

# Improving hydrogen generation from biomass and solid waste by combining sonophotocatalysis through carbon-based materials: challenges and future perspectives

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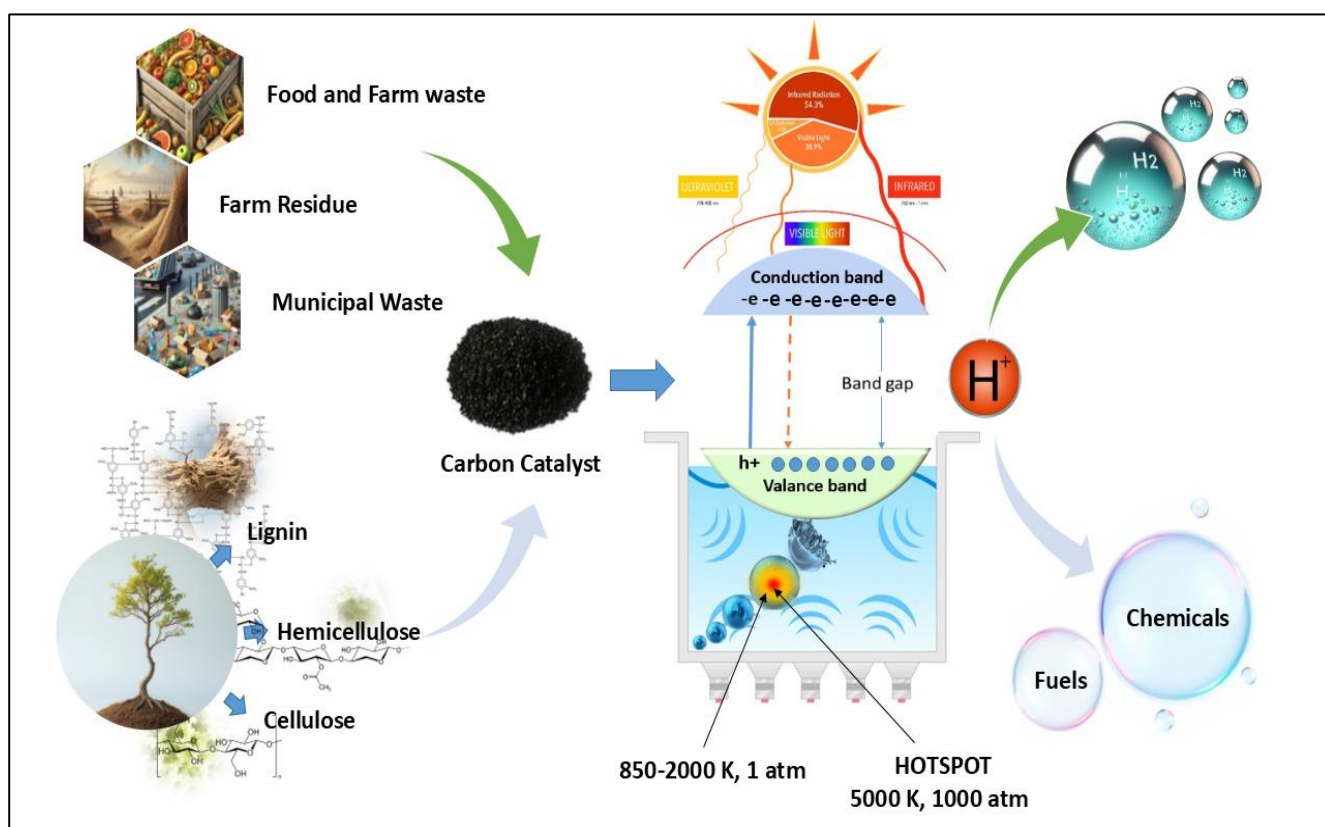
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## Abstract

Environmental pollution is becoming one of the main issues facing human civilisation as a result of the world's growing industrialisation. Using limitless solar energy, photocatalysis may be used to address environmental issues. Whilst, the incorporation of ultrasonic cavitation in various processes such as the Fenton-like process, electrochemical process, and especially sono-photocatalysis offer ideal conditions for increasing biomass fragmentation and conversion into target products and green hydrogen within a lesser reaction time. The scientific community and industry have taken a serious interest in the intensification of ultrasonic processes for a variety of applications, as well as the resulting synergistic effects based on conventional comparisons in literature. In light of this, the purpose of this review is to summarize the current research in the field, provide an overview of the recent studies on ultrasound-assisted different catalytic processes for hydrogen production and discuss their limits and validity, comparing them to conventional techniques. Finally, a few future perspectives in the form of suggestions on ultrasound-assisted photocatalysis for hydrogen production from various biomass models and different wastes, and highlighting the role and recent evolution of carbon-based materials in this field, are presented together with current issues and challenges to clarify future attitudes in the hopes of exploring this ideal combination for large-scale H<sub>2</sub> production.

**Keywords:** Sonication, lignocellulose biomass valorization; sono-photocatalysis; carbon-based materials; hydrogen evolution

# Graphical Abstract



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## 1.Introduction

The demand for energy and products is under immense strain as a result of the rapid economic development in emerging nations and the exponential expansion in the human population. In order to keep up with the ever-increasing energy demand, fossil fuel resources have been heavily mined since the start of the industrial revolution. The use of fossil fuels increases as a result. For instance, by 2050, world energy consumption is expected to rise by 77% while the worldwide population is projected to reach 8.9 billion, an increase of 36% [1]. Nevertheless, fossil fuels like coal, hydrocarbons, and natural gas have a limited supply on Earth. Additionally, the development of transport options and the expansion of industrial operations have led to the release of exceptionally high levels of particles and greenhouse gases into the atmosphere. Hence, to counteract the depletion and pollution caused by fossil fuels, it is critical to investigate cleaner and more efficient energy sources. An alternative energy economy based on hydrogen has therefore emerged from the hunt for new energy sources [2]. Hydrogen may be generated using many processing procedures, including thermal, electrolytic, photolytic, and biological methods. The thermal process encompasses natural gas reforming, renewable liquid and bio-oil processing, and the gasification of biomass and coal, while the electrical phase involves the electrolysis of water (H<sub>2</sub>O) utilising external energy sources. In addition, the photocatalytic method, water splitting using sunlight through biological and electrochemical materials [3]. As an alternative sustainable approach, the scientific and industrial communities have once again begun to thoroughly investigate the valorisation of biomass resources into high-value products [4, 5]. The academic community has devoted a great deal of energy over the past 20 years to determining the most efficient methods of converting biomass resources into targeted compounds. Various methods are utilised for this purpose including combustion, [6], pyrolysis [7], gasification [8,9], direct liquefaction [10], and recently several advanced oxidation processes such as Fenton, ozonolysis [11,12], electro-fenton [13] and photocatalysis [14,15]. The photocatalysis an enhanced oxidation process, can be initiated when a photocatalyst is exposed to light irradiation with energy equivalent to or exceeding the photocatalyst's band gap. Electrons transition

from the valence band to the conduction band, resulting in the formation of positive holes. Radicals ( $\cdot\text{OH}$ ) can be formed by the oxidation of water molecules by positive holes in the valence band, whereas ( $\cdot\text{O}_2^-$ ) species can be produced by the reduction of ( $\text{O}_2$ ) in the conduction band. Photocatalytic biomass conversion is a novel alternative method, replete with significant potential and technical challenges [16].

Ultrasound (US) energy has seen a dramatic uptick in its utilisation for the transformation of lignocellulosic biomass into valuable products in the past several years [17]. By causing physical changes on the surface of the biomass, such as de-texturization, fragmentation, and pore creation, the ultrasonication or sonochemical effect provides a physicochemical pretreatment approach. The cavitation bubble bursts, and the free radicals that were generated chemically help dissolve the bonds between the lignin, hemicellulose, and cellulose [18]. This solution overcomes the problems with the old ways that were mentioned before. Another environmentally friendly method that accomplishes reactions by employing created hydroxyl radicals is photocatalysis, which is likewise regarded as a greener approach since it does not require extra chemicals to manufacture free radicals, and enables the cleavage of chemical bonds in the specified compounds after the light absorption and triggering a chemical reaction on the catalyst [19-21].

In this context, the interesting advantages of combining the synergistic effect of photocatalysis and sonication which results in an increase in the production of hydroxyl radicals in the reaction mixture, and the turbulence effect created by ultrasound improves mass transfer between the liquid medium and the surface of the catalyst. A novel methodology integrating sonochemistry with photocatalysis, termed sonophotocatalysis (SP), has emerged as a sophisticated oxidation process and is now under investigation in the field of biomass valorisation [22]. Recent research has shown the use of this synergistic process for the degradation of organic pollutants such as phenol and phenolic compounds [23], the degradation of textile dyes [24], and the treatment of pesticides in wastewater [25]. While the potential uses of sonophotocatalysis surpass water and wastewater treatment, it has been investigated for energy production, including hydrogen generation via water splitting and other biomass sources. This

energy-focused application underscores the promise of sonophotocatalysis in tackling environmental and energy issues, fostering a sustainable and efficient future [26,27]. However, up until this point, many researchers have focused on developing carbon-based materials as a means to target metallic semiconductors in an efficient, environmentally friendly, and cheaper manner [28]. The unique characteristics of carbon materials, including their optical activity, electrical conductivity, chemical stability, mechanical strength, and porous structures, render them exceptional candidates with high activity and stability for sonophotocatalysis [29]. However, several challenges are associated with the use of carbonaceous materials in these catalytic processes [30]. One of the main challenges lies in the low light absorption capacity of carbonaceous materials which will limit their ability to efficiently absorb and use light energy, essential for catalytic reactions in sonocatalysis and photocatalysis. Another challenge lies in the wide energy bandgap of carbonaceous materials which will affect the range of light wavelengths that can be effectively used by carbonaceous materials, thereby further limiting their photocatalytic activity. Additionally, carbonaceous materials often exhibit unsatisfactory physicochemical stability, which can lead to degradation and reduced catalytic activity over time [31].

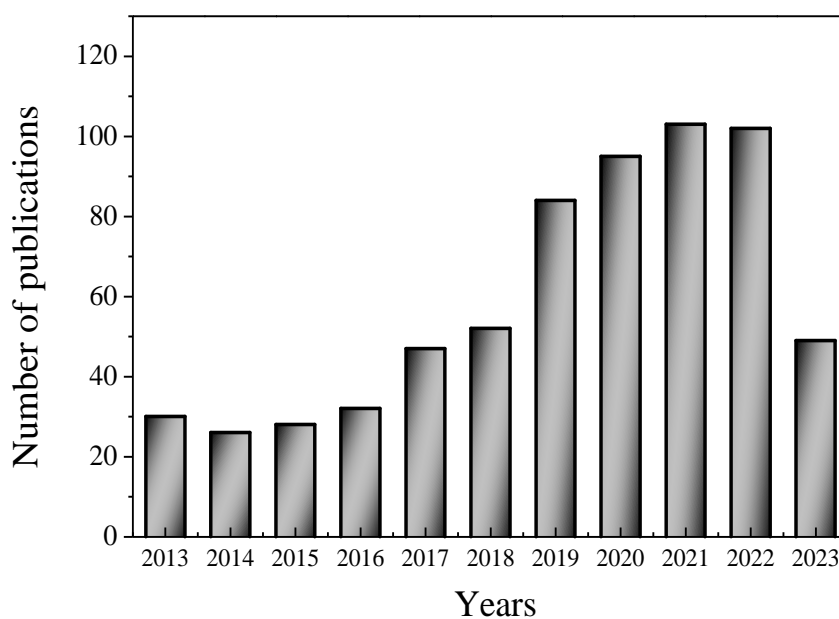
In the present review, we highlight the important results provided by literature studies, especially those from the last ten years, dealing with sonochemistry and ultrasound-assisted hydrogen generation techniques, and discuss the possibility of biomass valorisation as lignocellulosic and different wastes into high-value products and hydrogen production by photocatalytic and sonophotocatalytic means from a mechanistic and technological perspective with comparison to conventional techniques and clarifying the future attitudes. Various aspects of this field have been addressed in the sub-sections. The first section discusses the principle of ultrasonic technique from the principle to its application for green energy with an extensive discussion regarding the complementary impacts of cavitation events and ultrasonic power. Then, highlighting the advantages of hydrogen generation from different solid waste as plastic sources and biomass by heterogeneous photocatalysis and the evolution of carbon-based materials in this area, with the recent application in this field, also describing the potential of ultrasonic application towards this technology. As a potential technology for hydrogen generation, ultrasound-assisted approaches will

eventually be briefly reviewed along with its limitations and difficulties, investigating sonocatalysis, sonoelectrocatalysis, and sonophotocatalysis at a molecular level to shed light on how to integrate sonication with more traditional methods of producing hydrogen. This perspective would effectively push forward the progress of biomass valorization and understanding the mechanism of sonophotocatalytic as a great combination for hydrogen generation and we hope this review can inspire development of more creative work and help clarify some directions for research and development in this specific application area in the future.

