The Sono-Hydro-Gen Process (Ultrasound Induced Hydrogen Production): Challenges and Opportunities

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Abstract

Producing hydrogen using sound waves offers tremendous opportunities, which could lead to a clean, affordable and reliable energy source. Introducing high-frequency sonic waves to liquid water could provide an efficient way to produce efficient and clean hydrogen. This particular review makes a focus on the application of power ultrasound in hydrogen production and discusses the challenges, opportunities and future directions. This new, ultrasonic based hydrogen production technology is given the name of "*Sono-Hydro-Gen*". It is well known that hydrogen can be formed from the dissociation of water molecules subjected to ultrasound via the so-called sonolysis process. Factors affecting the hydrogen production rate and the theory beyond these effects are described herein. The average hydrogen production-rate reported from the *Sono-Hydro-Gen* process is 0.8 μMole per minute at an acoustic intensity of 0.6 W cm^{-2}. This review also compares the Sono-Hydro-Gen technology with the most commonly used technologies and it is found that this technology would lead to a prosperous and secure hydrogen energy for the future. Recent numerical and experimental investigations on the hydrogen production pathways have been reviewed showing various numerical simulations for different experimental configurations. Finally, performance and efficiency criteria are discussed along with the challenges associated with the *Sono-Hydro-Gen* process.

Keywords: Ultrasonic hydrogen production, Acoustic cavitation bubble, *Sono-Hydro-Gen*, Efficiency.

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Nomenclature

c	Speed of sound [m/s]	*Acronyms*	
C_{pUs}	Specific heat of water	HHV	Higher Heating Value
h	Energy input	GHG	Greenhouse gases
M_{us}	Mass of water [kg]	KI	Potassium iodide
p_a	Acoustic pressure amplitude [kPa]	PZT	Piezoelectric transducer
r	Bubble radius from the center [m]	SE	Sonochemical efficiency
t_{us}	Time [s]	*Greek symbols*	
ΔT	Temperature difference [K]	ρ	Density [kg/m^3]
u	Particle velocity	μ	Dynamic viscosity [kg/m/s]
x	Position in *x* direction [m]	ν_0	Velocity amplitude of the probe
z	Acoustic impedance	λ	Wavelength [m]

1. Introduction

Different carbon-cutting options are under the spotlight, and arguably, one of the most promising is the use of hydrogen as an energy carrier [1]. Highlighting the need for drastic cuts in carbon emission, hydrogen is one among the most powerful fuels and highly suitable for clean energy production [2]. Furthermore, hydrogen is an excellent high capacity and long-term energy storage medium that can be connected to the intermittent renewable energy technologies like wind and solar. Producing hydrogen that can be kept in gaseous or liquid form for an undefined periods without affect its energy content adversely can be a challenge. At times of peak power demand, the stored hydrogen can then be used to generate grid electricity using Combined-Cycle Gas Turbines (CCGTs) or distributed energy supplies using fuel cells, for example; alternatively, it can be harnessed to heat households, to fuel vehicles, and in many additional applications. In recent years, the idea of harnessing hydrogen as a mainstream energy option has been given a great importance. Many research studies are probing the opportunities and challenges of *hydrogen-for-*

future-energy considering an important and yet a very basic question: *how might we produce sufficient hydrogen, produce it sustainably enough, and clean to meet the needs for a low-carbon economy* [3,4]*?*

Answers to this very frequent question are provided in this review. The importance of hydrogen to power our economies and societies and the potential use of power ultrasound for producing hydrogen is also highlighted herein.

1.1 Hydrogen for future

Hydrogen is known to have the highest heating value per unit volume as a fuel. Demand for energy has grown substantially over the past century and continues to grow at a staggering pace. One of the primary sources of energy is fossil fuels. However, the combustion of fossil fuels (mainly hydrocarbons), in internal combustion engines is usually incomplete, resulting in reduced efficiency and the emission of various pollutants into the atmosphere [5]. In this regard, it is known that using hydrogen as fuel increases the efficiency of the internal combustion engines and reduces drastically GHG emissions. Hydrogen is considered as one of the key parameters for a clean and environmental energy source. In addition, it is a renewable source of energy. Hydrogen fuel is one of the most powerful fuels because it has a very high energy content of 141.8 MJ/kg which, can be defined as the amount of energy released while burning 1 (one) kg of the fuel [6,7]. It's heating value is approximately 3 (three) times higher than that of the natural gas [8]. Many countries are mainly using fossil fuels for energy production, which leads to a tremendous amount of pollutants. Consequently, hydrogen is considered highly suitable for clean energy. Recently, hydrogen is used in the Integrated Gasification Combined Cycles (IGCC) as a fuel blend because it is characterized by lower GHG emissions such as carbon dioxide (CO_2) and nitrous oxides (NO_x) [8,9]. In the following sections, different methods of hydrogen production will be briefly presented.

The benefits of using hydrogen as a fuel can be summarized as follows: it is an environmentally friendly, non-toxic, efficient fuel and a renewable source of energy, emitting very low levels of greenhouse gases when burnt. However, the challenge lies in an energy-hungry, low-carbon age, is to manufacture enough hydrogen and to do it cleanly and cost effectively.

1.2 Potential ultrasound application

Ultrasound has the power to explore and destroy. The ultrasound frequency is the sound frequency beyond which the human ear can react. In other words, it is a sound of frequency after which human will not hear. The ultra-power of sound has been widely used in the medical and clinical applications. However, it has many applications in the engineering field as well. The ultrasound method offers a potential option in routinely engineering applications for monitoring and diagnostics processes [10–12].

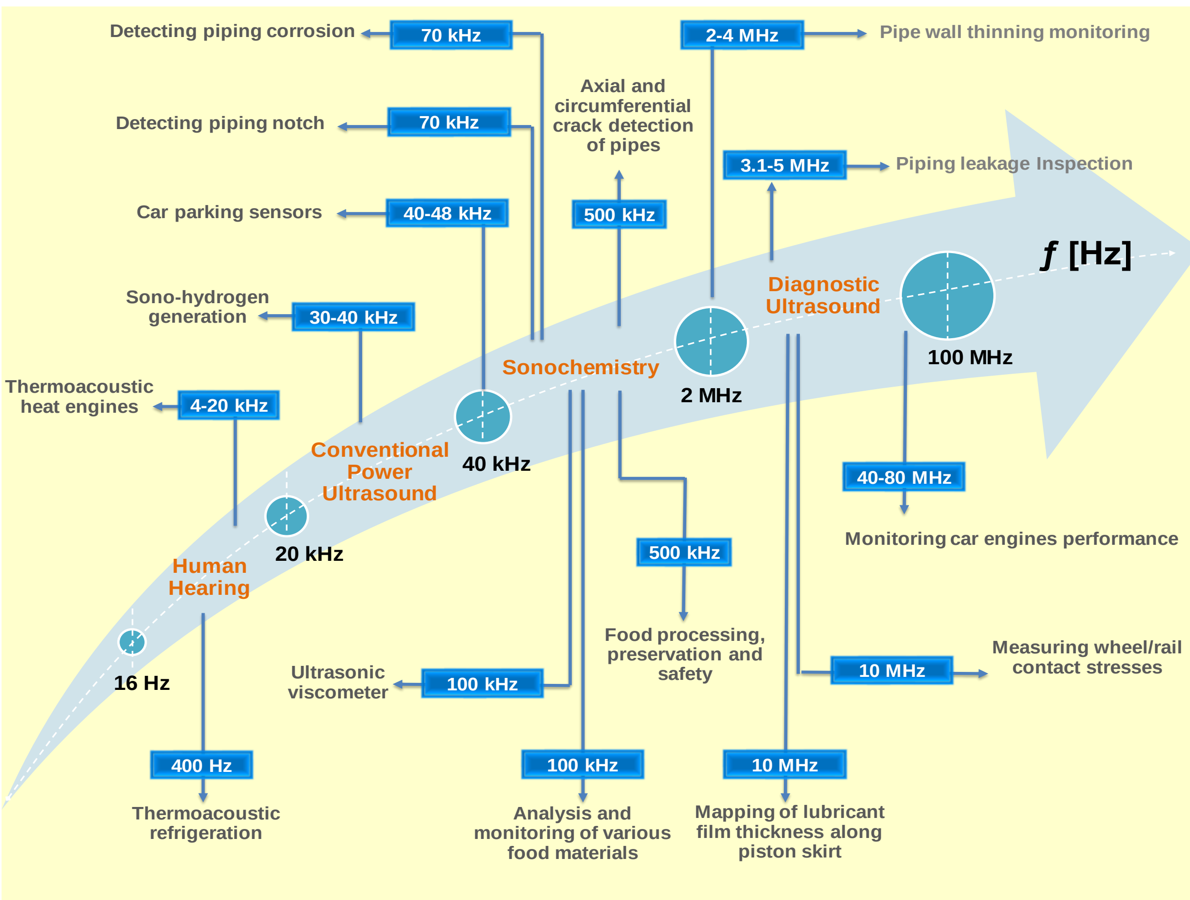


Figure 1: A summary of ultrasound applications at different corresponding ultrasonic frequencies

In Figure 1, a summary of several applications is presented with their corresponding ultrasound frequency ranges specified in previous studies. Ultrasound can be used, either by itself or in combination with emerging technologies; and it has been used for different applications including, thermoacoustic heat engines, thermoacoustic refrigerators, axial and circumferential crack detection, detecting piping corrosion/notch, pipe wall thinning monitoring, ultrasonic viscometer, monitoring food materials, monitoring food processing and preservation, car parking sensors, mapping of lubricant film thickness along piston skirt, monitoring car engines performance and measuring wheel/rail contact stresses. Mohany et al. [13] have reviewed the development and recent patents on thermoacoustic devices.

On the other hand, however, engineering ultrasound has taken a new form of life as productive applications to produce useful gases (e.g. hydrogen) by either sonication [14] or trans-esterification. One example is the *Sono-Hydro-Gen* process, which is the main topic of this review article. Each application in Figure 1 is associated to more than one literature report as summarized in Table 1 providing an overview of the ultrasonic frequency condition, a couple of references and a short description for different ultrasound applications but does not report on the recommended ultrasonic frequency or frequency range for each application. Other important parameters including, acoustic power, intensity, pressure amplitude and irradiation durations are not listed for the sake of clarity.

Table 1: A summary of ultrasound applications in the engineering field

Application	Frequency	Description	References
Thermoacoustic refrigeration	400 Hz	An eco-friendly refrigeration technology, which triggered energy to transfer heat from one side to another side.	Wetzel et al. [15]; Newman et al. [16];
Thermoacoustic heat engines	4-20 kHz	A thermos-acoustic technology that utilizes the power of ultrasound waves to pump heat from one side to another and contrariwise. This application uses the temperature difference to produce high amplitude sound waves.	Backhaus and swift [17,18]; Ghazali [19]
Ultrasonic in Extraction	20 kHz	The process of transferring a substance from any matrix to an appropriate liquid phase, assisted by sound waves (>20 KHz in frequency) that propagate through the liquid media.	Alzorqi and Manickam [20,21]; Chemat and Ashokkumar [22]
Sono-Hydro-Gen	20-1000 kHz	A new method for H_2-production from a mechanical source such as ultrasound. The power from ultrasound used to dissociate the water molecules into OH and H radicals, then these radicals recombine together to produce H_2 and O [23].	Pollet et al. [24] Son et al. [25]
Ultrasonic crack inspection	40-48 kHz	Ultrasonic crack inspection has the following procedure: Introduce ultrasound pulses to piping system, reflection and refraction at the inner wall surface, corner reflection at external crack, receiving surface echo and finally receiving crack echo.	Glushkov et al. [26]; Komura et al. [27]; Burrows et al. [28]

Detecting Piping corrosion	70 kHz	A new technology for detecting of corrosion in piping system of chemical plants using cylindrical waves.	Alleyne and Cawely [29];
Detecting notch	70 kHz	Detecting notch by using ultrasound waves	Lowe et al. [30]
Novel Ultrasonic Viscometer in Engines	100 kHz	To obtain the viscosity of the mixture it is mandatory to choose a lower operating frequency at 100 kHz.	Schirru et al. [31]; Markova et al. [32]
Monitoring of food materials	100 kHz	Quality control and monitoring of different food materials throughout food industry to guarantee and maintain high quality and safety food production.	Awad et al. [33]; Mason et al. [34]
Food processing, preservation and safety	500 kHz	The power of ultrasound is implemented at higher frequencies to induce physical, mechanical and biomedical effects on the foods properties and considered very promising in food preservation.	Juarez et al. [35] Chandrapala et al. [36]
Axial and circumferential Crack detection	500 kHz	Detecting axial and circumferential cracks in piping systems of nuclear power plants via the analysis of the short-time Fourier transformation.	Cheong et al. [37]; Reber et al. [38]
Piping inspection	3.1-5 MHz	Development of ultrasonic solutions for sewer inspection. Pipe deformation and anomalous conditions can be simulated.	Gomez et al. [39] Liu and Kleiner [40]
Pipe wall thinning monitoring	2-4 MHz	It is a non-destructive type evaluation of wall thinning in power plants for continuously monitoring the plant while under operation.	Kosaka et al. [41] Lee et al. [42]
Mapping of the film lubricant thickness used in a piston skirt	10 MHz	To outline the applicability of the ultrasonic methodology to both piston skirt film thickness measurement and show the possibility of deducing some piston secondary motions.	Mills et al. [43]
Measuring Wheel/Rail Contact Stresses	10 MHz	To investigate how rail components contact by reflection of ultrasound. The acoustic wave is emitted and bounced back from an incomplete interface. The higher the interaction load, the more reasonable will be the contact and consequently more wave will be transmitted.	Marshall et al. [44,45]
Monitoring Car Engines	40-80 MHz	Ultrasound scans have been widely used in medical application, but it has never been used in testing the performance of a modern combustion engines. It is used to measure the engine performance parameters. Such as monitoring the piston oil film, piston ring oil and lubricant film.	Dwyer Joyce [46,47] Avan et al. [48]

In closing, ultrasound has a wide history of use in routinely engineering applications for monitoring and diagnostic as summarized in Table 1. However, ultrasound can be used for producing useful gases such as hydrogen [24]. The ultrasonic power can produce acoustic cavitation-bubbles; the implosion of these bubbles after several successive growths will result in a tremendous amount of energy enough to produce hydrogen from liquid water via the sonolysis process. This *Sono-Hydro-Gen process* has a significant implication that takes place in the frequency range between 20-40 kHz. Producing hydrogen using the power of ultrasound is a significant challenge, although ultrasound offers an eco-friendly way to produce hydrogen by introducing ultrasound waves to liquid water.

1.3 Review objectives

Knowledge about hydrogen production using sonoreactors is insufficient for describing the best operating conditions. Recently, limited studies consider the *Sono-Hydro-Gen* approach for hydrogen production. However, factors affecting the hydrogen production rate is still unclear [24] The obvious advantage of this technique is to highlight the tremendous opportunity and the new venue that will be opened to many research studies for hydrogen production via sonication. This challenge lies in advancing our fundamental understanding of the novel approach and probing the different influential factors to obtain the optimum H_2-production rate. With the ability of ultrasound and with the potential of applying ultrasound waves to liquid water, ultrasound forms a powerful tool for the future of hydrogen generation.

This review article is a follow-up review by Pollet et al in Ultrasonic Sonochemistry [24] and is to provide further a comprehensive review on the *Sono-Hydro-Gen* technology. In this review, an intensive introduction to the use of the ultrasonic power in the engineering applications is carried out. Different methods of hydrogen production are demonstrated and compared to assess whether these methods are effective and environmentally favorable. Furthermore, the article promotes our understanding of the ultrasonic power on the *Sono-Hydro-Gen* technology along with enhancing the knowledge of the mechanism associated with hydrogen production as the mechanism is not yet understood and the most reported suggestions are controversial [49]. Factors affecting the hydrogen production rate and the theory beyond these effects are well analyzed and reported. Recent numerical and experimental investigations on the hydrogen production scheme are intensively reviewed showing different numerical simulation and different experimental configurations. Finally, performance and efficiency criteria are reviewed along with the challenges associated with the *Sono-Hydro-Gen* design.

