving the activity of sonocatalytic reactions.

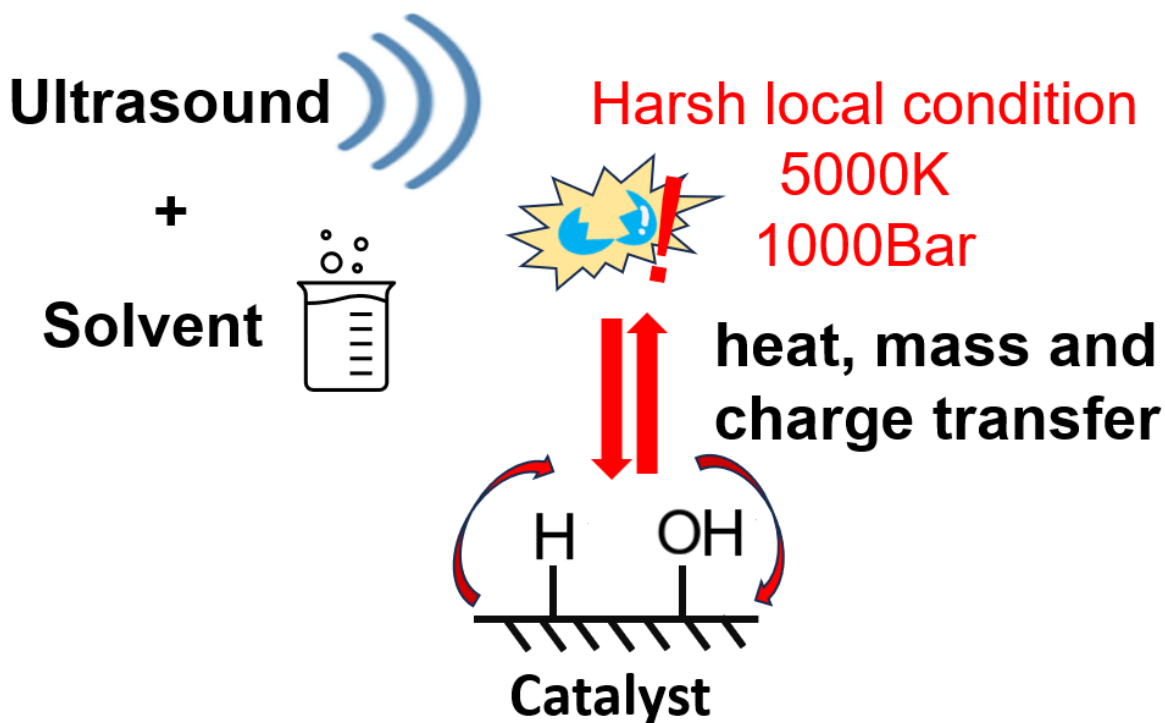


Figure 4. Heat, mass and charge transfer under ultrasonic liquid-solid interface.

### 3. Combining Computational Chemistry with Sonocatalysis

Addressing these challenges in sonocatalysis, particularly maximizing interactions with cavitation bubbles and harnessing the energies of bubble implosion involves designing catalysts that not only withstand the intense conditions of cavitation but also effectively utilize in-situ formed radicals. However, the mechanism of radical formation, the specific interactions between cavitation bubbles and catalytic materials during sonolysis, and the adsorption of in-situ generated radicals, reaction substrates and intermediates are unclear when relying solely on experimental techniques. To overcome these hurdles, a combination of experimental and theoretical tools is essential, with a focus on filling fundamental knowledge gaps in catalyst behavior under sonochemical conditions and the dynamic interaction of in situ formed radicals, as well as their roles in the reaction mechanisms and pathways. Theoretical calculations, such as first-principles calculations and hybrid Quantum Mechanics/Molecular mechanics (QM/MM) modelling, however, can elucidate the role of solvent and in-situ formed radicals, provide

insights by predicting the stability of these critical species, determine their role in the most favorable reaction pathways and the reaction mechanisms underlying sonocatalytic processes. First-principles calculations such as density functional theory (DFT) calculations can determine the detailed reaction mechanism for radical formation and most favored reaction pathways by comparing the activation energy barrier of each possible elementary reaction step. Meanwhile, QM/MM can model the chemical reactions and electronic properties of systems while maintaining the balance between accuracy (quantum mechanics for detailed reaction mechanisms, activation barriers and transition states) and computational cost (molecular mechanics for cost-effective prediction with larger systems over longer timescale). For more details and examples of different advance computational techniques help understanding reactions in condense phase, Varghese et al.<sup>33</sup> showed that the crucial role solvents can play in chemical process by influencing the reaction kinetics, thermodynamics, and selectivity of chemical reactions in their review paper.