## **2. Exploring the potential of sonochemistry as a tool for sustainable energy**

The impact of ultrasonic sound waves was initially noted by British maritime engineers in 1894. The sound waves produced by rotating propellers were referred to as hydrodynamic cavitation. In chemistry, researchers produce these sorts of sound waves by using powerful sound waves from an ultrasound generator with frequencies above 16 kHz [32–34]. Researchers exposed water to ultrasound and observed that the heat from the bubble collapse breaks down water ( $\text{H}_2\text{O}$ ) into very reactive hydrogen atoms and hydroxyl radicals ( $\text{OH}^\cdot$ ). Both atoms came back together to form hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and molecular hydrogen ( $\text{H}_2$ ) during the cooling phase that followed [35]. Within the bubble, localized temperatures and pressures reached 5000 K and 1000 atm, respectively, due to cavity implosion. Under these intense circumstances, it is frequently thought the formation of extremely reactive species including hydroxyl ( $^\cdot\text{OH}$ ), hydrogen ( $^\cdot\text{H}$ ), hydroperoxyl ( $^\cdot\text{HO}_2$ ) radicals, and hydrogen peroxide, occurs [36]. The presence of organic species or other substances in the irradiated water increases the likelihood of subsequent reactions. The degradation of organic substances is accelerated in these types of settings. Until now, the scientific discussion on the efficiencies of hybrid processes has been sparked by the significant progress in combining ultrasound power with various conventional processes, such as electrolysis, photolysis, catalysis, photocatalysis, and electrocatalysis, in relation to ultrasound-assisted pathways for hydrogen generation [37]. The use of sonication in "silent" procedures has been shown to produce encouraging results that might achieve up to 100% enhancement, according to the last ten of

experimental publications on this subject **Fig.1**. Significant achievements have been made in this area over the past 5 years, leading to a doubling of academic publications compared to scientific production before 2018 [38]. Sonophotocatalysis has emerged as a highly promising hybrid process, combining ultrasonic waves and photocatalysis to enhance chemical reactions. Ultrasonic waves generate cavitation bubbles that collapse violently, creating localized extreme conditions of high temperature and pressure, which produce reactive species like hydroxyl radicals ( $\bullet\text{OH}$ ) and hydrogen atoms. Simultaneously, photocatalysis uses light energy to activate semiconductors like titanium dioxide ( $\text{TiO}_2$ ), generating electron-hole pairs to drive redox reactions. The synergy between these processes offers several advantages: ultrasonic waves enhance mass transfer, improve catalyst surface cleaning, and increase reactivity, while photocatalysis accelerates energy conversion and pollutant degradation. This combination maximizes efficiency, accelerates reaction kinetics, and promotes hydrogen production and organic decomposition. As a result, sonophotocatalysis stands out as an innovative, sustainable solution for energy generation and environmental remediation, leveraging the complementary strengths of cavitation and photocatalysis to address pressing global challenges.



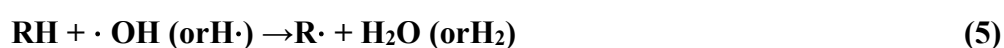
**Fig. 1.** The total number of recent research about ultrasound-assisted photocatalysis Combining "photocatalytic" and "ultrasound" as keyword searches (collected from the Web of Science Core Database).

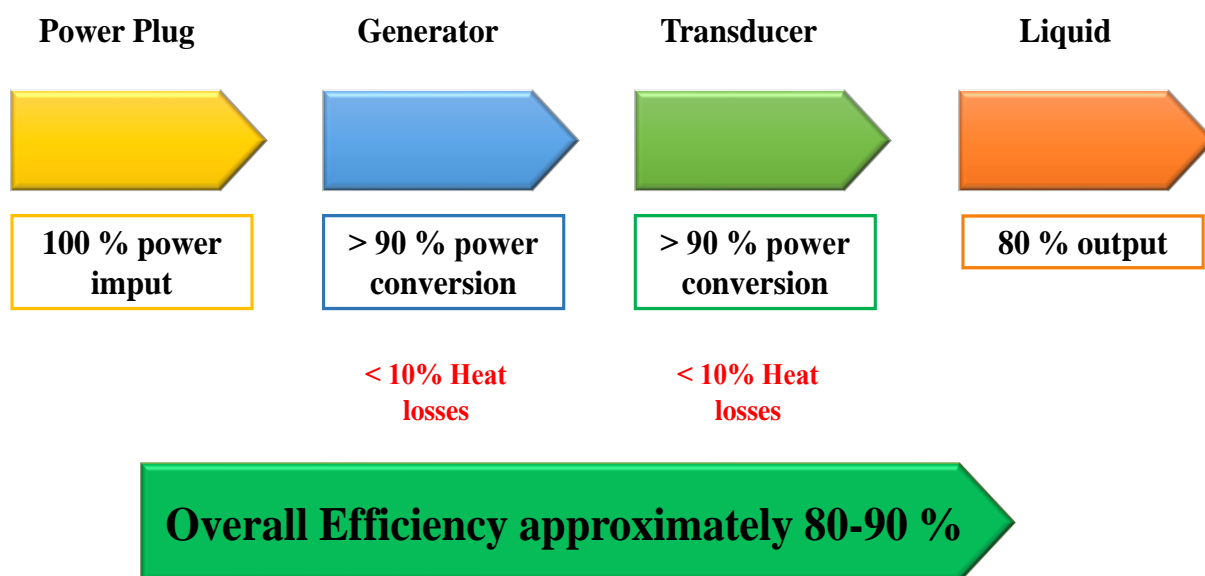


## 2.1. Fundamental concepts in cavitation assisted techniques

### 2.1.1. Cavitation phenomena

Due to the longitudinal pressure wave nature of ultrasound as it passes through various media, the characteristics of each contact are determined by the wavelength and velocity of the beam. The linked effects are not confined to the molecular level [39,40]. Ultrasound, in contrast to other forms of energy like heat and light, can be used to enhance physicochemical reactions by producing extremely transient local temperatures, high pressures, and mechanical stresses [41]. In theory, an effect known as ultrasound-induced cavitation may mediate such a mechanism [42,43]. In chemical reactions that take place in water, the cavitation effect is most prominent; this occurs when the aqueous phase is compressed and then expanded as a result of successive ultrasonic irradiation. Because the ultrasonic liquid's vapor pressure is lower than that of the compressed liquid, microbubbles made of gas and liquid vapour are created [44]. During expansion, the microbubbles quickly enlarge beyond their compressive size, a phenomenon known as microbubble overgrowth. A large amount of the ultrasound's energy is accumulated and built up within a very small volume of microbubbles in this type of cycle. Specific "hot spots" that reach pressures of up to 1000 atm and temperatures of up to 5000 K are created when microbubbles suddenly rupture, releasing tremendous amounts of energy [45]. Depending on the effectiveness of the ultrasound, the percentage of power that reaches the liquid can range from "80% to 90%". This is because there are a series of losses that occur between the power plug and the liquid, which might hinder the catalytic process as illustrated in **Fig.2**, as follows;



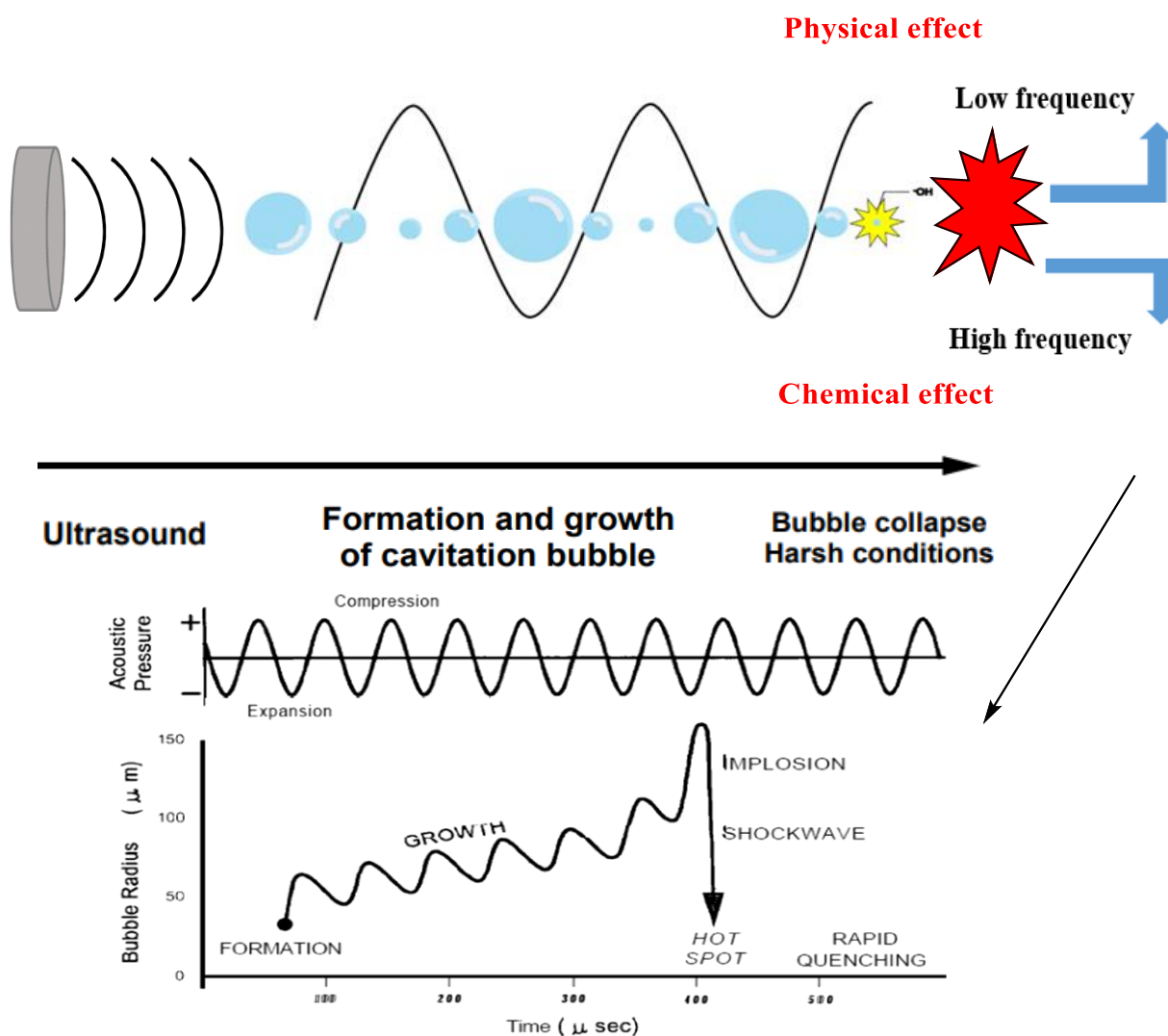


**Fig. 2.** Ultrasound efficiency and overall power attenuation from plug to liquid

### 2.1.2. Ultrasonic power's impact

The impact of ultrasound power on sonophotocatalysis performance is an important consideration for optimizing the process [46]. Ultrasound power is the degree of energy or intensity of the ultrasound waves used for the treatment. To maximize catalyst-pollutant interaction, ultrasound is an essential tool in sonophotocatalysis because it helps disperse the catalyst and improves mass transfer [47,48]. Furthermore, more power means more energy and stronger acoustic cavitation, which means more reactive species like hydroxyl radicals ( $\cdot\text{OH}$ ) are generated. Efficient and rapid degradation of pollutants can be achieved by the oxidation and breakdown of these reactive species [49]. For example, they looked at how different levels of ultrasound affected the sonophotocatalytic degradation of sulfadiazine (SDZ) onto a composite of Zn-Cu-Mg mixed metal hydroxide (MMH) @  $g\text{-C}_3\text{N}_4$  @ microfibrillated carboxymethyl cellulose (MFC3) [48]. Degradation accelerated with rising power values, because more cavitation microbubbles were generated, which in turn collapsed and produced more reactive oxygen species (ROS) [48]. In a similar vein, the sonophotocatalytic degradation of AB dye [50] dye onto ZnO/persulfate composites was studied in relation to ultrasound power (100-300  $\text{W L}^{-1}$ ). Increasing the ultrasound power resulted in better cavitation formation and the generation