In the next section, different hydrogen production methods will be briefly reviewed and tabulated to compare whether these methods are environmentally friendly and economically feasible.

2. Hydrogen Production Processes

Hydrogen is not only very powerful and efficient but also it is a renewable source of energy, as it can be produced via five main categories of technology, namely, thermochemical [50], (ii) electrochemical [51], (iii) photobiological [52], (iv) photoelectrochemical [53], and (v) *Sono-Hydro-Gen* [54], which are all summarized in Figure 2. A brief overview is made to describe each of these technologies stating the advantages and disadvantages of each of them and a comparison is drawn among all methods in terms of the process, the chemical reaction, the advantages and disadvantages, and the H2- production rate and cost.

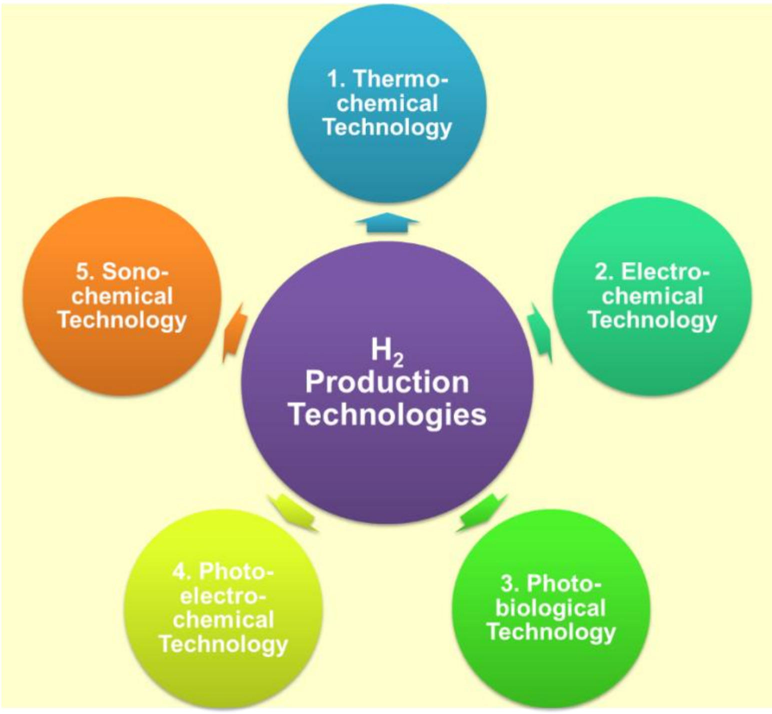


Figure 2: Hydrogen production through different sources of energy

(i) Thermochemical technology. This mean of hydrogen production involving the steam gas reforming (SMR) which is considered one of the widely used means of hydrogen production from a gas material such as methane, ethanol and methanol. However, the gasification processes are used when the raw material is solid such as: coal or biomass [55–57]. This technique has a wide sustainability problems, therefore, Dincer and Acar [4] reviewed and evaluate different hydrogen production methods for enhancing sustainability of such a technique. Stream gas reforming is not an environmentally friendly way for hydrogen production as reported by Haryanto et al. [58].

(ii) Electrochemical technology. This technique is in charge with water electrolysis (WE) to produce hydrogen. This technique is a high-energy demanding with an overall efficiency of 60%. In fact, it can be very efficient if the electricity cost is below 2 cents/kWh.

(iii) Photobiological technology. The photobiological technology uses the natural photosynthesis activity of bacteria and green algae to produce hydrogen. One main problem is that the production rate is very slow. Detailed reviews associated with this technique can be found in [59,60].

(iv) Photoelectrochemical technology. The photo-electrochemical technology is producing hydrogen in only one-step using the water-splitting phenomenon via illuminating a water-immersed semiconductor with sunlight. A better technique to produce more cleaner hydrogen is the so called "Photocatalytic water splitting" which can decompose oxygen and hydrogen by utilizing sunlight with the aid of photo-catalyst [61–64]. One obstacle of this methods is that the instability of the semiconductor materials in the aqueous phase. Other disadvantages is provided by Haryanto et al. [58].

(v) Sonochemical technology. Sonochemistry is defined as how the power of ultrasound can be utilized in chemistry. In fact, it has been well recognized that, hydrogen can be produced by introducing ultrasound waves to liquid water. As compared to the other non-renewable energy sources, hydrogen can be produced infinitely by simple means of separation from water molecules. This can be provided by the *Sono-Hydro-Gen* approach.

A summary

Table 2 presents a brief illustration and a comparison between the five main categories of hydrogen production in terms of fundamental theory and remarks. The thermochemical technology is associated with the steam gas reforming [65] which is not environmentally friendly method, whereas, the electrochemical technology related to water electrolysis requires high electrical power. The photobiological technology problem is that the production rate is very slow. The Photo-electrochemical technology is producing hydrogen in only one-step using the water splitting. In all cases, though, the need to drive costs and carbon down and to drive efficiency and production capacity up provides a striking solution right to the heart of the problem confronting hydrogen-for-energy.

The next section will focus on the sonochemistry technology, benefits of sonochemistry, *Sono-Hydro-Gen* theory and the system design. The originality of this interesting topic goes back to the sonochemistry field.

Table 2: A conceptual illustration of different H_2-production methods

H_2-production methods	**Theory beyond each method**	**Remarks**	**H_2-Production rate and cost**
Thermochemical (steam reforming)	$CH_4 + 2\ H_2O + h_{thermal} \rightarrow 4\ H_2 + CO_2$	Not environmentally friendly as carbon dioxide is produced.	9-12 tons of CO_2/ 1 ton H_2 [66].
Electrochemical (water electrolysis)	$H_2O + h_{electrical} \rightarrow H_2 + \frac{1}{2}\ O_2$	Not environmentally friendly as it requires high electrical energy.	53.4-70.1 kWh/ 1 kg of hydrogen [67].
Photobiological	$2\ H_2O + CO_2 +$ *Algae/Cyanobacteria* $+ h_{solar} \rightarrow O_2 + 4\ e^- + 4\ H^+ \rightarrow 2\ H_2$	Environmentally friendly, however, it has very low and slow production.	0.07-96 mmol $H_2\ L^{-1}\ h^{-1}$ [68]
Photoelectrochemical	$H_2O + h_{solar} \rightarrow H_2 + \frac{1}{2} O_2$	Environmentally friendly, however, it has limited durability because of the instability of the semiconductor material. This technology is still under development. The challenge lies on the durability material and the steady operation. This technology is very expensive.	17.3 \$/kg of H_2 [69].
Sonochemical	$H_2O + h_{sound} \rightarrow OH^* + H^*$ $OH^* + H^* \rightarrow H_2 + O$ [49]	Environmentally friendly, sustainable, durable and low-energy consumption.	0.8 μM min^{-1} at acoustic intensity of 0.6 W cm^{-2} [70].

3. Sonochemistry Technology

In this section, sonochemistry approach is will be reviewed presented as well as the Sono-Hydro-Gen process. The sonochemistry approach is defined as when ultrasound waves are introduced into a liquid medium to bring an unusual chemical environment [71]. Ultrasound waves are introduced to the sonication medium by ultrasonic transducers. Piezoelectric transducers selection and applications in sonoelectrochemistry are reported by Pollet [72]. The *Sono-Hydro-Gen* approach is one of several benefits beyond the sonochemistry, and it will be illustrated in a later section in details. The power of ultrasound can generate an acoustic cavitation bubble within a liquid. Pollet [14] summarized the main benefits of the acoustic cavitation bubble. Highlighting that it can produce a tremendous amount of energy, which can enhance a range of chemical reactions and can enhance the electrochemical diffusion processes.

3.1 Sonoelectrochemistry

Sonoelectrochemistry is defined as a combination of three fields including electrolysis, ultrasound and electrochemistry which is initially reported by Morigushi in 1930s. In the electrolysis process, hydrogen is produced at the decomposition potential in the molecular form which is taking place on the surface of electrodes via electrochemical reaction. Then the molecular hydrogen gas nucleate at the cavity of electrode surface to hydrogen gas bubbles at the cathode active sites. The hydrogen gas bubbles start to enlarge at the surface of the electrode. Early in 1990s, Sheng-De Li et al. [73] and Richard et al. [74] reported that the effect if introducing ultrasonic waves to an electrolysis process will increase the energy efficiency considerably.

In next sub-sections, fundamental aspects, many benefits, Sono-Hydro-Gen production approach, acoustic cavitation bubbles and important factors affecting the hydrogen production rate will be coherently reviewed.

3.2 Benefits from sonochemistry

The ultrasound is widely used for several applications in different fields including hardening by immersed metals [76], several medical and clinical applications, for example: drug delivery and other therapeutic applications [77], enhanced electrospinning [78], enhanced bladder cancer therapy [79] and accelerating chemical reactions and processes [80]. The ultrasonic waves and irradiation are associated with efficient chemical and physical effects for driving enhancing the chemical reactions and yields. The idea beyond using ultrasound is to use less hazardous chemicals and solvents and to reduce energy consumption. There are several benefits beyond the sonochemistry approach such as it can enhance the electrochemical diffusion processes.

Ultrasound waves used to enhance the chemical reactions and to provide an unusual chemical environment. For example, organic syntheses can be greatly improved by the use of ultrasound. A comprehensive review is performed on the ultrasound in synthetic organic chemistry concentrated on the applications in organic synthesis by Mason [81]. Many other researchers e.g. Cravotto and Cintas [82] and Bang and Suslick [83] have performed successfully synthetic organic reactions using ultrasound. Production of nanomaterials, environmental treatment, purifying water, corrosion of metals, cleaning of polymeric membranes, food processing, cavitation bubble dynamics and hydrogen production. Chen [84] performed a comprehensive review on the applications of ultrasound in water and wastewater treatment.

A summary of the recent different research disciplines utilizes the benefits of the sonochemistry technology is summarized in Table 3 including the area of research, recent or old references and a short description of each discipline.

Table 3: Summary of the recent available area of research using the sonochemistry

Area of research	Description	References
Organic syntheses	The ultrasound in synthetic organic chemistry.	Luche et al. [85]; Einhorn et al. [86]; Mason [81]
Production of nanomaterials	The ultrasound technology is used for preparing nanomaterials by the means of pulsed sonoelectrochemistry. Application of nanoparticles in electrochemical is also reported by Luo et al. [87].	Saez et al. [88]; Luo et al. [87]; Pollet [12] Muthoosamy and Manickam [89,90]
Environmental treatment	It can be used for water and wastewater treatment by using the advanced oxidation processes for the remediation of water, wastewaters, odors and sludge.	Simon Parsons [91]; Oller et al. [92]; Poyatos et al. [93]
Water disinfection or purifying water	The ultrasound is used also for purifying water	Esclapez [94,95] Panda and Manickam [96]
Corrosion of metals	The corrosion behavior of these coating on some metal studied by the electrochemical methods	Ashasssi and Bagheri [97,98]; Mason [99]
Cleaning of Polymeric membranes	The ultrasound waves are also used for cleaning of polymeric membranes for water treatment	Chai et al. [100] Howell and Velicangil [101]
Ultrasound in food processing	Ultrasound is promising for food processing because it has a significant effect on enhancing several food processes.	Chemat et al. [102]; Mason [103]; Chandrapala et al. [104]; Knorr et al. [105]
Cavitation bubble dynamics	The sonoelectrochemistry approach is used to investigate the dynamics of cavitation bubbles and flow velocities	Pollet et al. [72,106]; Ashokkumar et al. [107,108]; Lee et al. [109]
Ultrasound in separation	In recent years the use of high frequency ultrasound standing waves for droplet or cell separation from biomass has emerged beyond the microfluidics scale into the liter to industrial scale applications	Spotar et al. [110] Manickam et al. [111]
Sono-Hydro-Gen	The process is firmly illustrated in details	Merouani et al. [23,49] Son et al. [25,112]

3.3 Sono-Hydro-Gen system illustration

When the sound waves with high frequency passing through a liquid such as water, it will lead to vibration of liquid water mechanically, it is so-called “Water Sonolysis” or “Water Sonication”. Figure 3 shows and illustrates schematic of the sonoreactor model. The ultrasound probe immersed in a water container emits sound waves through the water by a frequency range between 20-40 kHz. Ultrasound also generates acoustic cavitation bubbles within the liquid that are generated at the tip of the ultrasound probe. The typical ultrasound wave has compression and rarefactions acoustic pressure that will accumulate energy inside the acoustic cavitation bubble. This energy is in the form of several thousand of temperatures in kelvin and several hundreds of pressures in atmospheres which is enough to dissociate the water vapor trapped inside the bubble, the so-called sonolysis process [113].

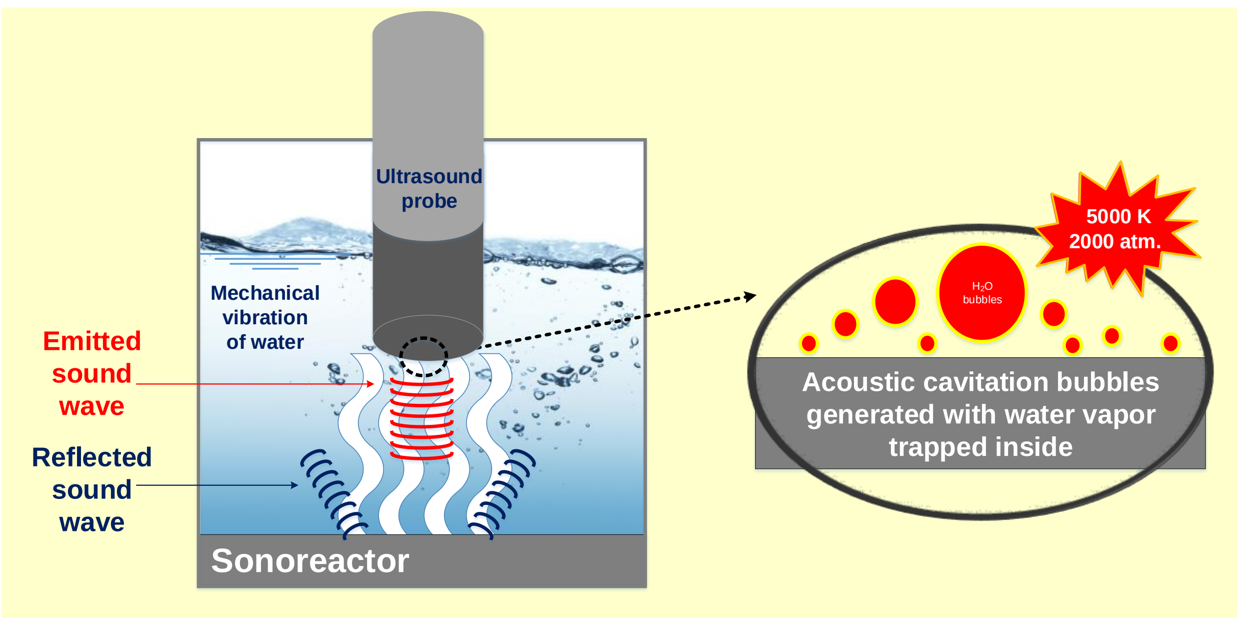


Figure 3: Schematic of the ultrasound generator and probe and the corresponding acoustic pressure waves [10]

These bubbles are so-called acoustic cavitation bubbles that take place when ultrasound is introduced to liquid water; the medium goes through a series of compression and rarefaction cycles. As rarefaction and compression high-frequency sound waves travel through water, the expansion will push apart the water molecules and give the strong negative pressure to overcome, the intermolecular forces while the compressions push the molecules together through the strong positive pressure. If the sound waves strong enough and in succeeding cycles this will lead to a sudden pressure drop at which the cavitation phenomenon occurs and creation of gaseous bubbles in liquid takes place. Sequence and dynamics of acoustic cavitation bubbles. The mechanism has 4 (four) consecutive and instantaneous stages as seen in Figure 4; (a) bubble formation, (b) successive growth, (c) collapse [12], (d) micro jets [77] and as reported by Lee et al. [114,115]. The first stage is the acoustic cavitation bubble formation due to the mechanical vibration of water when ultrasonic waves introduced. The second stage is the bubble enlarges and growth in successive cycles after which the bubble reaches the unstable mode at which it is about to collapse. The third stage is the acoustic cavity implosion at which a violent bubble collapse leading to release high energy. However, a detailed system description can be found in the recent perspective article by Rashwan et al. [10].