In the work<sup>17, 21</sup> previously discussed in section 2, DFT calculations reveal the reaction pathway, while also show that the dissociation of solvent under high-frequency ultrasound, and the stabilization of in-situ formed radicals by CuO catalyst, participating in the phenol's formation. These DFT results are combined and verified with various experimental techniques, including radical scavenging experiments, H<sub>2</sub>O<sub>2</sub> analysis and isotope labelling experiments. Moreover, DFT calculations can also analyze the electronic structure to understand the interaction between radicals and catalyst under ultrasound. For example, Fan et al.<sup>34</sup> calculated the frontier electron densities to provide information of active sites and prediction of the reaction sites on ciprofloxacin that are attacked by radicals, formed under ultrasound conditions. This synergistic approach effectively overcomes the limitations of experimental methods. Moreover, Bian et al.<sup>35</sup> performed DFT calculations to determine the thermodynamic parameters, electron energies, and reactivity of the reactants, intermediates, and products involved in the sonochemically induced electron transfer controlled atom transfer radical polymerization process. These calculations provided critical insights into the electron transfer mechanism, confirming that DMSO•• radical can react with Me6TREN to generate electron transfer essential for reducing CuIIBr/L+ to CuI/L+, a key step for radical polymerization. The detailed elementary reactions activation barriers from DFT results are further combined with kinetic modeling, providing a

comprehensive understanding of the polymerization kinetics and validating the multi-scale modeling strategy by accurately predicting the polymerization behavior under various conditions.

#### **4. Major applications of sonocatalysis**

Recently, the field of sonocatalysis has experienced a surge of innovative transformations, showcasing its increasing relevance in numerous industries (Figure 5). It is also worth noting that acoustic chemistry also finds extensive applications into catalyst synthesis. Sonochemistry has proven to be a valuable technique in catalyst preparation, offering advantages such as increased surface area, improved dispersion of active sites, and the creation of novel micro/nanostructures without requirement of high temperature calcination.<sup>36-40</sup> For instance, Wang et al.<sup>41</sup> showed that ultrasonic exfoliation process can help create carbon quantum dots on modified graphitic carbon nitride, leading to enhanced oxygen reduction reaction activity. Similarly, Hu et al.<sup>42</sup> also reported that core-shell nanoparticle catalysts synthesized by ultrasonic method exhibits significantly higher activity for oxygen reduction reaction induced by metal oxide on edges and vertexes. The catalyst prepared by ultrasonic hydrothermal-sintering also shown significant improvement for oxygen evolution reaction.<sup>43</sup> In a recent review paper, Guan et al.<sup>44</sup> reviewed the various ultrasound-assisted catalyst synthesis techniques, and their significant improvements in electrocatalysis reactions due to improved dispersion and catalyst surface area.

One of the most impactful applications for sonocatalysis is the degradation of organic pollutants in wastewater.<sup>24, 45-47</sup> Specifically, utilizing ultrasound with titanium dioxide-based catalysts can effectively break down complex organic compounds, like pharmaceutical residues and industrial dyes in wastewater. Afzal et al.<sup>23</sup> demonstrated that integrating biochar with titanium dioxide improves the adsorption of pharmaceutical residues (like ciprofloxacin and levofloxacin) in wastewater, while ultrasound, through cavitation-induced high-temperature and high-pressure hot spots, generates hydroxyl radicals from water molecules, both essential for the efficient degradation of ciprofloxacin. Motlagh et al.<sup>48</sup> also showed similar promising results (over 70% degradation efficiency in 2 hours without additional agents) for titanium dioxide nano-catalyst supported on biochar on ultrasound-assisted pharmaceutical levofloxacin degradation. In this case, Motlagh et al.<sup>48</sup> noticed that ultrasound not only aids in the dissociation of water molecules,