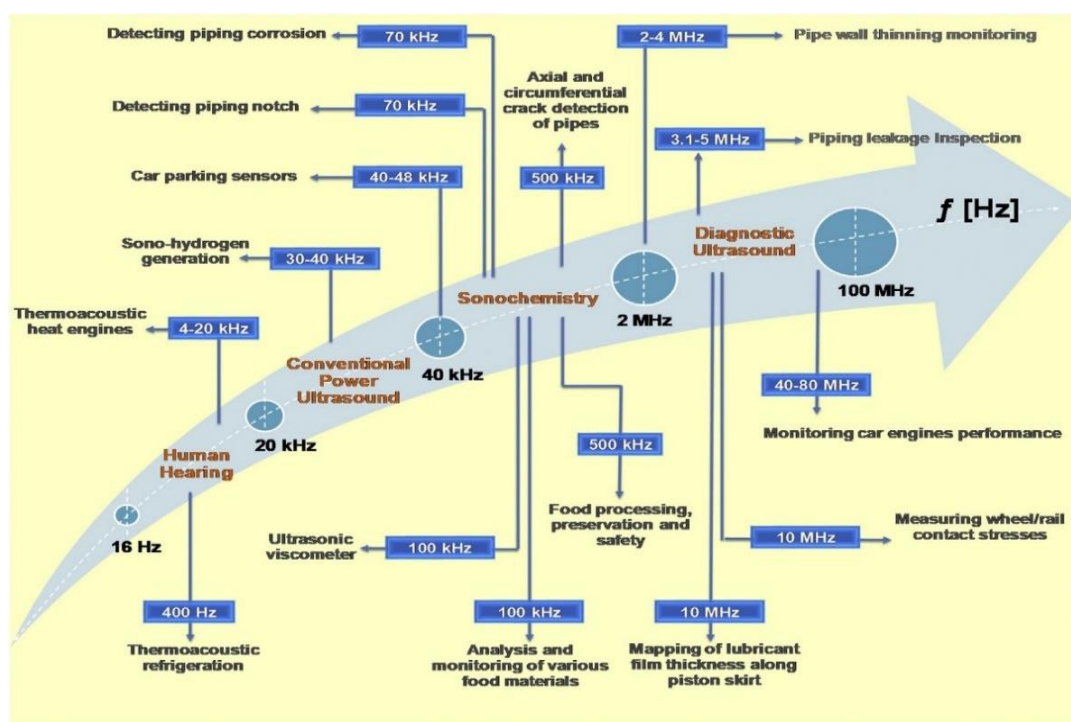
of more ROS, leading to higher degradation performance of the dye [51]. Several others have made the same observation. The most important thing is to determine how much ultrasonic power to use so that degradation performance is maximized. A small power output might not be enough to cause cavitation phenomena to produce enough reactive species. As a result, degradation rates are reduced and pollutant removal efficiency is diminished. However, problems like catalyst deactivation or damage might result from ultrasonic power levels that are too high. The mechanism of ultrasonic cavitation and the subsequent creation of "hot spots" is shown schematically in **Fig. 3**.



**Fig. 3.** Scheme of the acoustic cavitation phenomenon. Information on transient cavitation is taken from Ref insert [52]

### 3. The potential applications of ultrasonic technology

Various sectors can benefit from ultrasound's practical applications **Fig 4**, such as in the hardening of immersed metals [53], the delivery of drugs and other therapeutic applications [54], the improvement of electrospinning [55], the treatment of bladder cancer [54], and the acceleration of chemical reactions and processes [56]. In order to drive and improve chemical processes and yields, irradiation and ultrasonic waves are linked to efficient physical and chemical factors. The use of less energy-intensive and potentially harmful chemicals and solvents is the overarching goal, together with the utilization of ultrasound. Among its many advantages beyond sonochemistry is its ability to improve electrochemical diffusion processes, among others. The use of ultrasonic waves can both facilitate more efficient chemical reactions and create an altogether novel chemical environment. For instance, ultrasonication significantly enhances organic syntheses. Mason authored a thorough analysis of ultrasound's role in synthetic organic chemistry, with an emphasis on its organic synthesis functions [55].



**Fig. 4.** An overview of ultrasound uses at various ultrasonic frequencies. Reproduced with authorization from Reference [55]. Copyright 2019 Elsevier

A number of other researchers have also used ultrasound to successfully carry out synthetic organic reactions; for example, Bang and Suslick [57] and Cravotto and Cintas [58] are noteworthy examples. Nanomaterial production, water purification, metal corrosion, polymeric membrane cleaning, food processing, cavitation bubble dynamics, and hydrogen production are all areas of concentrated study. An extensive analysis of ultrasound's uses in water and wastewater treatment was carried out by Chen [59].

**Table 1** provides a synopsis of the various fields of study that have made use of sonochemistry in recent years, along with a brief explanation of each field, a list of relevant references (both current and historical), and an overview of the research area (where applicable).

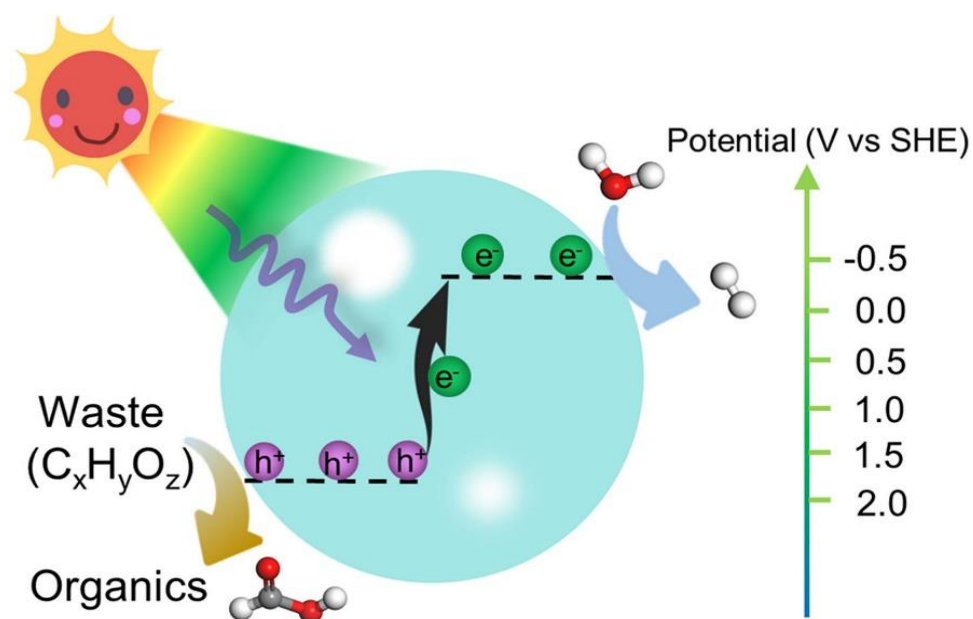
**Table 1-** Overview of the current research environment in sonochemistry

Area of research	Description	References
<b>Organic syntheses</b>	The ultrasound-assisted synthetic organic chemistry	[60,61]
<b>Production of nanomaterials</b>	Ultrasound technology is employed for the synthesis of nanomaterials by pulsed sonoelectrochemistry.	[62,63]
<b>Environmental treatment</b>	The superior oxidation processes make it useful for treating water and wastewater as well as smells and muck.	[64,65]
<b>Water disinfection or purifying water</b>	Ultrasound is also employed in the water purification process.	[66-68]
<b>Corrosion of metals</b>	Electrochemical studies of the coatings' corrosion behaviour on various metals	[69-71]
<b>Cleaning of Polymeric membranes</b>	Ultrasound waves are used for the process of cleaning of polymeric membranes	[72,73]
<b>Ultrasound in food processing</b>	Ultrasound is promising for food processing	[74-77]
<b>Cavitation bubble dynamics</b>	The sonoelectrochemistry technique investigates cavitation bubble dynamics and flow velocities.	[78-82]
<b>Ultrasound in separation</b>	Recently, high-frequency ultrasonic standing waves have been used to separate droplets or cells from biomass at the liter to industrial scale.	[83,84]
<b>Biomass valorization and transformation</b>	Recent advances in ultrasound-assisted catalysis provide novel techniques to enhance biomass and waste.	[85-88]

#### 4. Hydrogen generation from different solid waste sources and biomass

Usually, the process of heterogeneous photocatalysis can be explained by crucial steps illustrated in **Fig. 5**. In the process, a photon with sufficient energy is absorbed by a semiconductor, resulting in

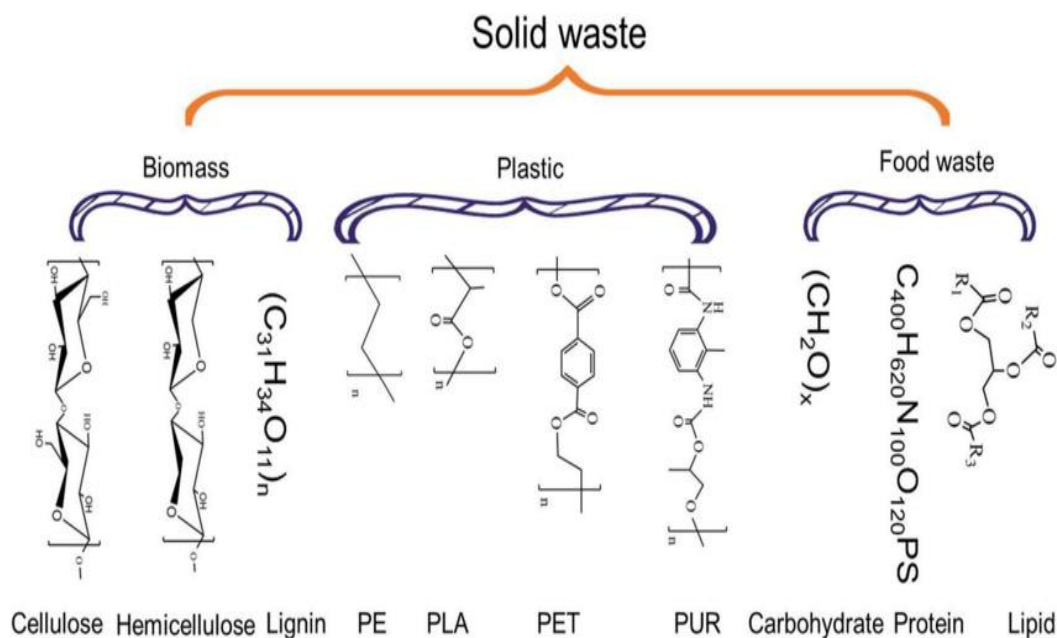
the creation of electron-hole pairs. These electrons and holes then separate from each other. They may either recombine on the surface or get trapped by defects before recombining in the catalyst. Finally, the electrons and holes, acting as strong reducing agents and strong oxidants respectively, either directly react with substrates or indirectly participate in reactions by generating reactive oxygen species (ROS) [89]. It is worth mentioning that the oxidation and reduction abilities of photogenerated holes and electrons primarily rely on the edge positions of the valence band (VB) and conduction band (CB), respectively. For an overall photocatalytic reaction to take place, it is crucial that the energy levels of the VBM and the CBM align with the redox potential of oxidation and reduction half- reactions. With their remarkable redox potential, the highly reactive ROS have the ability to oxidize a wide range of organic substrates. As an example, ( $\cdot\text{OH}$ ) hydroxyl radical, (redox potential +2.81 V vs. standard hydrogen electrode, SHE), plays a crucial role in photodegradation reactions. It is generated through the oxidation of surface hydroxyls or absorbed water. Additional reactive oxygen species can be generated during the process described above, such as the superoxide radical ( $\cdot\text{O}_2^-$ , redox potential: +0.89 V vs. SHE), singlet molecular oxygen ( $^1\text{O}_2$ ), and  $\text{O}_3$  species.