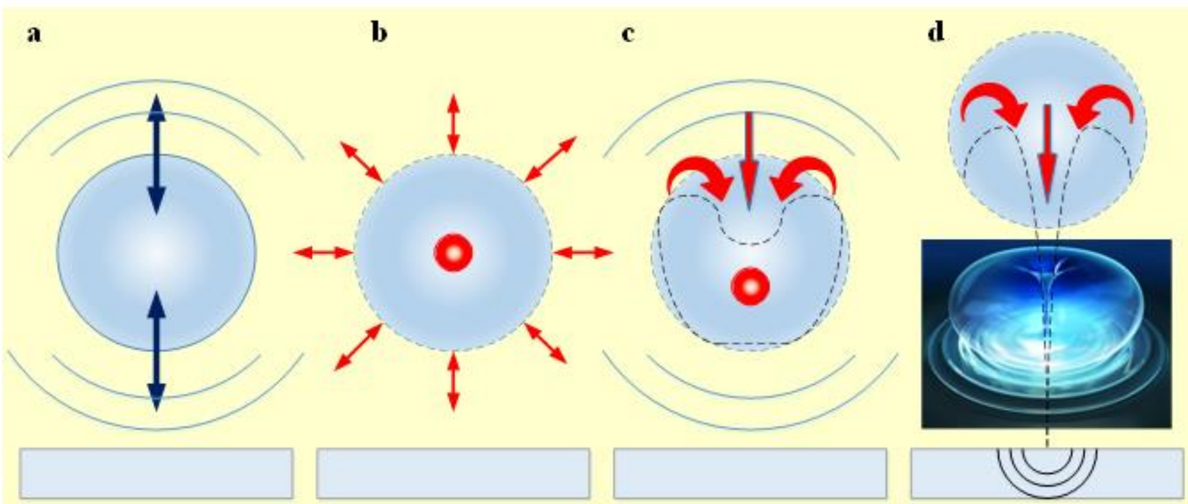


Figure 4: The sequence of acoustic cavitation bubble collapse

The reaction mechanism inside a single-bubble saturated with water vapor during a water sonolysis experiment has a great interest. The rapid heating phase is described as heat generated from the cavity implosion is enough for dissociate the water molecule (H_2O) into highly reactive hydrogen radicals H* and hydroxyl radicals OH*. While the quick cooling process is responsible for recombining the highly reactive radicals H* and OH* to form hydrogen H_2. Merouani et al. [116] reported the most two important reactions that 99.9% of the hydrogen is produced from the gas phase recombination reaction, the reaction can be given as follows:

$$H^* + OH^* \leftrightarrow \; H_2 + O \qquad (1)$$

However, another recombination reaction takes place at the surface of the bubble shell with a minor impact in H_2-production can be given as follows [117]:

$$H^* + H^* \leftrightarrow H_2 \qquad (2)$$

Merouani et al. [118] performed a water sonolysis (waster dissociation to OH+H). They reported that the sonolysis process of water by low ultrasound frequencies result in thermal dissociation of water into hydrogen radicals H* and hydrogen oxide radical OH*, this process is driven by a tremendous amount of heat accumulated inside the bubbles due to a very high temperature and high pressure resulted from cavitation bubbles collapse. Ultrasonic cavitation of water has a subsequent collapse of microbubbles. This is considered a unique phenomenon leads to hydrogen production during the water sonolysis process. Water sonolysis is a promising and clean technique to produce hydrogen, particularly if water is used as the hydrogen source. The effect of the *Sono-Hydro-Gen* parameters is not clarified yet.

In the next section, several factors affecting the H_2-production rate during the *Sono-Hydro-Gen* process will be intensively discussed.

4. Factors affecting the *Sono-Hydro-Gen* process

As a matter of fact, the rate of hydrogen production is governed by several important parameters as shown in Figure 5, foremost the acoustic frequency, acoustic intensity, dissolved gas and the water bulk temperature[116]. However, a way to quantify the hydrogen production rate has not yet been fully developed and still in need of many numerical and experimental investigations.

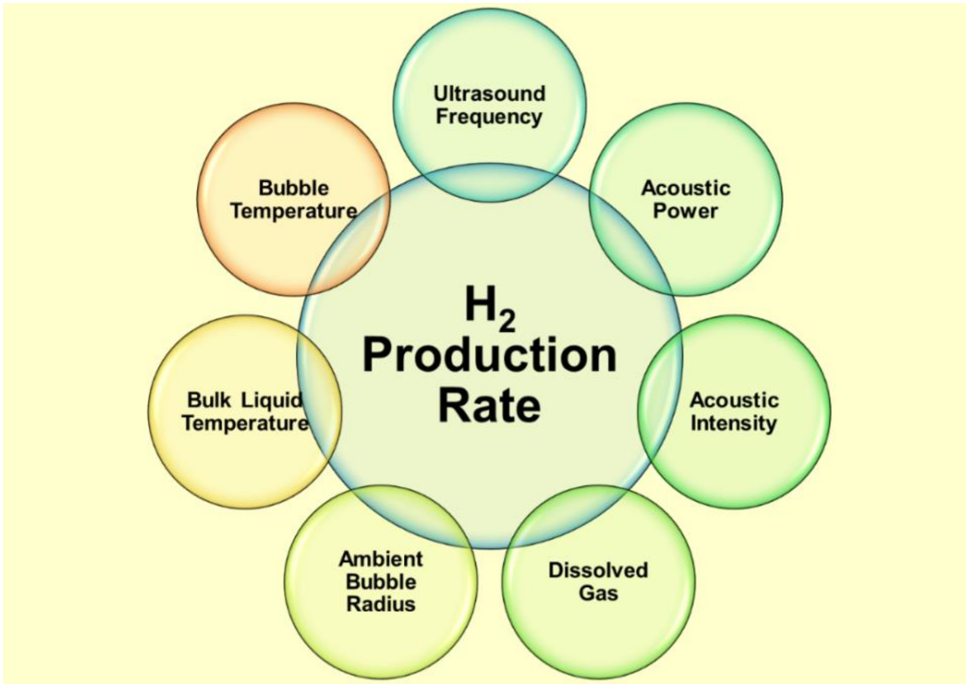


Figure 5: Factors affecting hydrogen production rate through the sono-hydrogen approach

In the following sub-sections, major factors govern the rate of hydrogen production are well illustrated including the theory of how each factor affects the production rate and a summary table is added at the end of different sections to summarize the effect of each parameter on some useful gases produced such as hydrogen peroxides H_2O_2 and hydrogen H_2.

4.1 Ultrasonic frequency

It is noticed that the amount of hydrogen produced from such a process is considered a highly frequency dependent as it is the most important parameter in sono-hydrogen generation. The hydrogen generation rate increases with the increase of applied frequency [119]. Several dynamic factors govern the hydrogen production rate with frequency, namely, maximum bubble core temperature and pressure, the amount of water vapor trapped and the collapse time. At low frequencies, the bubble will have more time to expand and enlarge this would allow more water vapor to be trapped inside the bubble core. As a result, the bubble collapse will be very strong and will generate a higher pressure and temperature, which will promote the chemical reaction producing more radicals. Whereas, at higher frequencies, the collapse time will be very short and the bubbles will not have enough time to generate radical as the reaction inside the bubbles will be very fast. Combining all these factors together, we figure out why the applied ultrasound frequency has a significant impact on the hydrogen production rate. In

Table 4 a summary of the conducted studies on H_2O_2 production using the ultrasound waves is presented while comparing different studies at different ultrasonic frequencies. It can be seen that the H_2O_2 production rate is increasing while increasing the frequency until it reaches an optimum point, then the rate goes down back, this can be attributed to the formation of bubble clouds that attenuate the acoustic intensity, which in turn will reduce the production rate of H_2O_2.

Table 4: A summary of the conducted studies on H_2O_2 production using the ultrasound waves

Frequency [kHz]	**Production rate of H_2O_2 (µMole/min)**		
	Petrier and Francony [120]	Jian et al. [121]	Merouani et al. [23]
20	0.7	1.1	-
200	5	5.2	-
300	-	-	2.5
500	2.1	3	-
585	-	-	4.2
800	1.4	2	-
860	-	-	3.4
1140	-	-	2.1

4.2 Dissolved gas

The effect of dissolved gas on the hydrogen production performance lies between two major physical properties; (1) specific heat capacity ratio ($\gamma = C_p/C_v$) and (2) thermal conductivity (k). The dissolved gas that has higher heat capacity could accumulate higher temperature. Whereas, dissolved gases with low thermal conductivity will have low heat dissipation, which will allow more temperature to be trapped inside the bubble. Consequently, selecting a dissolved gas with high heat capacity and low thermal conductivity will be the optimum selection for enhancing the dissociation process of water vapor, hence, more hydrogen generation in return. Summary of the numerical work carried out on the hydrogen production using ultrasound is presented in comparison on the hydrogen production rate at different frequencies and different dissolved gases from the available literature review and presented in Table 5.

Table 5: A summary of the conducted studies on **H_2**-production using the ultrasound waves

Frequency	**Dissolved Gas**	**H_2-production Rate**	**References**
20 kHz	Argon	0.8 to 5 µMol/min	Venault [70], 1997.
1000 kHz	Argon	13.6 µMol/min	Margulis and Didenko [122], 1984.
1000 kHz	Air	0.22 µMol/min	Margulis and Didenko [122], 1984.
1100 kHz	Argon	10^{-17} Mol/s	Merouani [118], 2014
1100 kHz	Argon	10^{-13} Mol/s	Merouani [116], 2016

4.3 Acoustic power

The hydrogen production rate is highly dependent upon the acoustic intensity. This is attributed to the fact that during the collapse the acoustic bubble is acting as a micro-combustor in which high-temperature chemical reaction takes place and highly reactive radicals are the product of such chemical reaction. The chemical reaction is governed by 3 (three) factors: bubble temperature, collapse time and the bubble size, which correspond to the amount of water vapor, trapped in the bubble. With the increase of the acoustic intensity, the expansion ratio of the bubbles will increase allowing more water vapor to be trapped in every single bubble. Similarly, the compression ratio increases leading to a higher bubble temperature. As a result, the increase in expansion and compression ratios of the bubbles will promote an unusual chemical reaction leading to produce more free radicals from the dissociation of the water molecules inside the bubbles. Furthermore, increasing the acoustic intensity will increase the collapse time, so the chemical reaction will have more time to produce more reactive-radicals. Combining all of these factors together leading to higher H_2 generation. Kerboua and Hamdaoui [123] performed a numerical estimation of hydrogen production at different operating conditions of acoustic power and frequencies. They confirmed the theory of increasing the acoustic intensity lead to an increase in the hydrogen production rate. Their results are extracted and reported in Table 6.

Table 6: **H_2-production** (Mole) at different acoustic power and frequencies. Data extracted from [123] by Kerboua and Hamdaoui.

	Acoustic Amplitude			
Acoustic frequency	1.5 [atm]	2.0 [atm.]	2.5 [atm.]	3.0 [atm.]
200 [kHz]	1.33×10^{-19}	2.53×10^{-17}	7.35×10^{-17}	1.30×10^{-16}
1000 [kHz]	2.98×10^{-33}	5.67×10^{-24}	1.64×10^{-21}	2.91×10^{-19}

4.4 Bulk liquid temperature

The cavitation is considered a dynamic phenomenon, which is strongly affected by the operating parameters such as bulk liquid temperature, static pressure, and geometry of sonoreactor. The reaction mechanism of the sonochemical process is influenced by the bulk temperature as pointed out by Sutkar and Gogate [124]. Any tiny changes in the temperature will alter the conditions of pressure and acoustic intensity of the liquid medium that may yield a dramatically different cavitational effect [125]. Therefore, studying the temperature change with the ultrasound irradiation is considered considerably important to understand the characteristics of the flow and acoustic fields inside the sonoreactor. Few studies have considered the quantitative determination of the parameters such as temperature and pressure field over an entire range of operation as a function of different operating parameters by Marangopoulos et al. [126] and Zeqiri et al. [127,128]. Kim et al. [129] studied the effect of ultrasound irradiation on the temperature and pressure distribution inside sonoreactor. Four different solvents have been investigated at various ultrasonic power for predicting energy density. Figure 6 shows the temperature change with respect to the sonication time at different ultrasonic power namely 300 and 450 W. In all liquid media, the temperature increases with time. However, the differences in the physical and thermodynamic properties of all liquid media are the reason behind the variation of the temperature trends with respect to time.

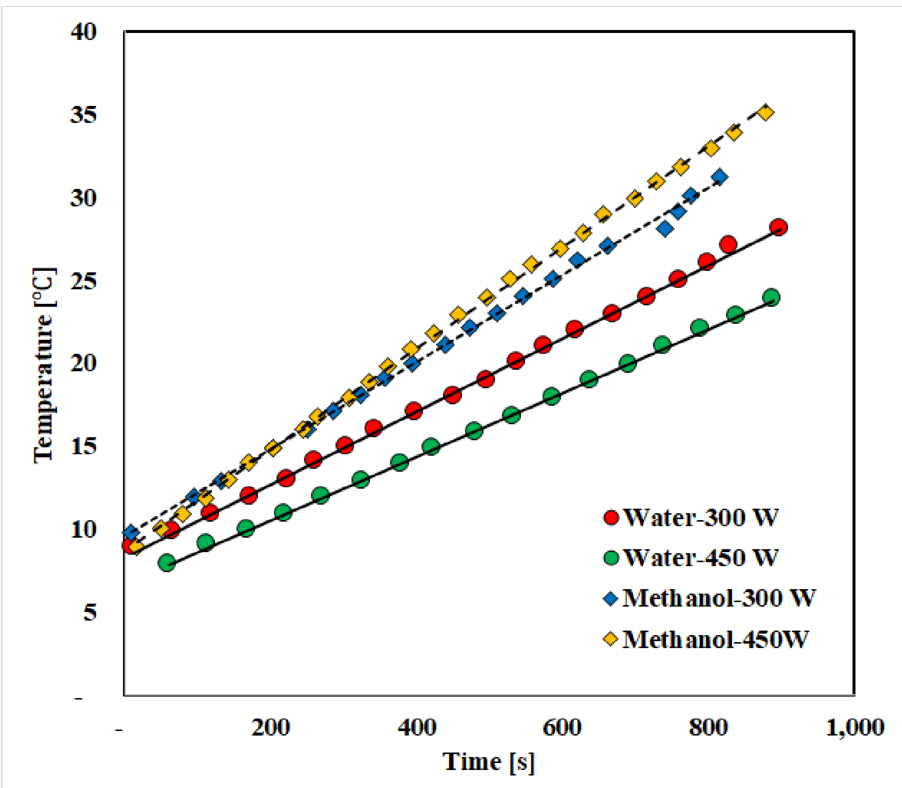


Figure 6: Temperature rise in various solutions with different ultrasonic power, reprinted from [129]

The effect of the liquid bulk temperature is scarce in the literature and the precise mechanism of this effect remains unclear. The liquid bulk temperature has a significant effect on bubble temperature and the hydrogen production rate in return. The liquid bulk temperature is very important as it is considered the surrounding medium of the acoustic cavitation bubbles. In fact, when bulk liquid temperature increases, the bubble temperature increases leading to liquid-vapor pressure increases and more vapor is trapped inside the bubble. However, increasing the bulk temperature will make the bubbles collapse less violent in turns affecting the decomposition process of water molecules causing fewer active radicals. Then the combination of these two important factors should lead to an optimum liquid bulk temperature at which the maximum hydrogen production rate is achieved.