but also provides acoustic energy to the catalyst particles ( $\text{TiO}_2$  and  $\text{ZrO}$ ), promoting electrons from the valence band to the conduction band, leaving behind holes in the valence band. As such, more electron/hole ( $e^-/h^+$ ) pairs can be created on the catalytic surface under ultrasound, ultimately generating more hydroxyl and superoxide radicals to enhance the degradation efficiency. Nuengmatcha et al. demonstrated that a hybrid catalyst composed of  $\text{ZnO}/\text{graphene}/\text{TiO}_2$  exhibited higher efficiency in degrading dye pollutant as well.<sup>25</sup> Similarly, ultrasound irradiation can excite the semiconductor particles ( $\text{ZnO}$  and  $\text{TiO}_2$ ) in the composite. In this case, graphene can act as an electron acceptor and transporter, reducing the recombination rate of the electron/hole pairs. By facilitating better charge separation and transfer, the sonocatalytic efficiency of the composite is improved, making it more active and effective in degrading pollutants.

In the pharmaceutical industry, sonocatalysis has emerged as a key technology for the synthesis of drug precursors and active pharmaceutical ingredients.<sup>49,50</sup> Duran-Valle et al.<sup>51</sup> demonstrated that combining ultrasound with alkaline-doped carbon catalysts enhances the synthesis of antibacterial chalcones, yielding better results and selectivity under mild conditions compared to traditional methods. During this sonocatalysis process, the ultrasound-induced cavitation does not only activate the catalyst with local harsh conditions, but also influences the surface properties of the carbon catalyst. The alkaline doping (Cs-Norit) modulates the basicity of the carbon surface, enhancing its Lewis basic character. The ultrasonic waves, through cavitation, can further create more active sites and alter the distribution of the Cs-dopant on the carbon surface, thereby improving the catalyst's performance. This sonocatalytic approach boosts reaction yields (from 13% to 25%), positively influencing catalyst activity, while also offering an eco-friendlier alternative by minimizing hazardous chemicals and energy use. It facilitates precise control over reaction conditions, essential for producing high-purity pharmaceuticals.

Sonocatalysis has also emerged as a promising method in biomass upgrading, offering a more efficient and environmentally friendly approach. Hu et al.<sup>52</sup> devised an efficient and cost-effective method for producing monosaccharides from soybean and corn straw using 1-H-3-methylimidazolium chloride with very high yield (achieving up to 53.27 mg from 0.2 g of soybean straw and 50.03 mg from 0.2 g of corn straw). The cavitation, high temperature and turbulence generated by ultrasound creates disruption, which enhances the reaction interface

between cellulose and the ionic liquids. The combination of ionic liquids and ultrasound accelerating cellulose cracking by selectively disrupting its hydrogen bonds within cellulose's polysaccharide chains, thereby enhancing its dissolution. This is majorly attributed to the enhanced movement rate of chloride ions under ultrasound, which accelerates the dissolution of cellulose. This process, aided by enhanced heat transfer and weakened hydrogen bonds, significantly boosts the yield of small sugar molecules. On the other hand, Toukoniity et al.<sup>53</sup> found that while acoustic irradiation does not improve selectivity in d-fructose to d-mannitol hydrogenation, it significantly prevents Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> catalyst deactivation. The cavitation bubbles created by ultrasound violently collapse on or near the catalyst surface, directing jets of solvent towards the catalyst surface. The resulting shock waves cause localized deformation of the catalyst surface, leading to intensive cleaning. This process cleans the catalyst surface, and exposes fresh, highly active areas, enhancing catalyst activity by improving mass transfer and reducing diffusion length in the catalyst pores. This clean effect ensures efficient delivery of reactants to the catalyst surface and rapid removal of products, thereby reducing surface poisoning or blockage risks, prevents catalyst activity decline over time.