**Fig .5.** Photocatalytic reaction of solid waste using semiconductors: a schematic. Inspired from Ref [95] and used with permission. Copyright 2022 Wiley Online Library.

Kawai et al. contributed to hydrogen generation method in 1981 using a photocatalyst of Pt/TiO<sub>2</sub> with substrates of microalgae, seaweed, rice straw, and grass. The rate of hydrogen production might reach 0.11 mmol g<sup>-1</sup> h<sup>-1</sup> when seaweed was utilized as a substrate [90]. Water does not dissolve unprocessed lignocellulosic compounds. So, to get more hydrogen out of the process faster, we need to make sure that active species like photo-generated holes and hydroxyl radicals may fully interact with the substratum by increasing the contact area between the catalyst and lignocellulosic materials. Jaswal et al. [91] confirmed that the hydrolysis products of the original lignocellulosic materials are appropriate hydrogen substrates by employing pine hydrolysate as a substrate and achieving a hydrogen production of 19.9 mL/g using a photocatalyst of 1% Pt/TiO<sub>2</sub>. H<sub>2</sub> was produced from cellulose, hemicellulose, and lignin by Wakerley et al. using photocatalysis in an alkaline water solution by synthesized CdS/CdO quantum dots (QD) with an average particle size of less than 10 nm

**Table 2.** The original lignocellulosic materials are fully contacted by the QD because to their small particle size, resulting in a huge contact area. On top of that, the photo-corrosion is mitigated and the HER is assisted by the CdO layer that coats the surface of the CdS nanospheres. Under the influence of visible light, the highest HER of 5.3 mmol g<sup>-1</sup> h<sup>-1</sup> was attained [92]. In addition, under visible light, this system functioned well for six days and could convert untreated lignocellulosic materials like wood and paper using solar energy at ambient temperature. The result is a low-cost method for oxidizing solid waste (biomass, plastic, food waste) **Fig.6** and driving proton reduction to create hydrogen gas in water.



**Fig.6.** Schematic depicting the classification of solid waste.

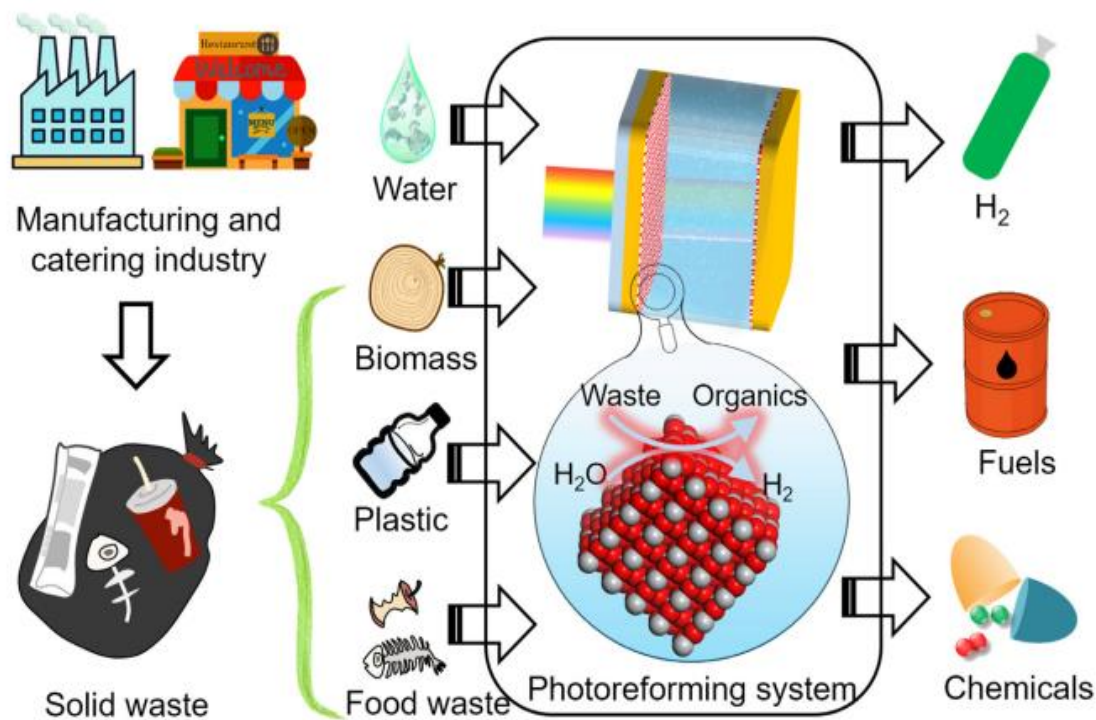
When compared to conventional fuels such as coal, gas, and petroleum, hydrogen energy offers several inherent advantages. Firstly, hydrogen is abundantly available in nature, as it is found in water, biomass, and waste plastics. Secondly, it is a high-quality energy source, second only to nuclear power; and thirdly, it produces zero emissions during reactions, with the exception of water, as combustion does not release carbon. So, it's a great clean energy option for getting to zero emissions. Solar energy is generally acknowledged as the undisputed leader in sustainable energy generation. The energy base supplied by solar radiation on earth significantly outweighs that of all other renewable energy sources combined. Solar-powered hydrogen generation is considered an efficient and occasionally renewable approach for converting energy and acquisition. Many research investigations have been conducted on solar-driven water splitting for hydrogen production for more than 20 years, and the process has shown some benefits [93-97]. A potential alternative to water oxidation that is rich, cheap, and pure has been investigated by researchers to tackle this issue [98]. Many biomass structures possess a significant concentration of protons, rendering them efficient transporters of  $H_2$ . Moreover, biomass may be oxidized with reduced bias, potentially yielding economic advantages from its oxidized byproducts [99]. As a result, the development of a circular



bio-economy centered on high-value, pollution-free hydrogen energy holds great potential to address the global energy crisis and environmental pollution. This approach leverages the abundant solar energy and the vast quantities of biomass waste produced worldwide (**Fig. 7**) [100].

**Table 2-** Photocatalytic hydrogen generation during the biomass reforming process.

Photocatalyst	Solvent	Substrate	Light source (mmol $g^{-1}$ cat h $^{-1}$ )	Atm	Time (h)	H <sub>2</sub> generation	Ref
CdS/CdO <sub>x</sub> (0.5 μM)	KOH solution(25°C) with Co (BF <sub>4</sub> ) <sub>2</sub>	Wooden branch	100mW cm <sup>-2</sup>	N <sub>2</sub> and 2% CH <sub>4</sub>	24	5.59	[92]
mpg-C <sub>3</sub> N <sub>4</sub>	CH <sub>3</sub> CN solution	Lignin	100 mW cm <sup>-2</sup>	O <sub>2</sub>	10	1.512	[101]
0.2-NiS/CdS (100 mg)	Aqueous solution	Lignin and lactic acid	455 nm LED 6W Xe lamp (300 W)	Vacuum	3	0.015	[102]
Activated <sup>NCN</sup> CN <sub>x</sub> (5 mg)	KOH solution (25° C)	Lignin	Xe lamp (100 mW cm <sup>-2</sup> )	N <sub>2</sub> and 2% CH <sub>4</sub>	288	23.52	[103]
TiO <sub>2</sub> -NiO (0.50)	Aqueous solution	Lignin	UV-Vis	Vacuum	5	6.60	[104]



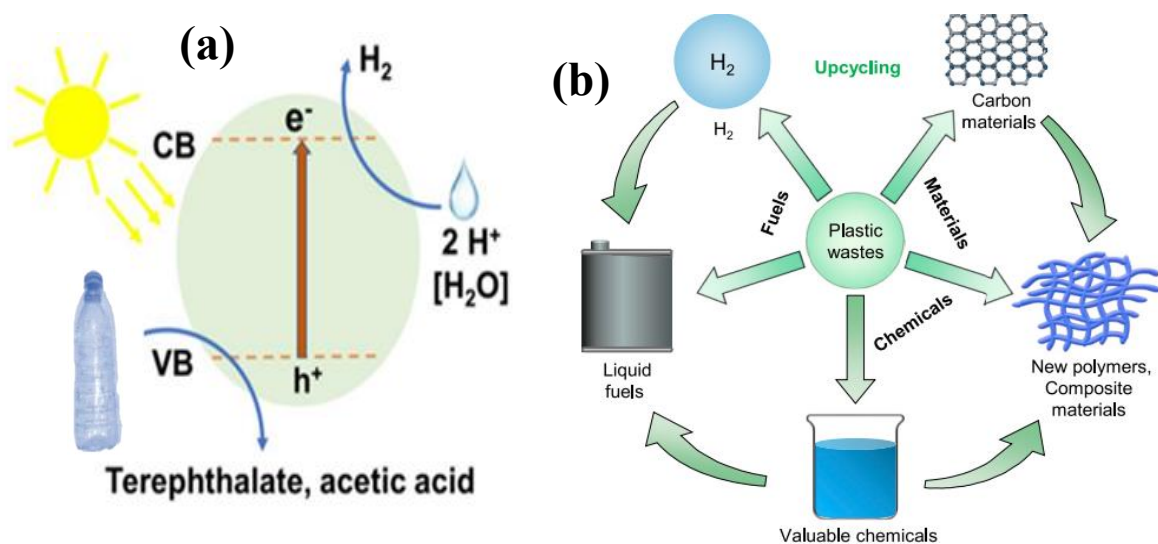
**Fig. 7.** Conversion of biomass feedstocks into compounds with added value and hydrogen production using photocatalytic reforming. Adapted with permission from Ref [100]. Copyright 2022 Wiley Online Library.