4.5 Bubble temperature

The bubble temperature is one of the important parameters that affect the mole fraction of the produced hydrogen. The maximum bubble temperature is associated with two operational conditions such as the frequency and the acoustic amplitude. Merouani et al. [118] reported the amount of H_2 production with respect to bubble temperature with different gas based models. It can be seen that there is an optimum hydrogen rate recorded in the range 5000-7000 K. The higher the bubble temperature, the higher the amount of hydrogen production as per Table 7. The results revealed that at a low acoustic amplitude and high frequency, the amount of hydrogen production is higher, while the hydrogen production is lower at a high acoustic amplitude and low frequency. On the other hand, to attain the maximum bubble temperature at the end of bubble collapse, higher acoustic amplitude and low frequency should be applied [118].

Table 7: H_2-production [Mol] at bubble temperature [K] by Merouani et al. [118]

Bubble Temperature [K]	**1500**	**2000**	**2500**	**3000**	**3500**	**4000**	**4500**	**5000**
H_2 Production [mol]	2×10^{-33}	1×10^{-25}	2.1×10^{-24}	3.5×10^{-21}	0.5×10^{-19}	7×10^{-18}	6.3×10^{-18}	5.1×10^{-15}

Merouani et al. [116] performed a comprehensive numerical study of hydrogen production using acoustic cavitation bubbles in water. Figure 7 (a) presents the effect of ultrasound frequency on the hydrogen production rate in case of air as a dissolved gas. While Figure 7 (b) depicts the production rate of hydrogen with respect to ultrasound frequencies at different acoustic intensities [W/cm^2].

To sum up this section, the overall generation of H_2 is controlled by the amount of water vapor trapped inside the bubbles. To quantify this amount of water vapor, a series of preliminary numerical and experimental studies need to be performed. A large number of parameters, including frequency and intensity, need to be explored to develop a sufficient understanding of the phenomena. Furthermore, optimization and regression/statistical analysis need to be conducted to examine the optimum point and the most significant parameter that would give the maximum H_2 production rate.

In the next section, recent numerical modeling and simulations concerning sono-reactors are presented.

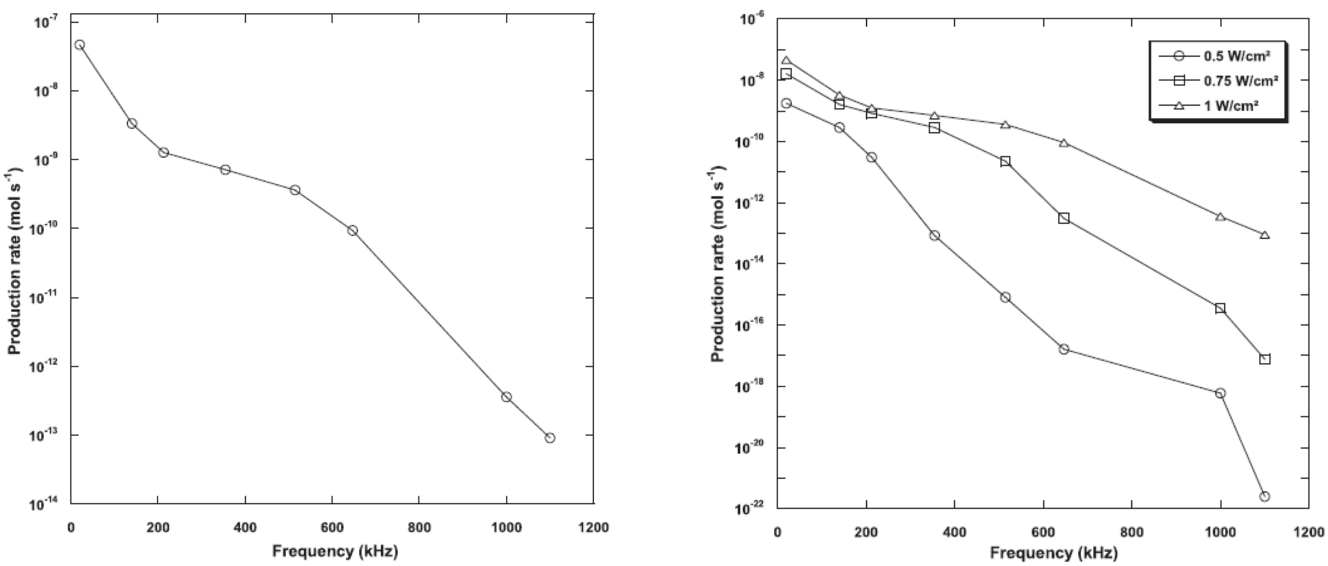


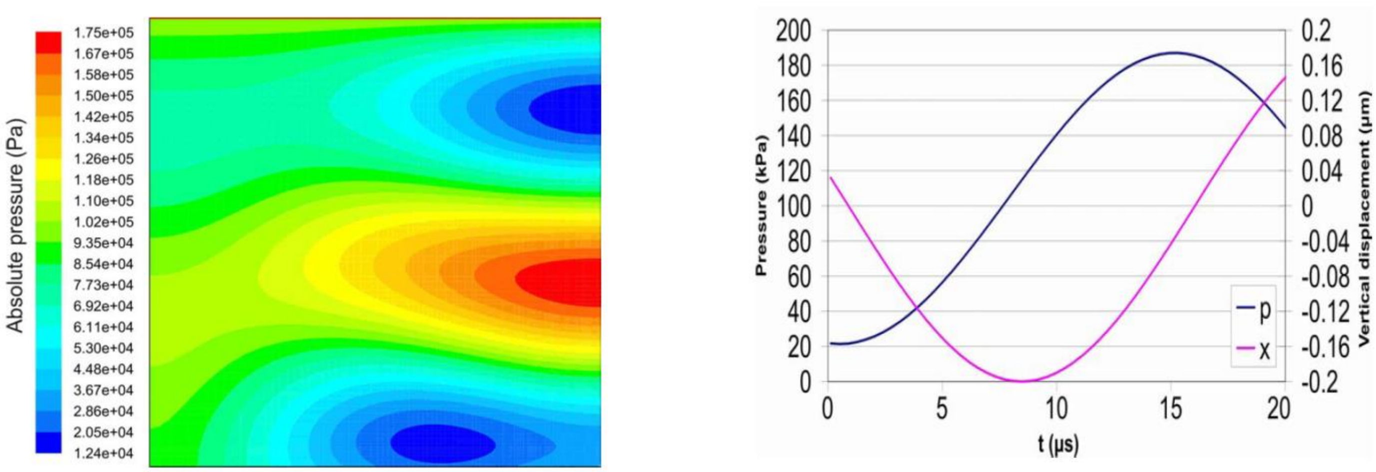
Figure 7: (a) The effect of the ultrasound frequency on hydrogen production rate, (b) the effect of acoustic intensity on hydrogen production rate by Merouani et al. [116]

5. Recent numerical modeling studies

In this section, recent numerical modeling and solution for the *Sono-Hydro-Gen* approach is presented starting from the hydrodynamic modeling for the gas inside bubble in liquid medium considering simulation about bubble behavior at different ultrasonic frequencies. Then we will be turning to some numerical simulation of the sono-reactor for characterizing the flow and the acoustic fields within the sonoreactor.

5.1 Hydrodynamic modeling

Hydrodynamic modeling and solution for the gas inside a bubble in a liquid medium is subjected to ultrasound waves triggers solving the *Navier-Stokes* equations for the gas inside the bubble. The conservation of mass for the gas inside the bubble assuming that the bubble has a symmetrical and spherical shape and the governing equations associated with the gas trapped inside a bubble subjected to ultrasound waves are introduced including mass, momentum, and energy are given by Kim et al. [130]. Numerical simulation of a near wall bubble collapse is performed by Osterman et al. [131] in an ultrasonic pressure field.



(a) **(b)**

Figure 8: (a) Oscillating pressure field in the domain; (b) the pressure fluctuation at the center of the bottom (blue) and the bottom displacement (pink) by Osterman et al. [131]

This numerical simulation has considered a 2-*D* and axisymmetric model. A pressure field is generated with the bottom of a container oscillating at 33 kHz. In this study, a validation of the model is successfully achieved by comparing a bubble collapsing near the oscillating wall as compared to the experimental work done by Philipp and Lauterborn [132]. Results considering the pressure contour oscillation and the pressure fluctuation are reported in Figure 8 (a) and (b), respectively. The comparison is made in terms of the dynamics sequence of the cavitation bubble collapse with respect to time. A sequence of the acoustic cavitation bubble captured in an experiment that is conducted by Philipp and Lauterborn [132] and a numerical work done by Osterman et al. [131]. The differences between the experimental and numerical simulation is that at the end of the collapse, it can be noticed that the differences lie on the bubble shapes and the bubble position. This can be attributed to that the numerical simulation did not consider the phase changes and the experimental work has some uncertainties due to the gravitational effects. Another difference can be found between both experimental and numerical work is that the counter-jet resulted from the bubble collapse is not captured by the numerical simulation, this is also attributed to that the phase change has not considered in the numerical simulations.

Many research studies conducted to investigate the acoustic cavitation bubbles. The cavitation bubbles can be characterized by the dynamics of oscillations and maximum pressure and temperature inside the bubbles before the collapse. Rooze et al. [133] performed an overview for characterization of acoustic cavitation bubbles reporting some recent experimental reports for characterization of the bubbles. In the textbook by Yasui [134], a comprehensive illustration is included for helping readers to understand the phenomenon of the acoustic cavitation and bubble dynamics.

5.2 Sonoreactor modeling

CFD simulation is performed on the acoustic cavitation in a crude oil upgrading sono-reactor and prediction of collapse temperature and pressure of a cavitation bubble by Niazi et al. [135]. In this study, ultrasonic waves introduced to liquid water contained in a sonoreactor via an ultrasound probe to investigate the pressure distribution.

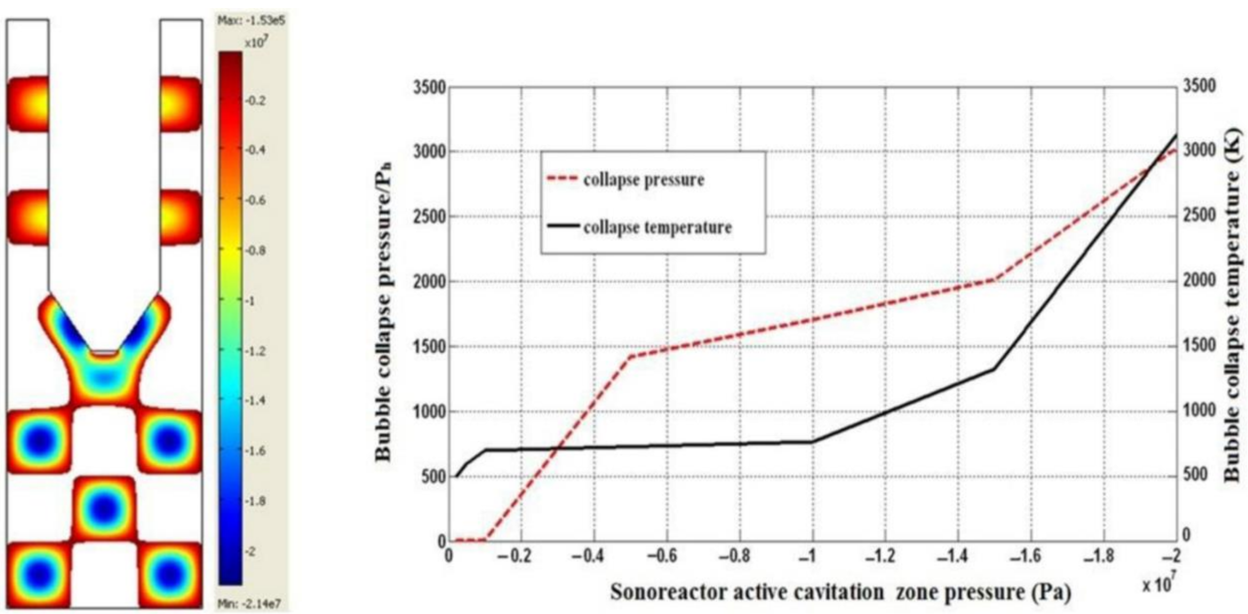


Figure 9: Active cavitation zones simulated by CFD technique for the reactor filled with saturated crude oil at temperature of 25 ℃ by Niazi et al. [135]

The experimental data is utilized from the Hielscher Ultrasound Technology website for a sono-reactor filled with water at 20 kHz and 2 kW. In the same study, CFD analysis of acoustic cavitation in a crude oil sono-reactor and prediction of collapse temperature and pressure of the cavitation bubble is conducted as well. Figure 9 presents the numerical results simulated to show the active cavitation bubbles zones in the sono-reactor filled with saturated oil at a bulk temperature of 25℃. The acoustic pressure threshold for acoustic bubbles is estimated to be 0.153 MPa with an initial oil bubble size of 10 µm. On the other hand, the figure is also showing the collapse pressure and temperature of the generated acoustic cavitation bubbles while crude oil is the working medium. The collapse pressure and temperature may go up to several thousands of Pa and Kelvins, respectively. The temperature and pressure fields due to the collapsing of bubbles under ultrasound conditions are predicted by Kim et al. [130] via the solution of *Navier Stokes equations* for the gas trapped inside a bubble. They compared the pressure profile of four different liquid mediums. The pressure profiles were different from each other; this can be attributed to the difference of the sound velocity and the density of each medium. They reported the pressure profile and temperature contours of water as seen in Figure 10.