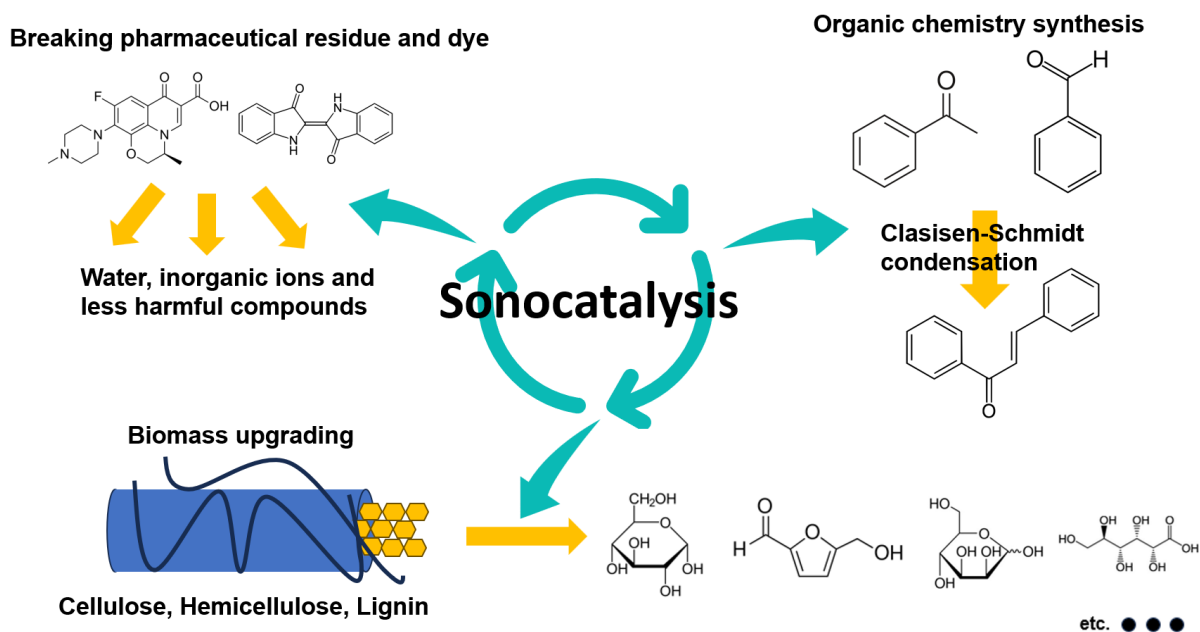


Figure 5. Three major applications of sonocatalysis: pharmaceutical residue and dye pollutant degradation, organic chemistry synthesis, and selective biomass upgrading.

## 5. Outlook and Future Perspectives

This review introduces the field of sono-catalysis, highlighting the synergy between ultrasound, solvent and catalysis to create local harsh conditions, enhanced heat/mass/charge transfer and surface-stabilized, in-situ formed radicals. It emphasizes the unique capabilities of ultrasonic waves in enhancing reaction selectivity and efficiency, and the critical role of catalyst design and solvent interactions.

Future research directions in sonocatalysis are encouraged to focus on existing challenges, including controlling ultrasound frequency for targeting selective chemical transformations. Additionally, the mechanisms underlying catalyst deactivation, particularly the impact of radicals generated by ultrasound and mechanical strain damage caused by ultrasound-induced cavitation collapse, need to be investigated. Understanding how these factors contribute to catalyst degradation and deactivation will be crucial in designing more durable and efficient catalysts utilized under ultrasound conditions. There is also a need to explore the efficiency of ultrasound-induced electron and charge transfer processes, which are pivotal for enhancing catalytic performance. Advanced computational tools, such as *ab initio* molecular dynamics (AIMD), are promising for simulating these processes in the condensed phase, providing deeper insights into the ultrasound-induced charge/electron transfer enhancement. Comprehensive studies combining experimental and theoretical techniques will not only elucidate fundamental aspects of sonocatalysis but also guide the development of more robust and effective catalytic systems for various applications. Overall, sonocatalysis is a powerful tool to unlock new opportunities in green chemical processes, and further investigations are encouraged for sonocatalysis to become a cornerstone of modern chemistry.

## 6. Acknowledgements

PNA, EKYL and FJ are grateful to the CNRS, Research Federation INCREASE, the University of Poitiers, and the Region Nouvelle Aquitaine. The ANR JCJC AminoSound Project ID ANR-20-CE07-0006 is acknowledged for financial support.

SHM and SJ extend their gratitude for the financial support received from the Natural Sciences and Engineering Research Council of Canada (NSERC) and the Faculty of Engineering, University of Alberta, Canada.

## 7. Conflict of Interest statement

All authors declare that they have no conflicts of interest.

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