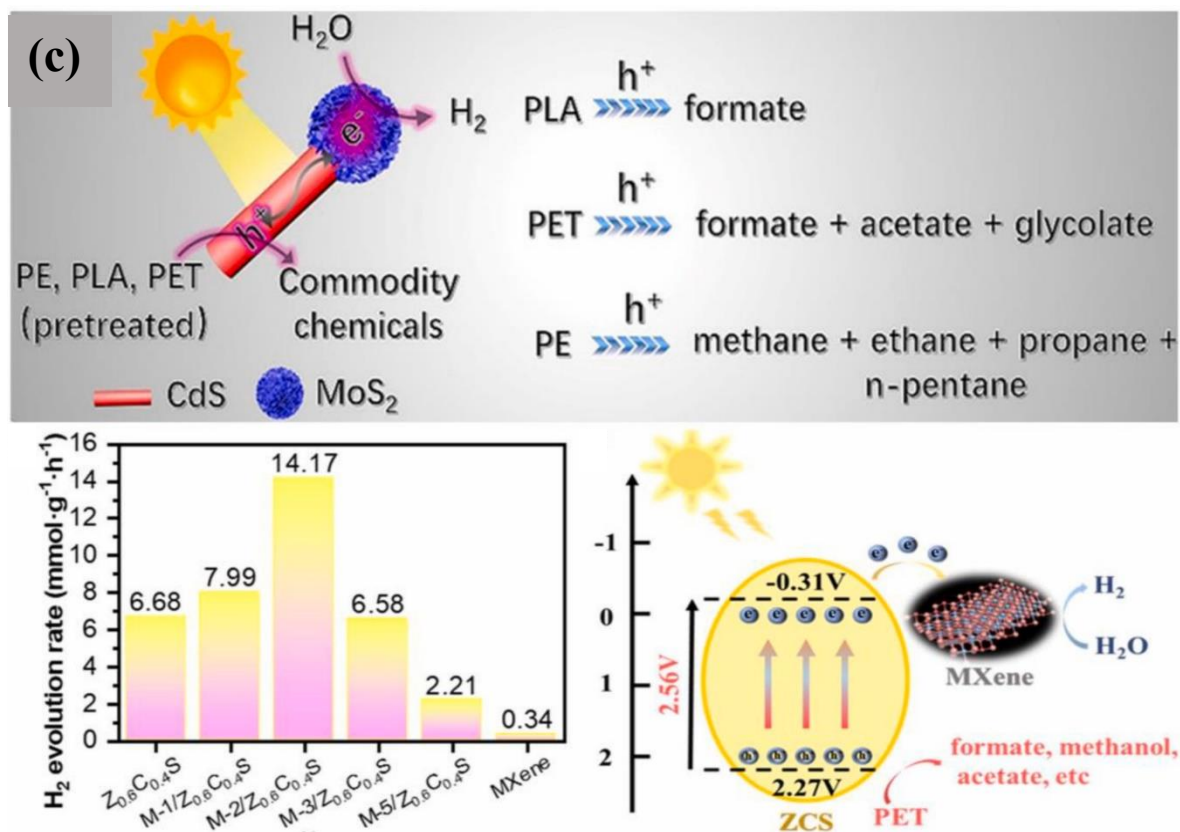
Actually, plastics also serve as sacrificial agents in conventional photocatalysis systems, where photogenerated holes oxidize them in whole or in part. Hole scavengers made of plastic polymers can enhance hydrogen generation and cogenerate value-added compounds at a lower cost than traditional, more expensive sacrificial reagents like triethylamine and methanol. **Fig. 8a**. However, the relatively strong C–C bonds present in hydrocarbon-chain-based plastics (non-polar polymers) pose challenges for photo-reforming processes [105]. For instance, research created a MoS<sub>2</sub>/CdS hybrid for the photo-reforming of pre-treated polyethylene (PE), polyethylene terephthalate (PET), and polylactic acid (PLA). Additionally, the MXene/ZnxCd<sub>1-x</sub>S composite photocatalysts exhibit superior activity for PET reforming compared to the individual components, with hydrogen generation being significantly dependent on the composition, as illustrated in **Fig. 8b** and **Fig. 8c**. Conversely, plastics that incorporate oxygenated, polar groups or esters (polar polymers), such as PET, PUR, and PLA, serve as more suitable feedstocks. Evidently, oxidative species are clearly generated during photocatalysis, resulting in low molecular fuels. The conversion of polylactic acid (PLA), PET, and polyurethane (PUR) to fuels by CdS/CdOx photocatalysis under sun irradiation was investigated. The H<sub>2</sub> product constituted 38.8%, 16.6%, and 22.5%, respectively [106]. Additionally, a two-step conversion route of C–C bond cleavage and coupling was designed via Nb<sub>2</sub>O<sub>5</sub> photocatalysis. PE, PP and PVC were degraded to CO<sub>2</sub>, which was further photo-reduced to CH<sub>3</sub>COOH (<40.0%) [107]. However, current photocatalysts face limitations in CO<sub>2</sub> reduction, resulting in low yields of multi-carbon (C<sub>2</sub>) fuels. The efficient conversion of plastic waste into multi-carbon fuels at room temperature and pressure requires rational photocatalyst design and optimization of C–C bond breaking and coupling mechanisms (Fig. 8b). Additionally, performed PET and PLA conversion using carbon nitride/nickel phosphide (CNX=Ni<sub>2</sub>P) photocatalysis in alkaline conditions [108-110]. Consequently, acetate and formate were produced, which may be utilized as fuels following the revisions in **Table 3**.

**Table 3-** fuel production from plastics using a sophisticated oxidation method

Type of plastics	Technology	System	Conditions	Products	Yield	Refs
LDPE	Photocatalysis	Polyacrylamide grafted TiO <sub>2</sub>	UV light	CO <sub>2</sub>	/	[111]
PET	Photocatalysis	Carbon Nitride/Nickel Phosphide	Solar light, T= 25°C	H <sub>2</sub>	77.2 μmol/g	[112]
PET	Photocatalysis	CdS/CdO <sub>x</sub>	Visible light	H <sub>2</sub>	278.2 mmol/g	[113]
LDPE	Photocatalysis	ZnO	UV light	CO <sub>2</sub>	/	[114]
PVC	Fenton	FeCl <sub>3</sub> , H <sub>2</sub> O <sub>2</sub>	T= 50 °C, C/S Ratio= 8.0	C <sub>1</sub> -C <sub>2</sub> acids	82.5 %	[115]
LDPE	Fenton	FeCl <sub>3</sub> , H <sub>2</sub> O <sub>2</sub>	Chlorosulfuric acid = 2.5 %, No light	C <sub>1</sub> -C <sub>2</sub> acids	62.4 %	[116]
PE	Fenton	FeCl <sub>3</sub> , H <sub>2</sub> O <sub>2</sub>	UV-Vis light, pH = 2.5 Time = 2 h	CO <sub>2</sub>	> 99%	[117]
PS	Photo-Fenton	FeCl <sub>3</sub> , H <sub>2</sub> O <sub>2</sub>	UV light, Time = 300 min	CO <sub>2</sub>	89.0 %	[118]
PVA	Electrocatalysis	H <sub>3</sub> PO <sub>4</sub>	T= 100 °C, External voltage = 0.55 V	H <sub>2</sub>	9.5 μmol/min	[100]
PVC	Electrocatalysis	TiO <sub>2</sub> /graphite cathode	T= 100 °C, pH= 3.0, Oxygen flow = 40 ml/min, Time = 6h, Applied potential = -0.7 V	Carboxylic acid	75.0 %	[119]
PP	Solar Thermo-coupled electrocatalysis	NaOH, KOH	T= 350 °C, Time = 60 min, External voltage = 1.5 V	Gas	33.1 %	[120]

The abbreviations LDPE, PET, PVC, PE, PS, and PVA stand for "low density polyethylene," "polyethylene terephthalate," "polyethylene," "polystyrene," and "polyvinyl alcohol," respectively. polypropylene (PP)





**Fig. 8.** (a) Diagrams illustrating photoreforming for concurrent photocatalytic plastic the degradation and hydrogen generation (b) Upcycling to create value-added items. (c) Schematic representation of the photoreforming of pretreated polymers utilizing a CdS/MoS<sub>2</sub> photocatalyst (PLA: polylactic acid, PET: polyethylene terephthalate, PE: polyethylene). Reproduced with authorization from Reference [106]. Copyright 2022 Elsevier

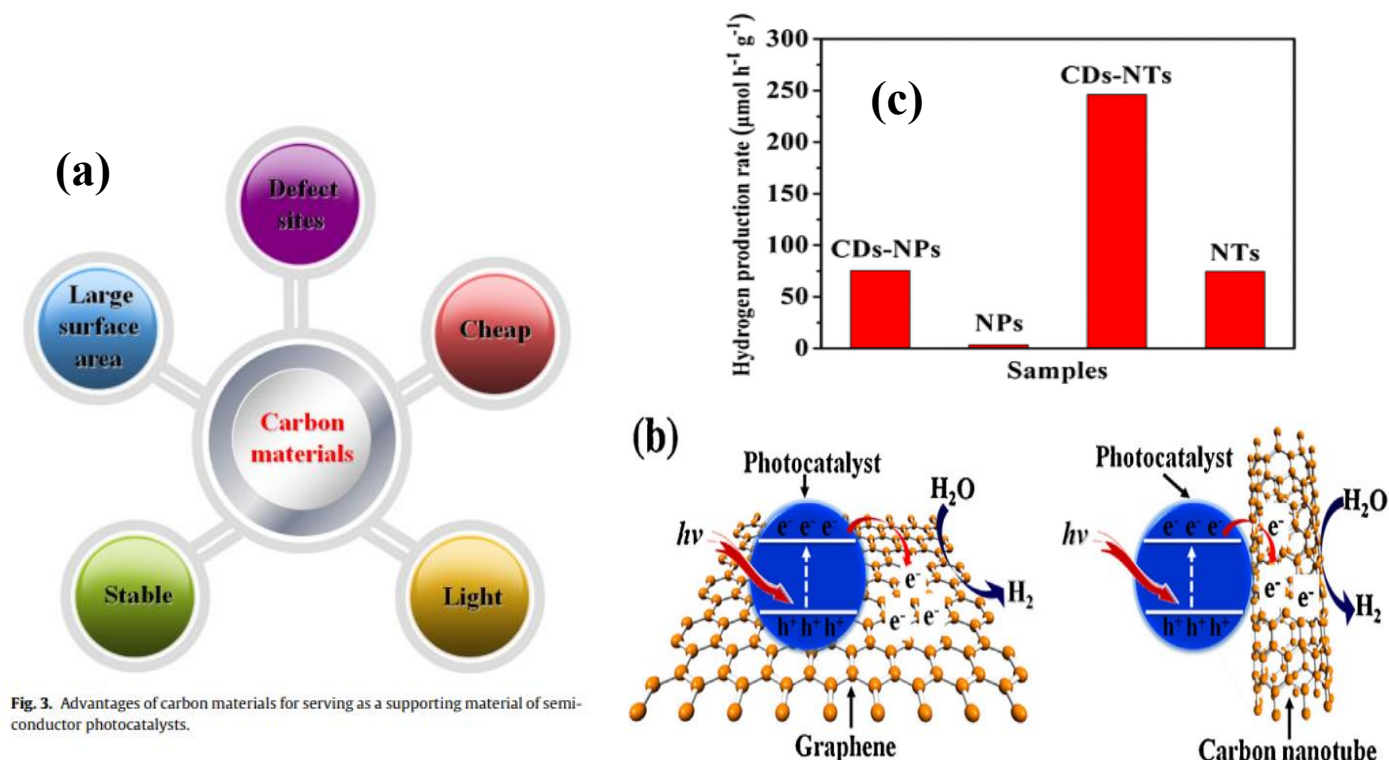
## 5. Development of carbon-based materials for sustainable energy (hydrogen generation)

The use of **carbon materials** as anchors for tiny semiconductor nanoparticles in photocatalysis has become a common practice due to their numerous advantages (**Fig. 9a**) [121–128]. First, carbon materials provide a large surface area framework that facilitates the distribution and immobilization of nanoparticles. For instance, Shen et al. [129] demonstrated the use of reduced graphene oxide (rGO) to stabilize ultra-thin Zn<sub>0.5</sub>Cd<sub>0.5</sub>S nanorods, significantly enhancing photocatalytic hydrogen (H<sub>2</sub>) production. Second, these carbon compounds can easily pair with nanoparticles while retaining their structure and properties due to their heat stability and chemical inertness. Carbon materials can

enhance water molecule adsorption and dissociation, according to theoretical research [130-132], which is good for hydrogen generation. Substantial surface functional groups, like COOH and OH, on carbon materials with modified surfaces, in particular, can serve as anchoring sites for the reagents, creating more reaction centers and improving the photocatalytic H<sub>2</sub>-production activity [133-137]. A notable example of this approach is the graphene-supported TiO<sub>2</sub> photocatalyst developed by Zhang et al. [138] using a sol-gel approach and a subsequent calcination procedure at 450 °C, starting with tetra butyl titanate and graphite oxide as precursors. The XRD results demonstrated that, regardless of the graphene content, all samples containing TiO<sub>2</sub> (anatase) exhibited an average crystal size of approximately 11 nm. Notably, the specific surface areas of pure TiO<sub>2</sub> and modified with 5 wt% graphene rose with increasing graphene content, measuring 15.9 and 24.1 m<sup>2</sup> g<sup>-1</sup>, respectively. Consequently, the hydrogen evolution rate of the 5 wt% graphene modified TiO<sub>2</sub> was 8.6 μmol h<sup>-1</sup>, which was 1.9 times more than that of P25 (4.5 μmol h<sup>-1</sup>). Of course, other factors also contributed to TiO<sub>2</sub>/graphene's increased photocatalytic activity in addition to its bigger surface area. Third, defect sites and oxygen-containing functional groups on carbon materials serve as nucleation sites, facilitating the uniform development and anchoring of nanoparticles. These properties address one of the primary challenges in photocatalysis: the rapid recombination of photogenerated electrons and holes. Carbon materials, due to their high conductivity, can capture photogenerated electrons from the conduction bands (CBs) of semiconductors or the lowest unoccupied molecular orbitals (LUMO) of dyes [139–149]. The reason behind this is that carbon materials like graphene and CNTs have a high work function, but the coupled semiconductors' or coupled dyes' CB is more negative than the work functions of graphene and CNTs, as shown in **Figure 9b**, with a value of approximately -0.30 eV vs. NHE [150] and -0.08 eV vs. NHE [151], respectively. The collected electrons either build up on carbon materials' surfaces or move quickly to reactive locations throughout their conductive surfaces. Carbon materials also have a low specific weight, which is great for structural purposes. Natural vegetables like guava, red pepper, peas, and spinach were used to make carbon dots by Wang et al. [152]. For four hours, these veggies were subjected to hydrothermal treatment at 180 °C. An

additional hydrothermal procedure was used to load the generated carbon dots (CDs) onto TiO<sub>2</sub> nanoparticles (NPs) and nanotubes (NTs).