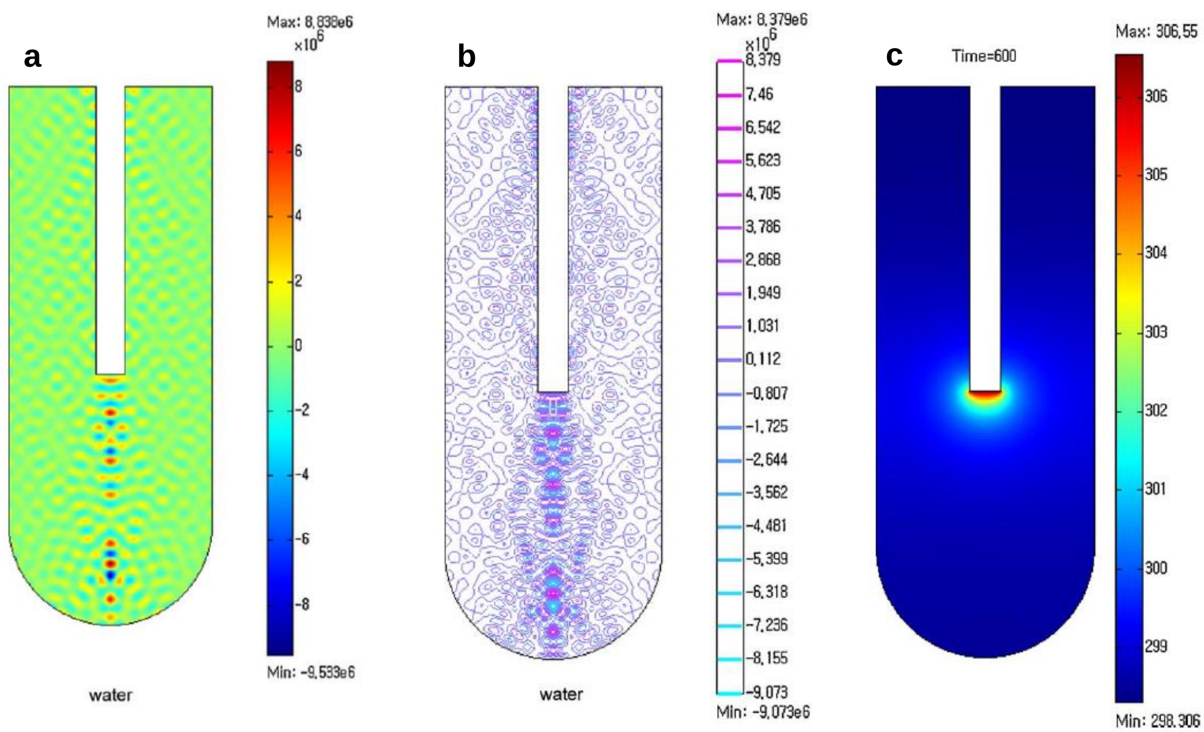
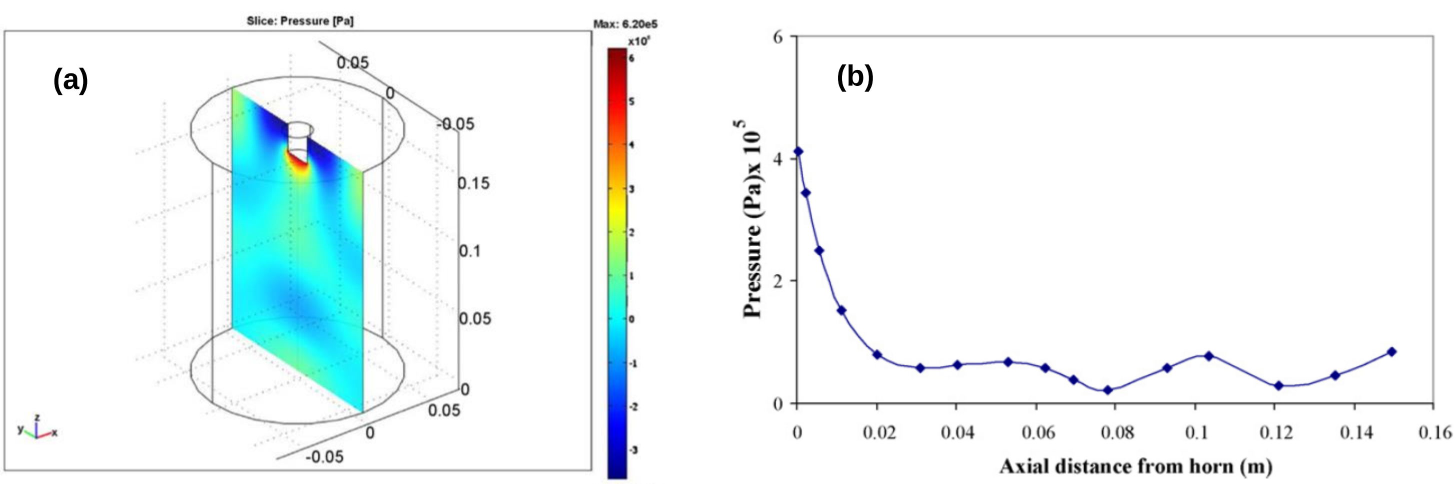


Figure 10: Pressure profile (a), pressure contour (b), and temperature contour (c) with ultrasonic power of 300 W, the liquid medium is water by Kim et al. [130]

Generally, the hot spot zone usually takes place near to the tip of the ultrasound probe. From the results of pressure profile, it can be concluded that the pressure profile oscillates starting from the probe tip to the way down to the bottom of the container. At an acoustic power of 300 W, the maximum and minimum pressure are recorded at 8.838 and -9.533 Pa, respectively. The temperature increased while increasing the acoustic power or the irradiation time. Sutkar et al. [136] performed a numerical analysis of the theoretical prediction of cavitational activity distribution in a sono-reactor. Numerical simulation is carried out and compared with experimental investigations. A 2 cm diameter ultrasonic probe with a maximum power of 240 W and a frequency of 20 kHz is been immersed in a cylindrical water bath (D= 13.5 cm × H= 17.5 cm). The results presented the variation of the pressure distribution inside the sono-reactor as seen in Figure 11. The pressure contours of the vertical transducer and its corresponding pressure amplitude in the axial direction of the ultrasound probe are shown in Figure 11(a) and (b), respectively. It is well recognized that the maximum pressure amplitude is close to the tip of the transducer probe and the pressure tremendously decreases in the way to the bottom of the reactor. The pressure contours of the longitudinal transducer and the corresponding pressure amplitude in the axial direction of the ultrasound probe are shown in Figure 11(c). Pressure fluctuation is observed along the length of the probe in the x-direction at z = 0.095 m as presented in Figure 11(d).



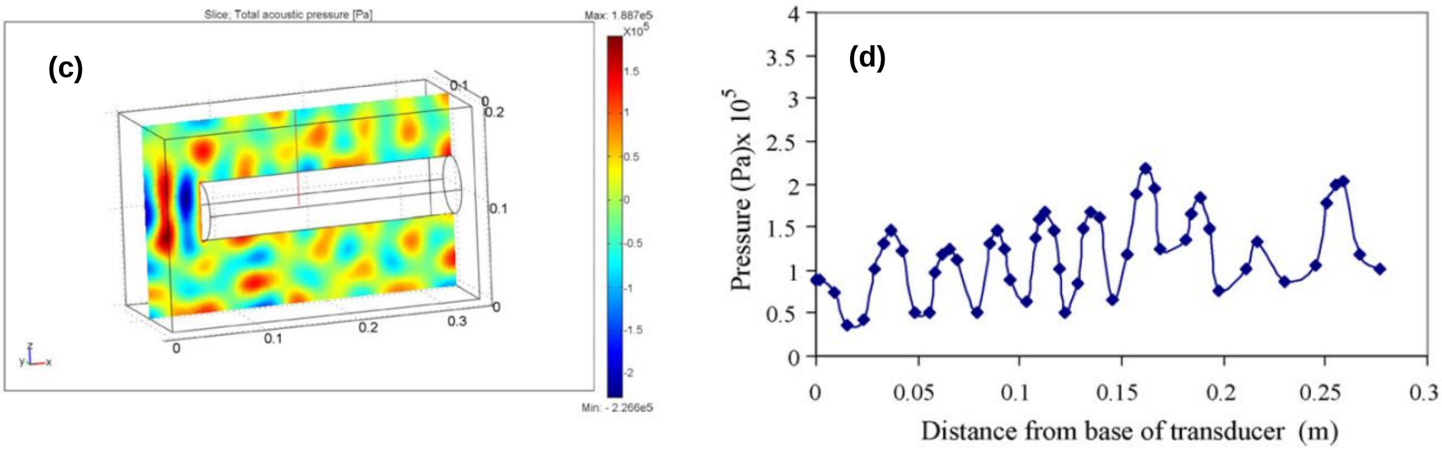


Figure 11: Pressure field distributions inside the sonoreactor; (a) pressure contours of the vertical transducer, (b) Axial pressure amplitude distribution, (c) pressure field of longitudinal transducer (d) radial pressure amplitude and the direction of transducer at z=0.095 from the bottom by Sutkar et al. [136]

Many research studies considered the improvement of the acoustic and flow fields of the sono-reactor. Wei [137] who performed a numerical simulation to design and characterize an ultrasonic transducer to overcome disadvantages of traditional transducers. Wang et al. [138] and Memoli et al. [139] performed characterization studies and improvement of a cylindrical-type sono-reactor.

In the next section, different experimental configurations have been summarized and reported. Analysis of the most important findings is quantified coherently.

6. Experimental configurations and analysis

An ultrasound-induced cavitation bubbles can be a source of acoustic waves due to bubble oscillation. The production of these sound pressure waves can be attributed to two reasons; the first reason is that these pressure waves is a result of the bubbles collapse, whereas the second reason is that these pressure waves are produced from the interaction between the bubbles, the wall and the reflected ultrasound waves from the walls. It is not yet clear that the production of these sound pressure waves is due to which of these reasons. Therefore, further experimental investigations should be carried out. An overview of different experimental configurations and recent experimental work procedure and their significance in understanding the *Sono-Hydro-Gen* production approach will be presented.

6.1 Different experimental configurations

There are three main configurations of sono-reactors as shown in Figure 12, the ultrasonic transducer horn or probe (Type-A), the ultrasonic transducer bath (Type-B) and the indirect irradiation ultrasonic bath (Type-C). In case of the ultrasonic horn, the transducer is immersed inside the liquid container, the ultrasound waves are introduced from the horn tip with a diameter smaller than the acoustic wavelength, consequently, the acoustic cavitation bubbles generated. While in case of the ultrasonic bath, it is mainly used for cleaning purposes, where the ultrasonic waves are introduced at the bottom of the liquid container. The generated bubbles are strongly affecting the ultrasound waves and the acoustic intensity. The decrease in the number of bubbles causes an increase in acoustic intensity in a tight liquid container.

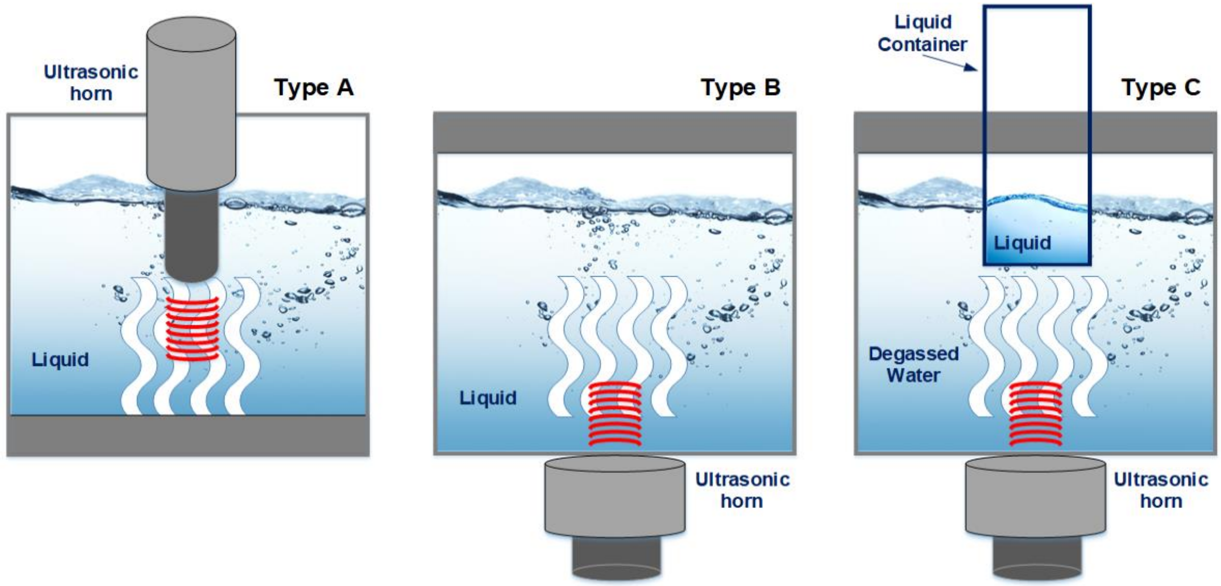


Figure 12: Different experimental configurations of the sono-reactor to generate acoustic cavitation bubbles

An indirect irradiation of ultrasound waves is also possible as shown in Figure 12(c). This configuration consists of an ultrasonic bath within which a small water container. The ultrasonic bath is filled with degassed water so as not to form bubbles [140]. The degassed water can be obtained by reducing the ambient pressure using a vacuum pump or by boiling. The benefits of this configuration are that the number of bubbles decreases with time because of degassing of bubbles leading to a decrease in gas concentration of the liquid inside the bath. The transducer emitting ultrasonic waves to travel through the degassed water until reaching the small liquid container. As known, the presence of bubbles in the bath will attenuate the acoustic intensity travels to the small liquid container. Therefore, the decrease in the number of bubbles in the bath will increase the acoustic intensity in the small container. Recently, Yasui et al. [134] recommended that the liquid surface inside the small water container should be aligned with the same level of the degassed water in the liquid bath in order to obtain the same irradiation condition.

Traveling and standing waves governing equations associated with the ultrasonic transducer immersed in the sono-reactor are summarized and well-illustrated by Kinsler et al. [141]. They provided an intensive illustration of different wave shapes such as a plane sound wave traveling through a liquid medium. Furthermore, a spherical wave can be formed if the acoustic wave source is point source that emits an acoustic wave into a liquid medium. The authors gave also an example considering a circular plane disc emits an acoustic wave into a liquid medium. In fact, the circular disc is acting similarly to the tip of the ultrasonic probe that emits ultrasound waves to the liquid medium inside the sono-reactor. The equation governing the spatial distribution of the pressure amplitude of a circular disk emits an acoustic wave into a liquid medium; the acoustic pressure distribution can be given as follows on the axis of symmetry:

$$P_a(x) = 2\,\rho_0\,c\,\nu_0 \left|\sin\left(\frac{\pi}{\lambda}\left(\sqrt{x^2+a^2}-x\right)\right)\right| \tag{3}$$

where ν_0 is the velocity amplitude of the vibrating disc, λ is the wavelength of the acoustic wave, x is the position from the disc to the point of measurement on the axis of symmetry and α is the radius of the circular disc. The disc is acting similar to the tip of the ultrasound horn or probe that emits ultrasound waves to water inside the sono-reactor. The generated acoustic cavitation bubbles by ultrasound are significantly affecting the density and sound velocity in the medium. In general, the density, the sound velocity, and the acoustic pressure amplitude decrease because of the generation and presence of bubbles under an ultrasound probe. As a matter of fact, the decrease in acoustic pressure amplitude has been studied and can be found in [142].

Koda et al. [143] compared 3 (three) different experimental setups for the sake of calibrating the sonochemical efficiency of different sono-reactor. The experimental setup (a) is built, operated and tested in the National Institute of Advanced Industrial Science and Technology (AIST). An ultrasound transducer of 45 mm is mounted at the bottom of a water bath to sonicate a sample of a volume of 50 cm^3 as seen in Figure 13(a). While the experiments of Nagoya University and Shiga University of Medical Science are presented in Figure 13 (b) and (c). All experiments are used to create a standard method to calibrate the efficiency of the sono-reactor and sonication efficiency and results of this comparison study are presented in the next section.