This photocatalytic H<sub>2</sub>-production activity was clearly higher in the carbon dot-modified TiO<sub>2</sub> nanostructures than in the unmodified form, especially when irradiated to a 300W Xe lamp, the optimum carbon dot-modified TiO<sub>2</sub> nanoparticles and nanotubes exhibited H<sub>2</sub>-production rates of 75.5 and 246.1 μmol h<sup>-1</sup> g<sup>-1</sup>, respectively, which are 21.6 and 3.3 times more than those of pure TiO<sub>2</sub> nanoparticles and nanotubes (**Fig. 9c**). The key factor for the performance increase of carbon dot-modified TiO<sub>2</sub> nanostructures was the exceptional electron transport capability of carbon dots. Finally, carbon materials are widely available and can be mass-produced at low cost. Resources such as activated carbon, carbon fibers, CNTs, and graphene are readily accessible and scalable. These materials influence the nucleation and growth of semiconductor nanoparticles, reducing aggregation and improving structural stability [153–156].



**Fig. 9.** Advantages of carbon materials for serving as a supporting material of semiconductor photocatalysts.

**Fig. 9.** (a) Advantages of carbon materials for serving as a supporting material of semiconductor photocatalysts. (b) Comparison of work function of CNTs and graphene with the CB of several typical semiconductor photocatalysts. illustration of the role of graphene and carbon nanotube as electron acceptor and transport channel in photocatalytic H<sub>2</sub> production. (c) Comparison of photocatalytic H<sub>2</sub>-production activity

over carbon dot modified TiO<sub>2</sub> nanostructures. Adapted with permission from Ref [157]. Copyright 2016 Elsevier

## 6. Short overview of ultrasound-assisted different catalytic process for hydrogen production findings and limits

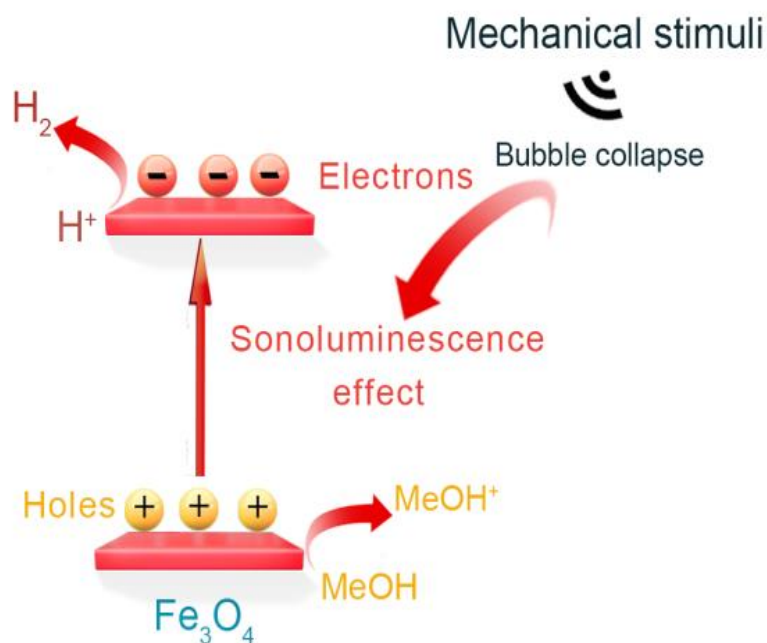
The integration of ultrasound into clean hydrogen synthesis processes has emerged as a promising and efficient technique, particularly when water is used as the hydrogen source. Studies have demonstrated that combining ultrasound with other methods such as catalysis [158], photocatalysis [159], sludge digestion [160], and anaerobic fermentation [161] of wastewater significantly enhances hydrogen production compared to each method applied individually.

Harada [162] investigated the production of hydrogen from water using photocatalysis aided by ultrasound with an alternating irradiation technique. Ultrasound and light were sequentially irradiated in this approach. Sono photocatalysis was employed for the purpose of extracting hydrogen from seawater. Hydrogen synthesis by water sonolysis does not arise from the interaction between acoustic waves and water, but rather results from acoustic cavitation. It is widely accepted that when pure water is subjected to sonication, the major products are H<sub>2</sub> (hydrogen gas) and H<sub>2</sub>O<sub>2</sub> (hydrogen peroxide), with a ratio of approximately 1.25 (H<sub>2</sub>:H<sub>2</sub>O<sub>2</sub>) [163]. The intense temperatures and pressures generated during cavitation lead to the formation and dispersion of reactive radical species both within and outside the bubbles [164,165]. Notably, water sonolysis generates hydrogen at a rate of 10–15 micromoles per minute [166], which is up to 500 times higher than the rate achieved using photocatalysis alone (approximately 0.035 μM/min) [167].

In addition to hydrogen generation, ultrasound has proven highly effective for degrading contaminants via sonocatalytic degradation, where ultrasonic waves penetrate pollutants during the reaction process. This method offers both efficiency and practicality in environmental applications (Xu et al., 2023a) [168,169].

Zhang et al. [170] concluded that the method of H<sub>2</sub> synthesis from magnetic Fe<sub>3</sub>O<sub>4</sub> via Sonocatalysis, illustrated in **Fig. 10**, elucidates the complex interaction of several physical and

chemical processes involved in this phenomenon. The process begins through sonocatalytic water splitting, enabled by the  $\text{Fe}_3\text{O}_4$  catalyst, whereby  $\text{H}_2\text{O}$  serves as the reactant and MeOH functions as the sacrificial agent. The activation of  $\text{Fe}_3\text{O}_4$  by ultrasonic vibrations is driven by ultrasonic cavitation, where the rapid oscillation, expansion, and collapse of microbubbles create localized regions of intense heat and pressure [171–173]. The sonoluminescence effect adds to this setting by illuminating the space between the valence and conduction bands of  $\text{Fe}_3\text{O}_4$  caused by the release of energy and light from the burst bubbles. Oxygen atoms usually react with sacrificial agents to make oxidation products, while this electron excitation primes the catalyst for activity, allowing the reduction of hydrogen ions ( $\text{H}^+$ ) to form  $\text{H}_2$ . Crucial to this procedure is the sacrificial agent MeOH. In order to ensure continuous  $\text{H}_2$  generation, it is oxidized during the reaction, consuming the holes created during water splitting and preventing them from recombining with electrons to provide additional reactive electrons to the system. Usually, this oxidation process produces formaldehyde, formic acid, and other oxidation products [174–179].



**Fig. 10.** The illustration of sonocatalytic  $\text{H}_2$  production process over  $\text{Fe}_3\text{O}_4$  adapted with permission from Ref [170]. Copyright 2024 MDPI

Even so, the plate and ultrasonic probe can be submerged in the electrolyte simultaneously or kept apart. Whenever an electrolyte is isolated from an ultrasonic probe or plate, an inner electrochemical



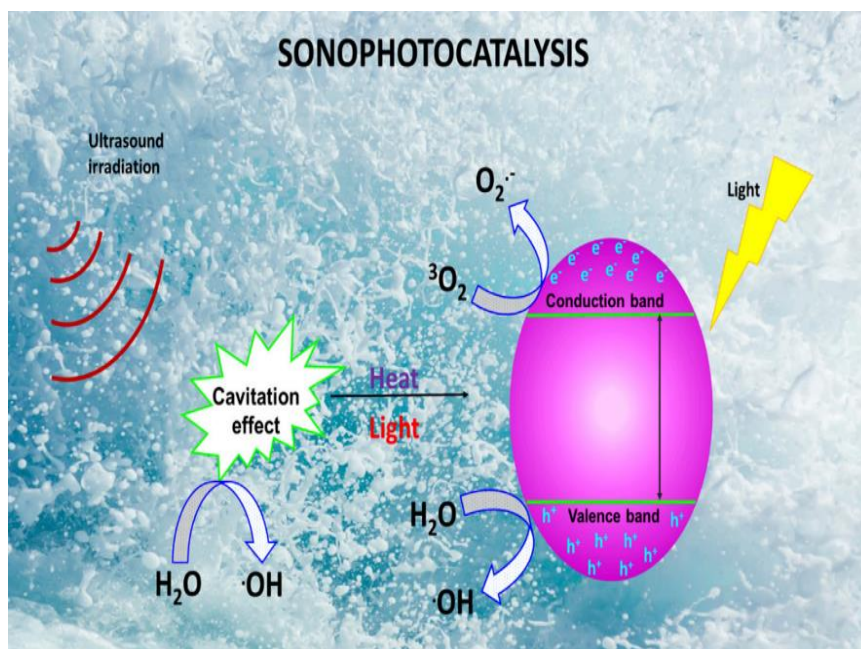
cell is typically employed; in both instances, the "face-on" geometry dictates that the ultrasonic-emitting source should be directed towards the surface of the working electrode. Walton et al. [180] investigated hydrogen evolution in a 1 M H<sub>2</sub>SO<sub>4</sub> solution at a platinized platinum electrode under 38 kHz ultrasonic insonation. Compared to silent conditions, they observed a 2.1-fold increase in current due to the application of ultrasonics. Interestingly, despite the reversible nature of proton reduction at platinized platinum electrodes, the increase in current was not attributed to improved diffusion of H<sup>+</sup> ions. Instead, the enhanced current resulted from the removal of hydrogen bubbles from the electrode surface, improving the efficiency of the reaction. In a similar study, McMurray et al. [181] demonstrated that ultrasonic waves (20 kHz, 26 W/m<sup>2</sup>) significantly accelerated both the oxygen reduction reaction (ORR) and the HER at a vibrating titanium electrode. Their findings highlighted those ultrasonics enhanced mass transport and activated an activation-controlled **process**, making both reactions more efficient.

The chemical breakdown of environmental contaminants through photoacceleration of semiconductors in the presence of ultrasonic irradiation is known as the sonophotocatalysis process [182-185]. Joseph et al. [186] classified the various AOPs as follows: (i) Photolysis; (ii) Photocatalysis; (iii) Sonolysis; (iv) Sonocatalysis; and (v) Sonophotolysis. The name sonophotocatalysis encompasses all of these processes. The degradation of environmental pollutants is achieved through sonophotocatalysis reactions, which are produced when these AOPs are combined. This process involves acoustic cavitation, in which ultrasound and light are passed through a semiconductor that exists in an aqueous media.