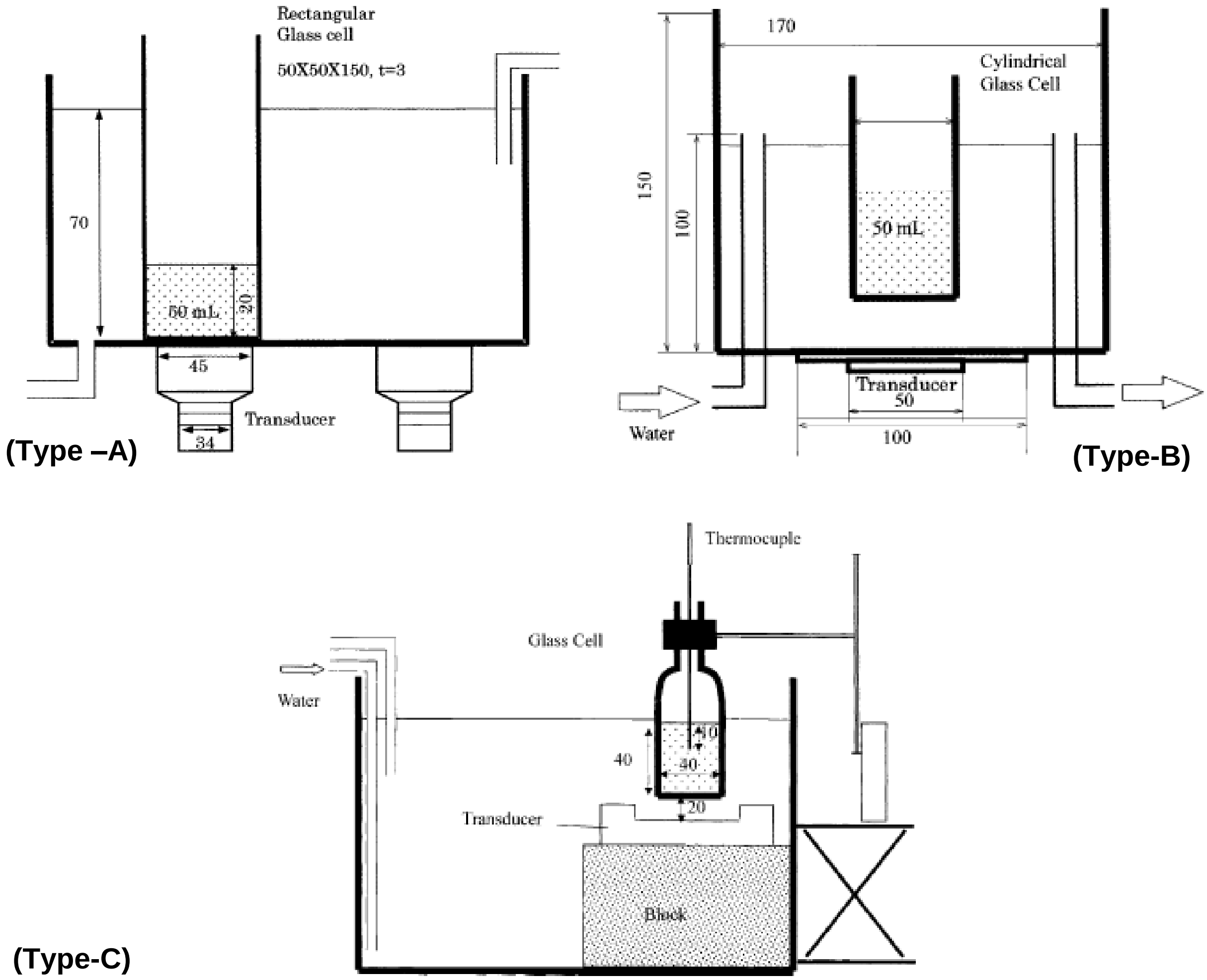


Figure 13: Different experimental configuration of sonoreactors in three different laboratories (a) AIST 96k, (b) Nagoya 130k and (c) Shiga 200k by Koda [143]

6.2 Recent experimental reports

An optimization of a sono-reactor (Type-A) subjected to a frequency of 20 kHz was investigated numerically by Klima et al. [144] and compared with experimental results. The second significant important parameter affecting the sono-reactor performance is the acoustic power intensity, which comes after the ultrasound frequency. In this study, the effect of acoustic power intensity on the sono-reactor characteristics and the ultrasound fields are studied. In case of low intensity, the prediction of the intensity distribution is well simulated, while the opposite in case of high intensity, the estimation of the intensity distribution is more complex. Figure 14 (b) presents the intensity distribution around the tip of the ultrasound probe. It is found that the higher intensity takes place close to the probe tip. Figure 14 (b) presents a comparison between the experimental results and the numerical analysis and it can be seen that both are fitted closely.

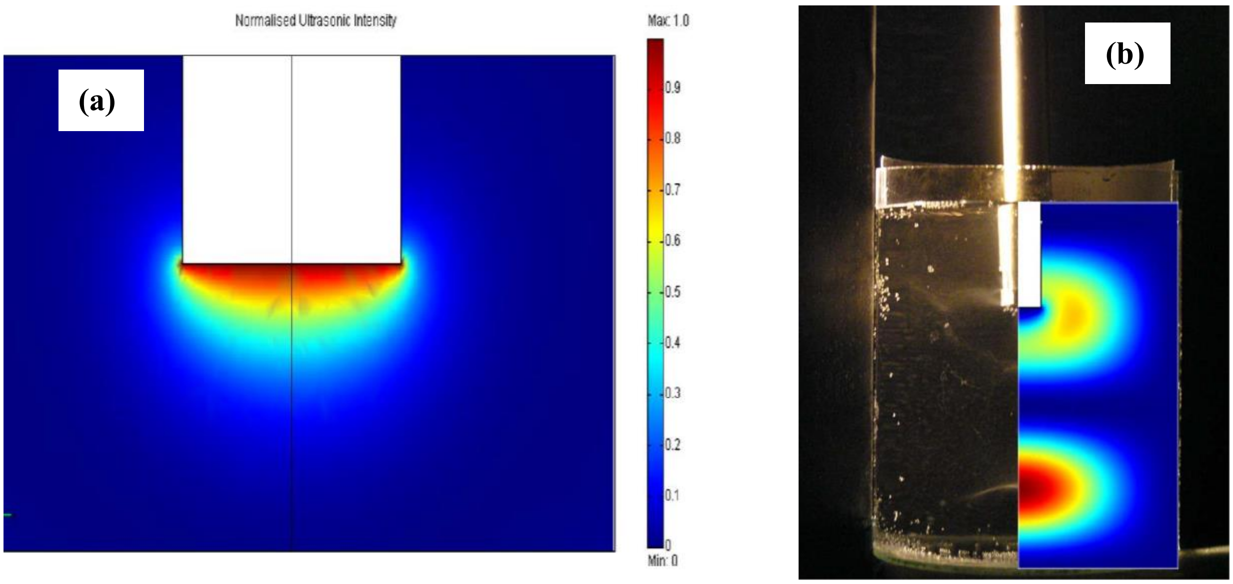


Figure 14: (a) Detail normalized ultrasonic intensity distribution at the ultrasonic horn tip, (b) comparison between the experimental sonoreactor (water, 20 kHz, ultrasonic power = 10 W) and the predicted intensity distribution for the same geometry by Klima et al. [144]

Son et al. [25] performed an experimental investigation on the acoustic emissions spectral using Type-B experimental configuration. They considered different experimental parameters including the liquid height and transducer power. Figure 15 (a) presents their experimental setup of the sono-reactor, which is consisted of an acrylic cylindrical sono-reactor (11 cm diameter and 110 cm height) with a transducer type piezoelectric transducer PZT. A 36 kHz frequency transducer mounted at the bottom of the sono-reactor. The water container is filled up with water at different liquid heights such as 100 cm. A power meter is mounted at the exit of the ultrasonic transducer controller to control the power input. A hydrophone is used to record the acoustic emission spectra and it is fixed at the mid-point of the sono-reactor. They investigated 3 (three) different electrical input power and they reported the total relative sono-chemiluminescence H_2O_2 generation and the calorimetric heat power. They reported that the H_2O_2 generation is 10.7, 30.6 and 25.6 μM at 30, 60, 90 Watts, respectively. Figure 15 (b) shows the effect of increasing the transducer power on the Sono-chemiluminescence images. The water inside the sono-reactor is mechanically vibrated. The results showed that at low acoustic input power bright zone appeared as an indication for a traveling wave and a standing wave is observed in the remaining part of the sono-reactor. At an average acoustic power, the bright conical zone is reduced and the stripes are concentrated along the sono-reactor axis. Whereas, at high acoustic input power, the bright zone is reduced because of a cloud of bubbles is formed near to the transducer, note that the transducer is mounted at the bottom of the sono-reactor. They reported a very important observation that higher acoustic power is not significantly affecting the hydrogen production yield, but it can be a reason of emitting acoustic emissions at harmonic frequencies. The more the bubbles are generated, the more the acoustic energy will be attenuated.

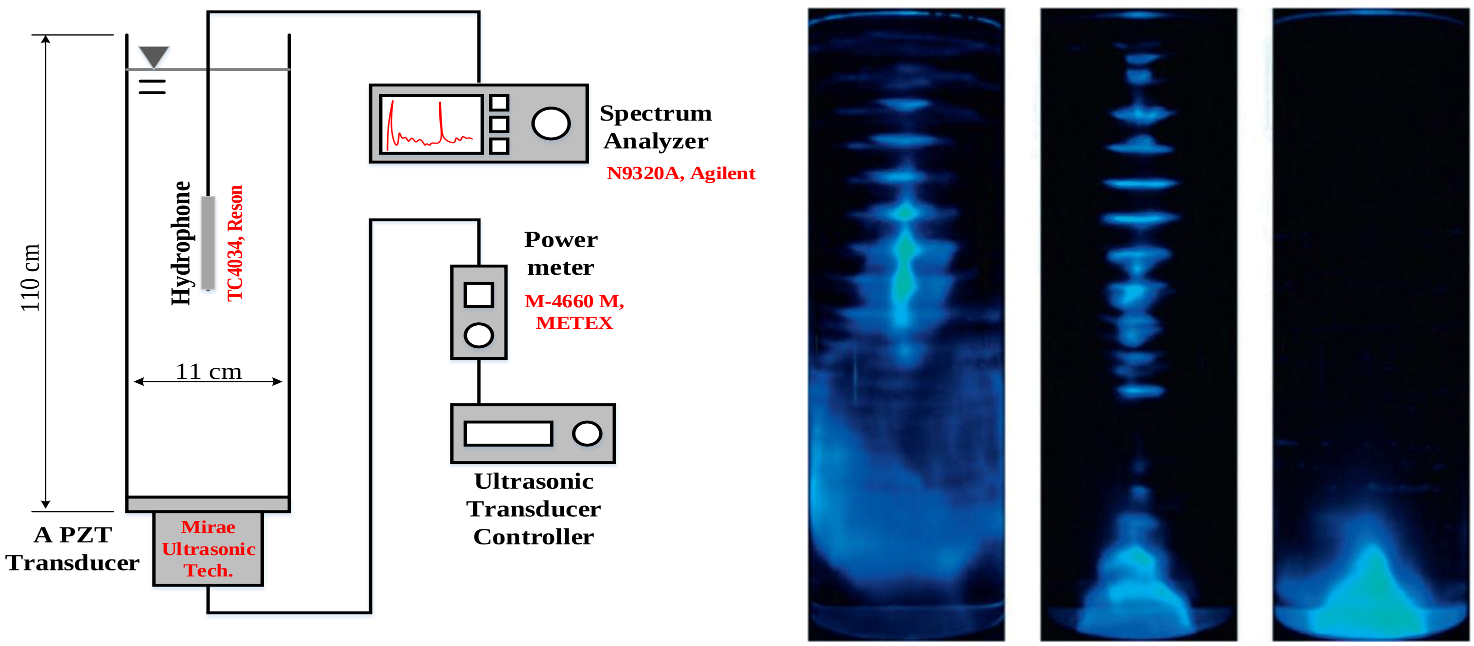


Figure 15: A redrawn experimental schematic of water sonoreactor (a); The Sono-chemiluminescence images under different input power for 30, 60 and 90 W (b) by Son et al. [25]

A recent experimental work performed by Merouani et al. [23] studied different methods for estimating the active bubbles in a type-B sonoreactor. Experiments involving *Sono-Hydro-Gen* and H_2O_2 production was carried out using an ultrasonic reactor containing 300 ml of distilled water. They conducted the experiments in cylindrical water-jacketed glass reactors. They reported some experimental procedures including that water temperature should be kept at 25°C by a water jacket recirculation around the cylinder. They reported that the bubble radius and H_2O_2 production rate are highly ultrasonic frequency dependent.

A different configuration has been suggested by Cotana et al. for studying water photo-sonolysis for hydrogen production. H_2O_2 and H_2 are the main products of this sonochemical reaction mechanism because of the recombination of the highly reactive radicals, produced from the dissociation of the water molecules at the first chemical reaction. They reported the sonochemical reactions steps in Table 8, corresponding to H_2 and H_2O_2 production. The experimental photo-sonolysis reactor consisted of a rectangular reactor with one glassed side to introduce photonic energy into the reactor. It had 3 (three) main ducts as highlighted in the figure with two piezoelectric transducers mounted in the bottom of the reactor. In order to generate the ultrasonic field in water, two piezoelectric transducers connected to the ultrasonic transducer controllers were attached to a power meter. The transducer generated the ultrasonic waves at a frequency of 22.5 kHz with a minimal input power of 50 W only. The *Sono-Hydro-Gen* experiments were carried out using the following procedure; first, the sono-reactor was filled with 0.1L of distilled water, second, the water above the water surface was injected with an inert gas, thirdly, the water was subjected to the ultrasonic actions at different pressure conditions namely, 1.0, 1.5, 2.5 atm. They performed a parametric study analysis to investigate the effect of the sono-reactor pressure on the hydrogen production rate.

Table 8: Sonochemical reaction steps by Cotana et al. [145]

$\mathbf{H_2O \rightarrow H^* + OH^*}$	(1)
$\mathbf{H^*+OH^* \rightarrow H_2 + O}$	(2)
$\mathbf{OH^*+OH^* \rightarrow H_2O_2}$	(3)
$\mathbf{H_2O_2 \rightarrow H_2O + \frac{1}{2} O_2}$	(4)
$\mathbf{H^*+OH^* \rightarrow H_2O}$	(5)

Figure 16 presents the hydrogen production in µMol with respect to time (in minutes). The results show a linear relationship between the produced hydrogen and time. Furthermore, the highest production rate took place for 1.0 atm pressure condition and the production rate of hydrogen decreased as the pressure inside the sono-reactor increased. This could be attributed to as the sono-reactor is pressurized, the acoustic cavitation bubbles cannot be oscillating freely which in turns reducing the amount of heat absorbed by the bubbles and affecting the hydrogen production rate.

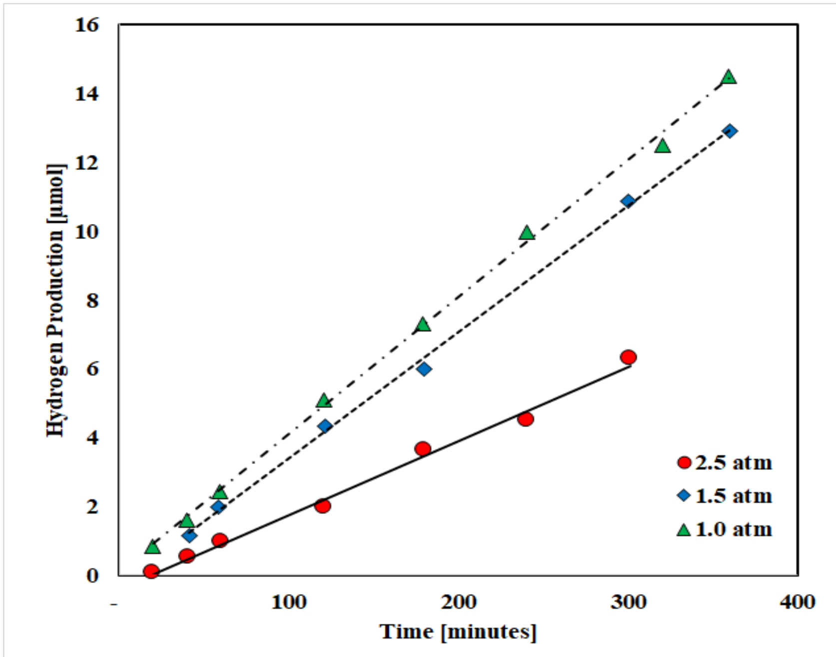


Figure 16: Hydrogen production versus time for different pressure conditions reprinted from [145]

The performance and efficiency assessment criteria of the sonoreactors are presented in the next section. Detailed description in view of the calculation procedure of the sonochemical efficiency based upon the energy density, ultrasonic power dissipated in the liquid medium inside the sono-reactor and cavitational energy. A comparison is made regarding the sonochemical efficiency from previous reports and studies in the literature.