The activation of the band gap in the semiconductor and the generation of sonic bubbles during sonophotocatalytic degradation are influencing the sonochemical processes (**Fig. 11**). The primary objective of integrating these two AOPs is to minimize both operational costs and the duration of the deterioration process. Consequently, sonophotocatalysis is anticipated to be more energetically and economically efficient, as can be determined using Equation (1).

$$\text{Synergy} = \frac{K_{\text{sonocatalysis}} + K_{\text{photocatalysis}}}{K_{\text{sonophotocatalysis}}} \quad (1)$$

where  $k_{UV}$   $\neq$   $k_{US}$ ,  $k_{UV}$  and  $k_{US}$  standing for the sonophotocatalytic, photocatalytic, and sonocatalytic rate constants, respectively



**Fig. 11.** Schematic overview of a sonophotocatalytic treatment process.

With the use of a powdered  $\text{TiO}_2$  catalyst and ultrasonic irradiation at 200 kHz and 200 W, Harada [187] initiated the sonophotocatalytic breakdown of water for hydrogen generation. In comparison to sonolysis ( $0.796 \text{ cm}^3/\text{h}$ ) and sonophotocatalysis ( $1.086 \text{ cm}^3/\text{h}$ ), the hybrid method reduced the amount of hydrogen produced by 26.68%. Using a variety of catalysts (metallic, semiconductor photocatalyst, mixed oxide, transition metal oxide, or insulator material), Sasikala et al. [188] replicated a comparable experiment in an aqueous methanol solution. Increased cavitation bubble production in the presence of suspended materials and efficient methanol scavenging of  $\text{HO}\cdot$  radicals under 40 kHz and 200 W increased the hydrogen rate. Using 0.4 g of new solid solutions of lanthanum, gallium, and indium oxides suspended in 200 and 1100 mL of water/ethanol (10% vol/vol) solutions, Luigi et al. [189] studied the production of hydrogen in water-ethanol solutions through heterogeneous photocatalysis, sonocatalysis, and sonophotocatalysis. Using CdS nanorod arrays as a catalyst, Zhao

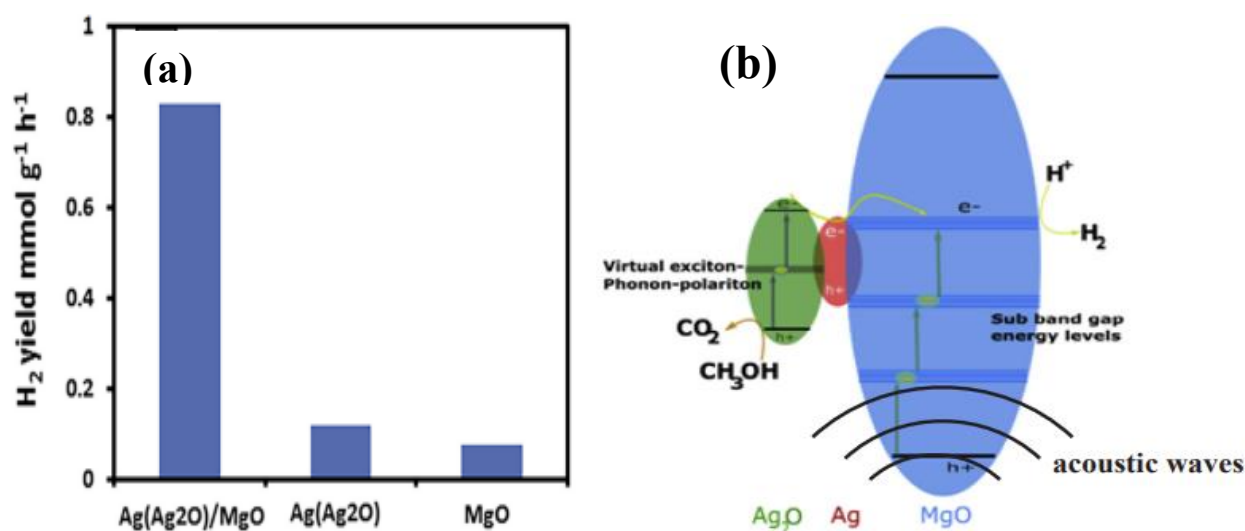
et al. [190] investigated the production of hydrogen using an ultrasonic-assisted photocatalytic process at various ultrasonic frequencies (21, 27, 50, and 68 kHz). Since 27 kHz is quite close to the resonance frequency of the nanorods utilized in the studies, it is likely that this is the reason why the maximum amount of hydrogen was observed at this frequency. According to the same study, the use of a catalyst in photoacoustic hydrogen production is a synergistic process. This is because the photo-generated electrons and holes are forced to move in opposite directions by the ultrasound source, which increases the density of free electrons on the catalyst surface.

Another investigation by Singh et al. [191] examined the use of ultrasound in conjunction with photocatalysis. The researchers used a rGO-supported CdS catalyst, operated at a 20 kHz frequency with input powers ranging from 10 to 40 W, in aqueous solutions containing 4 mM and 10 mM of Na<sub>2</sub>SO<sub>3</sub> and Na<sub>2</sub>S, respectively. Adding acoustic power to the photocatalysis system improved mass transfer of hydrogen from the catalyst surface to the bulk, leading to a 40% improvement in hydrogen generation under 20 kHz and 35 W. Consequently, this improvement was thought to be caused by the mechanical energy of ultrasound, which separates hydrogen bubbles from the photocatalyst's surface, creating more free space for reactants and reactive sites to be adsorbed. In the same study, the influence of temperature was also examined [191]. They found that increasing the temperature from 299 K to 318 K improved sonophotocatalysis of water by 70%. Hydrogen generation increased, according to the authors [191], because of shockwaves and mechanical energy produced when acoustic cavitation bubbles burst.

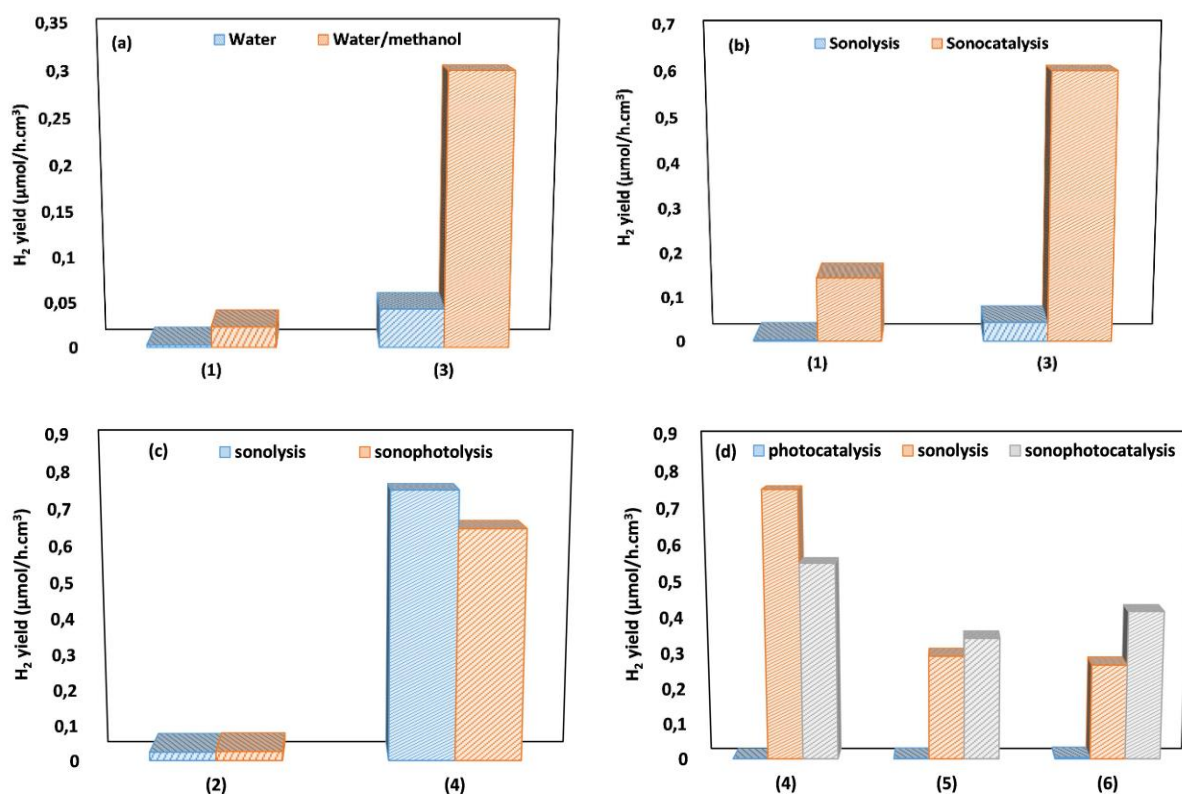
A recent study involved the irradiation of a combination of Ag or Ag<sub>2</sub>O doped MgO with infrared (IR) light in the presence of a methanol/water solution [192]. Under these circumstances, hydrogen generation was detected upon sonication (**Fig. 12.a**), but no hydrogen was produced in the absence of IR light or when the mixture was irradiated solely with the water/methanol solution without any catalyst [192]. Furthermore, no hydrogen was generated when the Ag (Ag<sub>2</sub>O)/MgO catalyst was subjected to IR irradiation in the absence of sonication. The findings indicate that hydrogen generation results from the synergistic effects of sonication and infrared light, implying that

sonication facilitates a multi-step, sub-band gap excitation of electrons in MgO, thereby augmenting the catalytic activity of Ag/Ag<sub>2</sub>O-coated MgO nanoparticles, as illustrated in **Fig. 12.b** [192].

In order to facilitate a succinct comparison of conventional, sonochemical, and coupled procedures for hydrogen generation, a comprehensive review of prior results is illustrated in **Fig. 13**. We standardized all the data by expressing them in uniform units of volume and time. Replacing the usual approach of photocatalysis with methods utilizing ultrasound, as seen in Fig. 13 (d). Nonetheless, we continue to interrogate the comparative efficacy of sonolysis and integrated methodologies. The various behaviors illustrated in **Fig. 13 (a), (b), and (c)** indicate insufficient research in this domain and a superficial understanding of the operational mechanisms of ultrasound, attributed by some authors to the sonochemical process, the mechanical effects of ultrasound [193], the enhancement of electromagnetic radiation absorption properties [194], as well as the reduction in catalyst size [195] and the improvement of mass transfer [196,197]. Isolated analyses of various interpretations within comprehensive experimental research would, in our opinion, yield clearer answers to this topic.

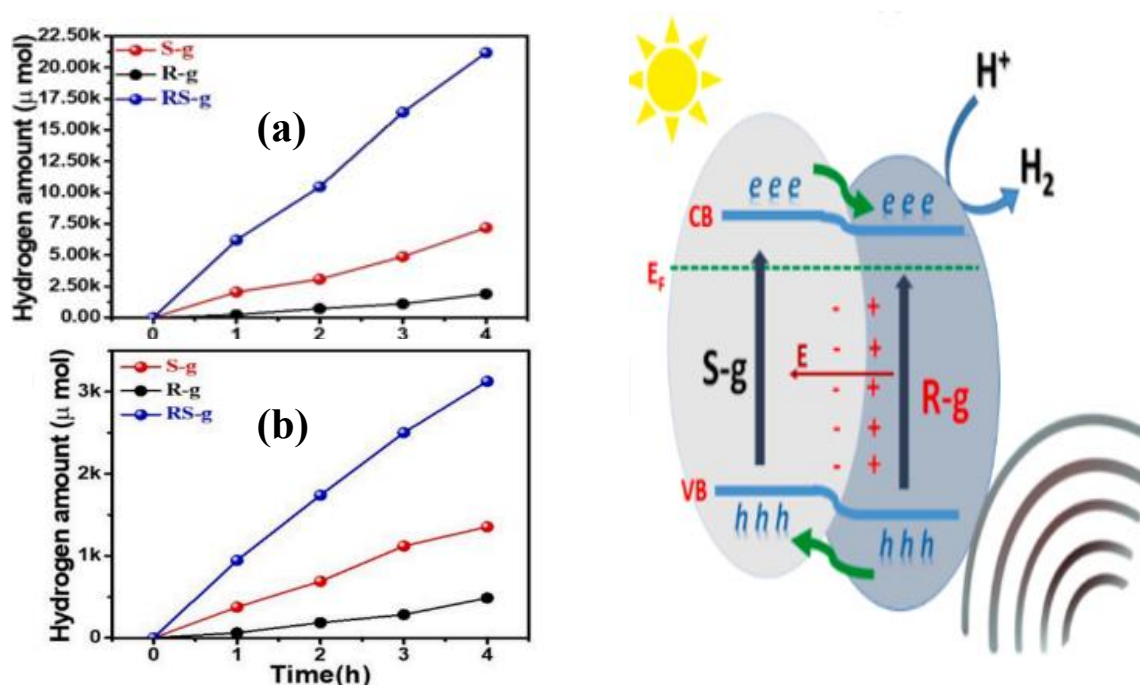


**Fig. 12.** (a) Hydrogen production under infrared irradiation with Ag (Ag<sub>2</sub>O)/MgO, Ag (Ag<sub>2</sub>O), and MgO catalysts in a water-methanol combination subjected to sonication. (b) Schematic representation of the proposed sono-photochemical multistep excitation process in Ag (Ag<sub>2</sub>O)/MgO. Modified with authorization from Reference [192]. Copyright © 2018 Elsevier



**Fig. 13.** Comparison of hydrogen generation rates by sonolysis of pure water and a water/methanol combination (a), sonolysis vs sonocatalysis (b), sonolysis versus sonophotolysis (c), and photocatalysis coupled with sonolysis and sonophotocatalysis (d). Production rate assessed based on the linear progression of H<sub>2</sub> yield as a function of time. Modified with authorization from Reference [198]. Copyright © 2019 Elsevier

The isotype heterojunction of g-C<sub>3</sub>N<sub>4</sub> nanosheets and nanorods was created in a separate study by Bajiri et al. [199]. This photocatalyst enhances HER performance due to its diverse morphologies (**Fig. 14**). The aim of the study was to determine the optimal conditions for hydrogen production by comparing the performance of two distinct morphologies and optimizing the use of glycerol as a sacrificial material. Researchers have also shown that sonophotocatalysis, which combines visible light (LED 90 W) with ultrasonic irradiation, produces outstanding HER results. With regard to photocatalytic HER, the developed isotype heterojunction catalyst outperformed the others that have been disclosed in the literature.