7. Performance assessment criteria

In order to scale-up sonochemical reactors for industrial use, one needs to investigate the efficiencies and the factors affecting the sonochemical process. The most important two parameters for developing a performance and efficiency criteria are the energy density and the ultrasonic power dissipation. In fact, energy density E_{US} is related to the temperature change of the liquid medium during irradiation time t_{US} that can be measured in a flask filled with the required liquid to estimate the "real" power of ultrasound. This experiment was primarily suggested by Zarzycki et al. [146]. The energy density with the irradiation time and reaction volume is calculated using the following equation as follows:

$$E_{US} = P_{US} . \frac{t_{US}}{V_l} \quad (4)$$

A standard method to calibrate the sonochemical efficiency of sono-reactor was initially reported by Mason et al. [147]. They carried out the sonication experiment using distilled water at 25℃. They performed a calibration analysis and concluded the ultrasonic power dissipation into a liquid can be calculated by the following equation:

$$P_{US} = C_{pUS} \frac{\Delta T}{\Delta t} \tag{5}$$

where m_{US} is the mass of water in kg, C_{pUS} is the specific heat capacity of water at a constant pressure of 4.19 kJ/kg/K and ($\Delta T/\Delta t$) is the temperature rise per second. For the study, 50 cm^3 of water is used at the initial room temperature. The temperature rise is monitored by immersing a thermocouple in a solution and is held at the half height of the solution. The results of the ultrasonic power obtained by the ultrasonic power-dissipation equation. Continuous introduction of ultrasound waves results on a temperature increase in the range from 5 to 10 Kelvins. The potassium iodide solution (KI) is used to quantify the amount of energy absorbed and it can be defined as when ultrasound is irradiated into an aqueous KI solution, the I^- is oxidized to I_2 then they recombine together to form I_3^- as following: $I_2 + I^- \leftrightarrow I_3^-$, the concentration of KI solution is (0.1 mol-dm^{-3}). In a later study by Asakura et al. [148], the efficiency of the sonochemical process is quantified and it can be given by the ratio of the number of reacted molecules m_I over the ultrasound energy as follows:

$$\eta_{sonochemistry} = \frac{m_I}{E_{US}} = \frac{C_I}{P_{US} t_{US} / V_I} \tag{6}$$

where C_I, t_{US} and V_I refers to the concertation of the ion I_3^- of a (0.1 mole dm^{-3}) KI solution, the irradiation time of ultrasonic energy and the solution volume, respectively. The production yield of CI_3^- against the ultrasonic energy (kJ) which is calculated by multiplying the calorimetric ultrasonic power in (W) by the sonication time (s). The production yield of CI_3^- corresponding to the sonochemical effect when ultrasonic power of 1 W is introduced to a liquid for 1 s can be obtained by Figure 17 (a). Actually, the CI_3^- values correspond to different ultrasound frequencies can be obtained by Figure 17 (b). In case of different water volumes, Figure 17 (c) shows that the CI_3^- is independent of water volume. Here we draw attention that the sonochemical efficiency is not laboratory dependent and not a volume or quantity dependent. On the other hand, in case of the sonoelectrochemical process, the sonoelectrochemical efficiency can be described as the ratio of the theoretical energy consumption of the electrolysis process (W_{Theo}) over the summation of the electrochemical energy consumption aided with ultrasound (W_{UsA}) and the amount of energy consumed by the ultrasonic transducer (W_{Us}) which is can be described as follows [24]:

$$\eta_{Sonoelectrochemical} = \frac{W_{Theo}}{W_{UsA}+W_{Us}} \times 100\% \quad (7)$$

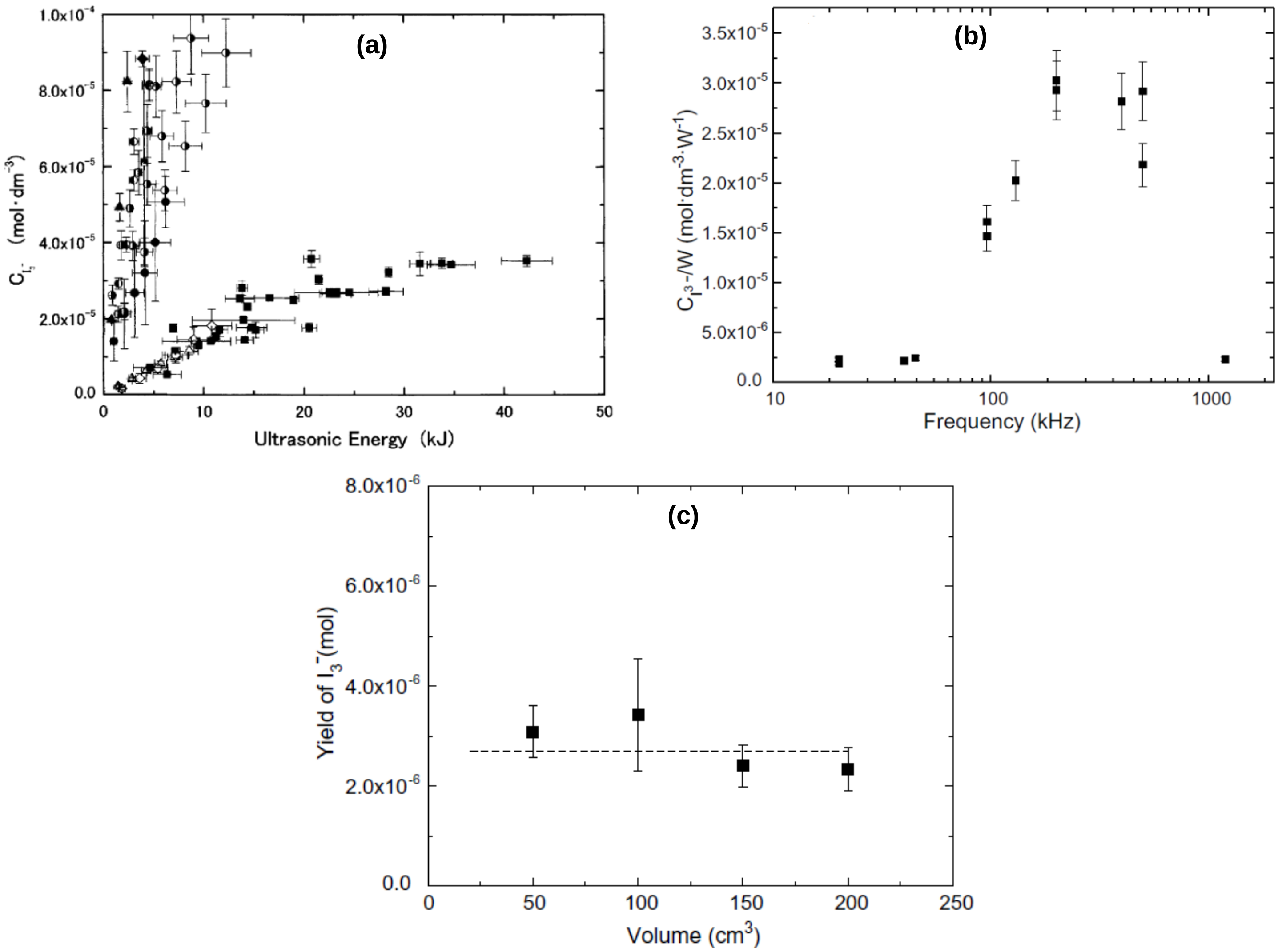
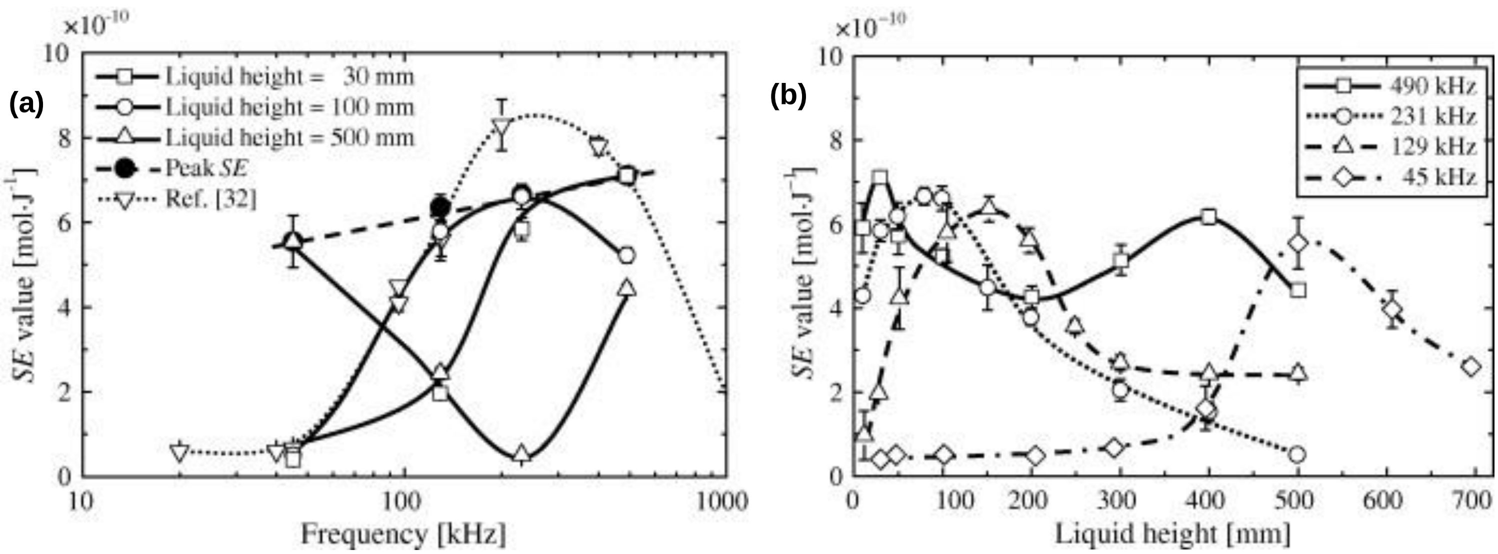


Figure 17: The chemical effect per unit power (mol.dm^3.W^{-1}). The frequency dependence of chemical effects per unit power concentration CI_3^- by Asakura et al. [148]

The sonochemical efficiency has been also quantified by Asakura et al. [148] as a function of ultrasonic frequency and the liquid height inside the sono-reactor. They examined 4 (four) different frequencies including 45, 129, 231 and 490 kHz while varying the liquid height from 10 to 700 mm, which is corresponding to the different water volumes. Distilled water was used in this experiment saturated with air at 298 K. The sonochemical efficiency of the reactor was evaluated by potassium iodide (KI) dosimetry and calorimetry. The sonochemical efficiency results were reported with respect to the ultrasonic frequency and the liquid height inside the sono-reactor. Figure 18(a) and (b) present the effect of the ultrasonic frequency and the liquid height of the sono-reactor on the sonochemical efficiency, respectively. At different liquid heights, the efficiency is reported with respect to the different ultrasonic frequencies. It can be seen that for the height of 100 mm, the findings are in good agreement with the experimental work performed by Koda et al. [143] at different ultrasonic frequencies. The sonochemical efficiency was also examined with respect to the liquid height, which behaved as one or two peaks for each ultrasonic frequency. The ultrasonic irradiation time varied in the range of 60-1,800 seconds, based upon the liquid height and the maximum temperature rise of the KI aqueous solution after introducing the ultrasonic waves.



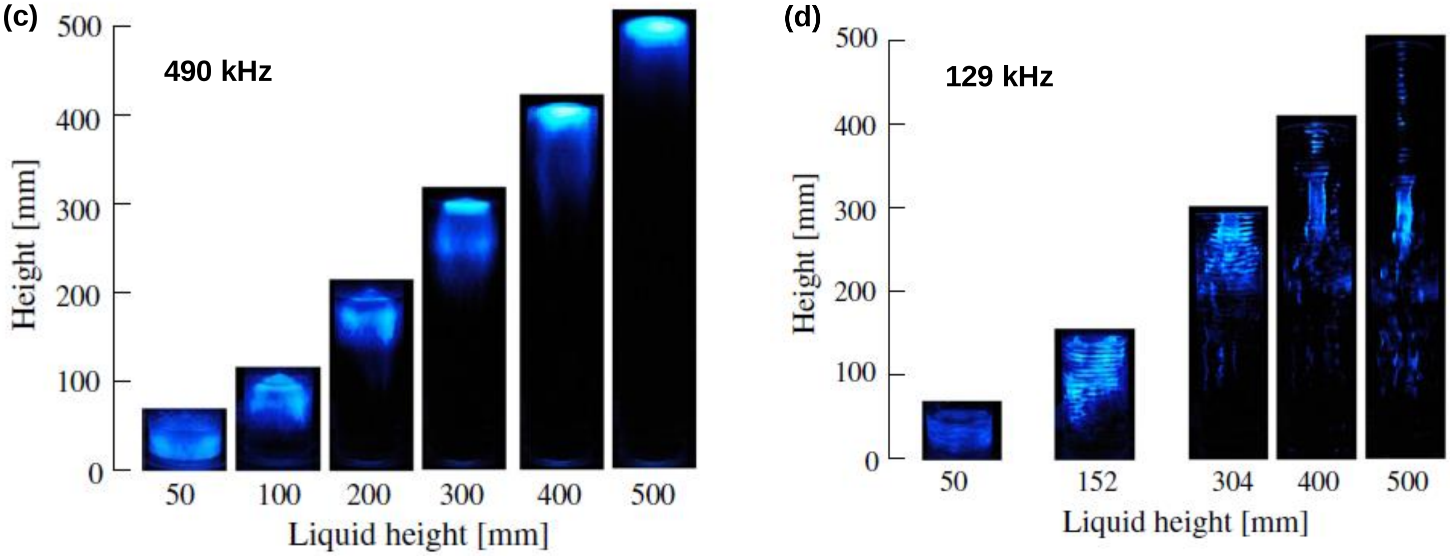


Figure 18: The effect of ultrasound frequency and the sonoreactor's liquid height on the sonication efficiency by Asakura et al. [148]

Sonochemical luminescence was investigated in the purpose of visualizing the sonochemical reaction field. In Figure 18 sono-reactor images were taken at different ultrasonic frequencies and liquid heights. In (c) high intensity is visualized near the top of the acrylic glass pipe and the highest intensity is found at the 400 liquid height. In case of a lower frequency of 129 kHz, the ultrasonic standing wave is observed in the way traveling from the transducer at the bottom to the top of the sono-reactor. Furthermore, the standing wave is elongated while increasing the liquid height.

A comparison was made between two experimental studies conducted by Koda et al. [143] in 2003 and Asakura et al. [148] in 2008 on the sonochemical efficiency using the KI dosimetry method. They reported the ultrasonic frequency dependence of the sonochemical efficiency (SE-value) for KI-solution oxidation as shown in Table 9. As shown, the maximum sonochemical effects were recorded at an ultrasonic frequency range of 200-500 kHz. Both studies gave very similar values to the SE.

Table 9: Ultrasonic frequency dependence of the sonochemical efficiency (SE-value) for KI oxidation

***f* [kHz]**	**Koda et al.** [143]**, 2003**	**Asakura et al.** [148]**, 2006**
	SE_{KI} (mol J^{-1}) $\times 10^{10}$	SE_{KI} (mol J^{-1}) $\times 10^{10}$
20	0.6 ± 0.02	-
40	0.6 ± 0.02	-

45	0.67 ± 0.06	5.5 ± 0.6
96	4.5 ± 0.2	-
96	4.1 ± 0.2	-
130	5.6 ± 0.4	6.4 ± 0.3
200	8.3 ± 0.6	6.7 ± 0.6
400	7.8 ± 0.2	-
500	7.1 ± 0.2	7.1 ± 0.1
1200	0.64 ± 0.3	-

Here it is emphasized and after comparing the sono-reactor efficiencies from several studies that, the sono-reactor efficiency is not affected either by the laboratory scale experiments or by the sample volume. It can only give some information and predict the average ultrasonic energy induced by the evaluation of KI oxidation.