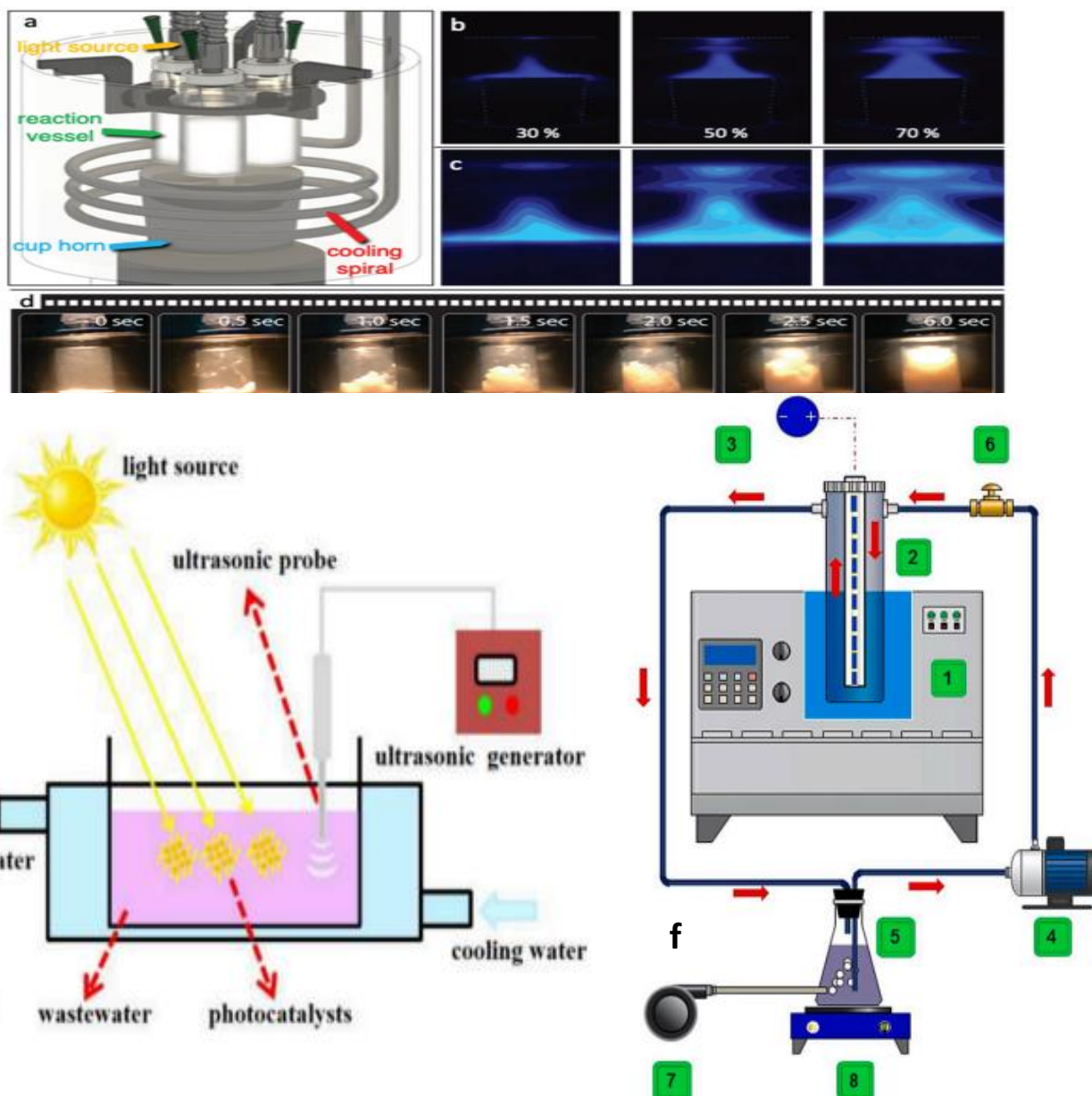


**Fig. 14.** a) Sono photocatalytic HER over R-g, S-g, and RS-g samples. b) Photocatalytic HER over R-g, S-g, and RS-g samples (c) schematic diagram of isotype heterojunction for hydrogen production. Adapted with permission from Ref [199]. Copyright 2023 Elsevier

A synergistic phenomenon occurs in this system when visible light and ultrasound are used together. Ultrasonic radiation can affect the photocatalytic process in a way that promotes the formation of active oxygen agents ( $H_2O_2$ ), which is a byproduct of water sonolysis, function as sacrificial agent by absorbing holes produced by light [200]. There is a strong correlation between the  $H_2O_2$  and  $H_2$  creation during sonophotocatalysis, which demonstrates a considerable improvement in hydrogen production [201]. Another possible explanation for this improvement is that the mass transfer barrier is significantly reduced when ultrasonic cavitation combines liquid circulation current with intense turbulence on a micro-scale, homogenizing the medium [202]. In addition, the formation, coalescence, and release of ultrasonic cavitation bubbles at the interfaces between phases enhance mass transfer and accelerate reaction kinetics. Based on these findings, we conclude that sonophotocatalytic HER outperformed both photocatalytic and sonocatalytic processes individually (Fig. 13).

Building upon existing research in ultrasound-assisted catalytic processes for hydrogen production, which has yielded promising results but still faces several challenges, the development of optimized reactors for sonophotocatalysis is vital to overcoming these limitations. Sonophotocatalysis presents significant opportunities for improving pollutant degradation, enhancing energy efficiency, and advancing hydrogen production from renewable sources such as biomass, water, or waste materials. However, to unlock the full potential of this technology and scale it effectively for broader applications, it is crucial to design innovative reactors that incorporate key elements. These include the precise placement of sonotrodes, which create intense cavitation zones, the strategic positioning of light sources to ensure even photon distribution for efficient catalyst activation, and the design of flow systems that facilitate optimal and stable energy transfer. These elements work in harmony to maximize the reactor's performance and efficiency. **(Fig. 15)** illustrates the various sonophotoreactors, emphasizing their design and functional components. It provides a detailed view of how the arrangement of these reactor elements directly impacts performance, highlighting the crucial role of reaction instrument design in optimizing sonophotocatalysis. While batch reactors are widely used in laboratory settings, they are limited in their scalability for industrial applications. To address this issue, continuous flow reactors and hybrid systems are emerging as more scalable solutions. These systems offer the advantage of continuous operation, minimize energy losses, and enhance the synergy between ultrasound and light, thus improving overall efficiency [203–205]

Looking forward, the future prospects for sonophotocatalysis lie in refining these reactor designs to tackle energy dissipation challenges, improve process productivity, and make the technology adaptable to large-scale industrial settings. This progress will enable sonophotocatalysis to reach its full potential in key industries such as wastewater treatment, sustainable energy conversion, and the production of green chemicals. By continuously enhancing these reactor designs, this technology will play a crucial role in addressing global environmental and energy challenges, offering more sustainable and efficient solutions for the future.



**Fig. 15.** (a) Schematic illustration of the SonoPhotoReactor (SoPhoReactor, SPR); (b) real photographs during the luminol tests for different amplitudes/powers (power density per volume above the surface of the horn  $0.16 \pm 0.1$ ,  $0.33 \pm 0.2$ , and  $0.52 \pm 0.2 \text{ W cm}^{-3}$  for 30, 50, and 70% amplitude, respectively); (c) power mapping of sonochemiluminescence to highlight the most intense regions of ultrasound waves derived from luminescence tests as a result of OH radical formation due to “acoustic cavitation”; and (d) photographs of the reaction vessel after ultrasound irradiation (30% amplitude) loaded with  $1 \text{ g L}^{-1}$  of catalyst (captured with an iPhone X). e) Schematic diagram of the sonophotocatalytic reactor. f) .Sonophotocatalytic reactor set-up: (1) ultrasonic bath, (2) reactor vessel, (3) LED source, (4) peristaltic pump, (5) reservoir, (6) sampling pump, (7) aeration pump and (8) magnet pump) Adapted with permission from Ref [203-205]. Copyright 2023 Elsevier



## 7. Conclusion and future perspectives

Finally, our research, as described in the first part of this review, shows that sonophotocatalysis is an effective method for treating wastewater, with advantages over both sonocatalysis and photocatalysis alone, such as shorter reaction times and higher removal rates. Second, ultrasonic and light irradiation work together synergistically to clean the catalytic surface in real time, addressing issues with catalyst opacity and porosity. Furthermore, sonophotocatalysis removes catalyst fouling, increases free radicals' production ( $\cdot\text{OH}$ ,  $\cdot\text{O}_2^-$ ), and removes resistance to mass transfer from the liquid phase to the catalyst surface. In terms of sonophotocatalysis for hydrogen production, the findings indicate that the impacts of ultrasound irradiation are primarily physical in nature. This is because the mass transfer of hydrogen from the catalyst surface to the bulk phase is improved, while charges are separated, leading to an increase in photoactivity. Consequently, hydrogen bubbles detach from the catalyst's surface. Additionally, the mechanical energy and shock waves produced when acoustic cavitation bubbles collapse further enhance adsorption characteristics and reactive sites. The introduction of ultrasonic vibration into the photocatalytic conversion of biomass for hydrogen generation has numerous benefits, including the acceleration of the photocatalytic process. On a larger scale, it would improve the process's efficiency and sustainability. From a kinetics perspective, the photocatalytic process can be improved by promoting the mixing of the photocatalyst with biomass, enhancing the formation and transfer of reactive oxygen species (ROS) to the biomass matter, and improving mass transfer as a result of shock waves and jetting phenomena. The oxidation of biomass and the creation of radical intermediates can be directly influenced by ROS production induced by ultrasound.

One key advantage of using ultrasonic fields for photocatalytic biomass conversion is their ability to regulate the concentration of dissolved oxygen in the medium. Another point to note is that when different carbon materials are used, the effectiveness of organic pollutant decomposition in the water phase when exposed to ultrasound is enhanced. To further improve the degradation of target pollutants, sonocatalysts doped with carbon materials and integrated into carbonaceous matrices cause an increase in the production of free hydroxyl radicals in the solution. Moreover, the

incorporating sonocatalysts based on carbon not only enhances free hydroxyl radical production by increasing the number of reaction nuclei, but it also facilitates the adsorption of various molecules onto the carbon surface, ready to be oxidized by newly formed free radicals. Additionally, several challenges are still facing the appropriate application of sonophotocatalysis for hydrogen production and as examples include:

- The optimization parameters of the ultrasonic process to assist the photoreforming of biomass and different solid wastes is imperative, such as the intensity of ultrasound, the frequency, and the time need to be intensively investigated in future studies.
- The optimization of the sonophotocatalytic reactor's geometry is critical to achieving a homogenous reaction medium.
- Further research is needed to understand the future of sonochemistry and ultrasound-assisted approaches for hydrogen production, by highlighting the significant mechanical impact of hydrodynamic cavitation compared to the more conventional acoustic cavitation of a technical-economic point of view.
- From a sustainable and large-scale application perspective, it is highly advised that further studies be conducted on improving the ultrasonic process for biomass dissolving in water.

In all probability, this review provides valuable insights into methods for enhancing the qualities by combining ultrasonic and photocatalytic processes, which circumvents the drawbacks of each approach independently. This summary also offers an innovative approach to adjust the photocatalytic characteristics, which encourages the method's practical application. Finally, effectively advance the progress of hydrogen production from different wastes by highlighting the perfect and economical combination of (sono)photocatalysis, and which introduces sonophotoreforming to the industrial world as a new dimension for the hydrogen production in large scale and supported for the advancement of our ecological system.

## Declaration of competing interest

The authors declare that there is no known competing interests or personal relationships that could have appeared to influence the research work reported in this manuscript.

## Data Availability

The data presented in this manuscript is available with the corresponding author and can be provided on reasonable request.

## Acknowledgments.

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