Investigation of acoustic cavitation bubble energy in a large-scale sono-reactor was performed by Son et al. [112]. They investigated cavitational energy distribution inside a sono-reactor consisting of an acrylic glass container with dimensions of (W= 0.6 m × L=1.2 m × H= 0.4 m). The ultrasound PZT transducer with a diameter of 5 cm was placed at the center-side of the container. The maximum transducer capacity was 400 W. The sono-reactor was filled with 250 L of tap water. The calorimetry method was not used here to calculate the input power because of the large heat dissipation due to the large surface area of the container; consequently, the input power was measured using a multi-meter. An ultrasonic cavitation meter was used for measuring the average cavitation energy in the sono-reactor. The maximum power capacity was 240 W while examining different ultrasonic frequencies namely 35, 72, 110 and 170 kHz. The maximum cavitation energy was recorded at an ultrasonic frequency of 72 kHz at varying ultrasonic probe locations corresponding to different irradiation distances as seen in Figure 19(a). At a higher ultrasonic frequency of 170 kHz, the cavitation energy was approximately 1.0 W.

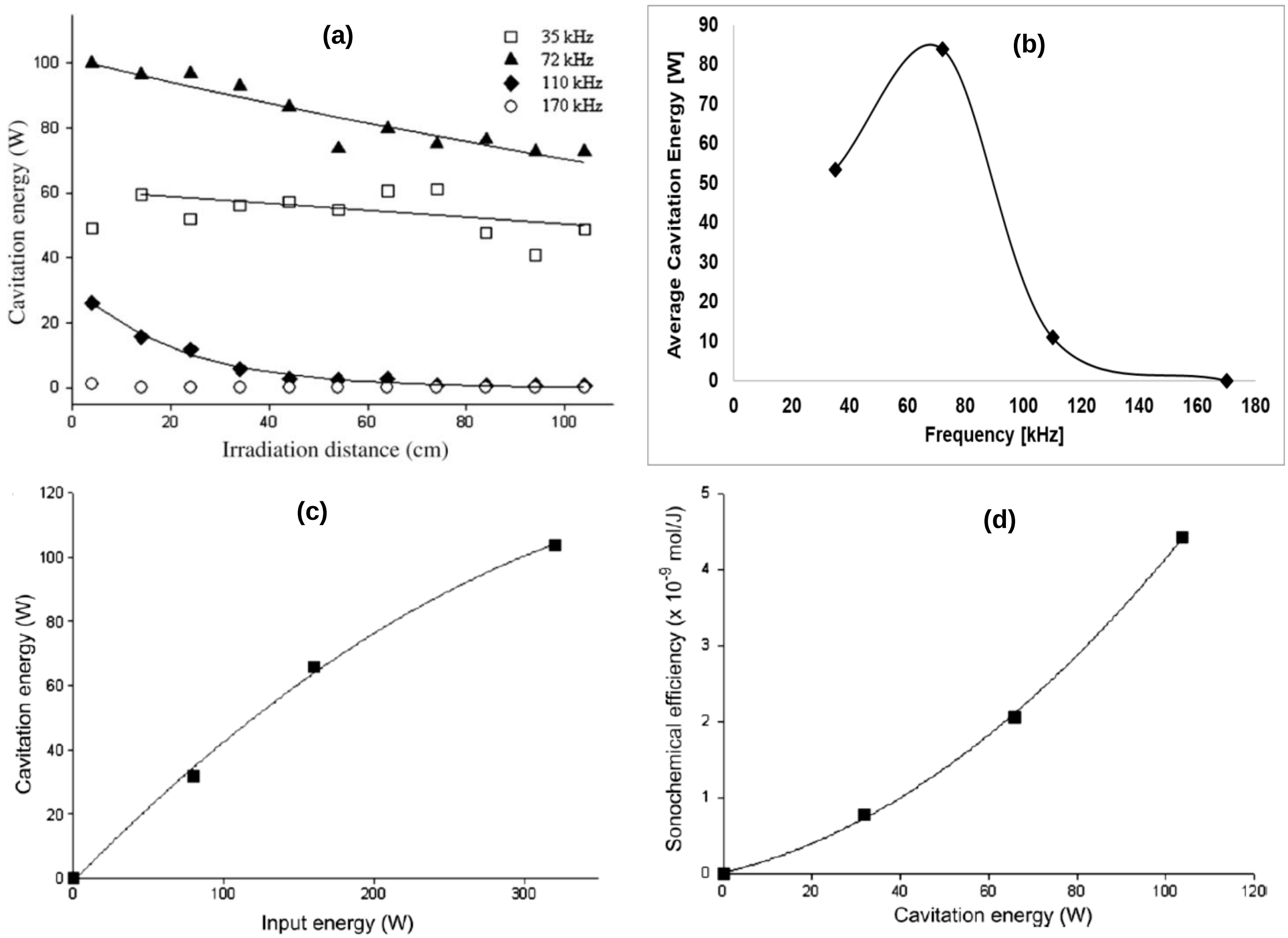


Figure 19: Empirical relationships (a) Cavitation energy *vs*. irradiation distance at different ultrasonic frequencies, (b) Average Cavitation Energy *vs*. Ultrasonic Frequency, (c) Cavitation energy and input energy (W) and (d) Sonochemical efficiency and cavitation energy (W) by Son et al. [112]

Son et al. carried out experimental analyses to quantify the relationship between the average cavitation energy and the ultrasonic frequencies as shown in Figure 19(b). They reported the average cavitation energy corresponding to different ultrasonic frequencies. It can be noticed that the cavitation energy is much higher in case of low frequencies. The results in this study showed that at higher frequencies more than 100 kHz, the ultrasonic waves could not propagate efficiently. Furthermore, they developed an empirical correlation to quantify the cavitation energy and sonochemical efficiency which can be quantified by the following equations:

$$Cavitation\ energy = -0.0008\ (input\ energy)^2 + 0.4699\ (input\ energy) \quad (8)$$

$$Sonochemical\ efficiency = 0.0003\ (cavitation\ energy)^2 + 0.0140\ (cavitation\ energy) \qquad (9)$$

To correlate the relationship between the cavitation energy and the sonochemical efficiency, the input energy is used. In Figure 19 (c) and (d), the empirical correlation between the cavitation energy against the input power and the sonochemical efficiency against the cavitation energy are presented, respectively.

Next section gives a snapshot on how previous reports quantified the hydrogen production rate in case of water electrolysis.

8. Hydrogen production quantification

This area of research also requires a great attention as a very few studies quantified hydrogen generation via *Sono-Hydro-Gen* process. Hydrogen production quantification plays an essential role to understand the effect of several factors affecting operating conditions and for the sake of upgrading the sono-reactor to a large or an industrial scale.

In the case of water electrolysis, it is well known that this process is a costly and highly energy demanding technology with a consumption rate of 4.5-5 kWh per m^3 of H_2. The overall efficiency of the process is in the range of 30-40%. One solution to reduce energy consumption is to use an ultrasound aided-water electrolysis system. In a combined study of water electrolysis with ultrasound, Symes [149] performed an experimental investigation of water electrolysis in the presence of an ultrasonic field. They used a graduated glass column filled with water to collect gas bubbles. The hydrogen gas formed was measured from the water level change in the column. The equation of the efficiency of hydrogen gas production is illustrated as follows:

$$\boldsymbol{\eta}\ (\%) = \frac{V_{real}}{V_{ideal}} \times 100\% \qquad (10)$$

where V_{real} is the actual hydrogen gas production per unit of time read by a digital hydrogen flow meter (Red-y compact flowmeter (L/min)) [150]. While the ideal hydrogen production rate for the water electrolysis process can be given as follows:

$$V_{real}(cm^3) = \frac{SIt}{nF} \times \frac{RT}{P} \qquad (11)$$

where *S* is the stoichiometric coefficient, *I* is the applied current, *t* is the electrolysis time, *n* is the number of electrons transferred, *F* is the Faraday constant (96,484 C/mol) and *P* is the pressure in Pascal. R and T are the universal gas constant and the temperature in Kelvins, respectively. The results revealed that the production efficiency of hydrogen is increased at a range of 5-18% while the energy efficiency is increased by 10-25%. In another combined study by Zadeh [150], the ultrasound aided water electrolysis technology is used and the results showed an improvement in production efficiency by 4.5% and energy efficiency by 1.3%.

To sum up, few studies considered hydrogen production quantification only in case of ultrasound-aided water electrolysis. However, methods to quantify the amount of hydrogen from the *Sono-Hydro-Gen* process is not yet clarified.

In the next section, challenges associated with the *Sono-Hydro-Gen* process are clarified and reported. These challenges related to each of the following: the complexity of the acoustic field, enhance understanding of the chemical reaction mechanism, intensity distribution inside the sono-reactor, attenuation of ultrasound waves, factors affecting hydrogen production rate, energy conversion and scaling-up the sono-reactor.

9. Ultrasound and Energy consumption

In case of the ultrasound efficiency, the overall ultrasound power reaches the liquid is approximately 80-90% due to the sequence of losses in the way between the power plug to the liquid as illustrated in Figure 20 [151].

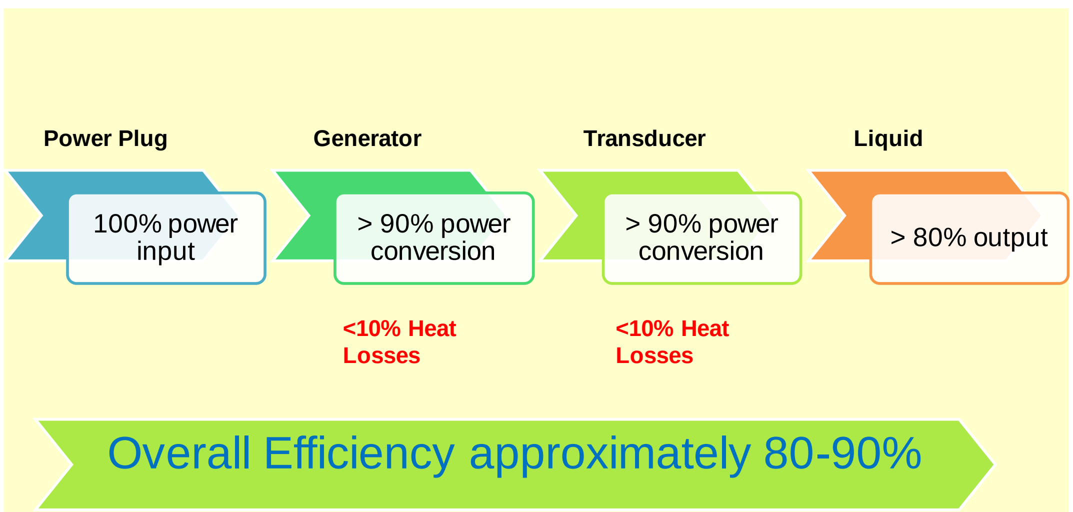


Figure 20: Overall ultrasound power attenuation from the plug to liquid and ultrasound efficiency

On the other hand, the energy consumption of the sonochemical (*Sono-Hydro-Gen*) process is compared with the sonoelectrochemical process in terms of the ultrasound frequencies and the energy consumption and they are all summarized in Table 10.

Table 10: Energy consumption of hydrogen production by different technologies

Technology	Process	Frequency [kHz]	Energy consumption	References
Electrochemical technology	Electrolysis process 0.1 M KOH	N/A	6.3 kWh/m^3 H_2	Zadeh [150]
Sonoelectrochemical technology	Electrolysis process aided with ultrasound 0.1 M KOH	20 kHz	5.12 kWh/m^3 H_2	Zadeh [150]
Sonochemical Technology	*Sono-Hydro-Gen* process	20 kHz	0.8 µM/min	Venault [70]
Sonochemical Technology	*Sono-Hydro-Gen* process	300 kHz	0.83 µM/min/W	Fischer et al. [152]
Sonochemical Technology	*Sono-Hydro-Gen* process	1000 kHz	0.42-0.68 µM/min/W	Buettner et al. [153]
Sonochemical Technology	*Sono-Hydro-Gen* process	1000 kHz	0.22 µM/min	Margulis et al. [122]

10. Challenges for *Sono-Hydro-Gen* reactor design

In this section, the need for further research studies is presented. The main challenges associated with the efficient design and operation of sono-reactor are summarized in Figure 21.

The challenges are revealed from previous and ongoing studies considering sono-reactor experiments and given in details as follows:

1) The acoustic field: has not been fully understood both numerically and experimentally. The acoustics field is highly complex due to several reasons such as, for example, the inhomogeneous spatial distribution of bubbles. Consequently, the speed of sound is time and position dependent. Furthermore, the liquid container's walls are vibrating due to the pressure oscillation of the liquid medium. These vibrations emit acoustic waves back to the liquid medium, which will significantly affect the acoustic field.
2) Mechanism of H_2 production: this challenge lies in advancing our fundamental understanding of the mechanism of hydrogen production as the mechanism is not yet understood and the most reported suggestions are controversial. Water-sonication experiments are still under investigation. Some research studies hypothesized that most of the H_2 production is generated

inside the bubble during the gas phase. Whereas others contradict this hypothesis by reporting that, H_2 is formed on the bubble shell by the recombination of the generated radicals from the water dissociation.

3) Intensity distribution: many difficulties are found in the determination of the intensity distribution inside the sono-reactor. Determination of ultrasonic intensity is well investigated in case of low-intensity ultrasound. However, the challenge comes in when high power ultrasound is used when exceeded the cavitation threshold. Other difficulties in the determination of ultrasonic intensity distribution lie on that ultrasound is mainly characterized by the power delivered to the system determined by calorimetry. Furthermore, the ultrasound field inside the sono-reactor is known to be a non-uniform sound field because most of the ultrasonic energy is consumed at the tip of the ultrasonic probe.
4) Attenuation of the sound waves in the liquid medium. As ultrasound waves are emitted and propagate through the liquid medium, the acoustic intensity of the sound decreases along the distance from the ultrasonic probe to the bottom of the liquid container. The attenuation takes place due to reflection, refraction, and absorption of the sound waves by the generated bubbles. As a result, active and passive acoustic zones will exist inside the sono-reactor. It is essential to understand the effect of these changes within the sono-reactor with probing the possibilities to reduce these attenuation effects.
5) Factors affecting the ultrasonic production of hydrogen: a number of factors affecting ultrasonic production including, the frequency of ultrasound, type of dissolved gas, acoustic power, and the bulk liquid temperature. Data concerning the effect of ultrasound frequency and the water bulk temperature are very limited and scarce in the literature and need to be furtherly investigated.
6) Energy conversion: one of the most important factors in the development of an industrial process is the energy conversion from ultrasound waves to the required effect. The importance lies in the change of liquid properties as per the ultrasonic characteristics, which is considered very limited in the literature.
7) Large-scale sono-reactor: some researchers highlighted that the implementation of industrial sono-reactors have not reached fully commercialization yet because most of the research studies are considered the lab-scale sonoreactors and they do not provide enough information about the optimum design and optimum operating conditions of the sonoreactors. It is

necessary to understand the sono-reactor characteristics including the cavitational energy and acoustic intensity distribution.

8) Quantification of the produced hydrogen: few studies have considered quantification of the hydrogen production rate. Detailed quantification is essential to understand clearly the effects of different operating conditions, which is necessary for upgrading sono-reactors from the lab-scale to industrial or conventional scale.

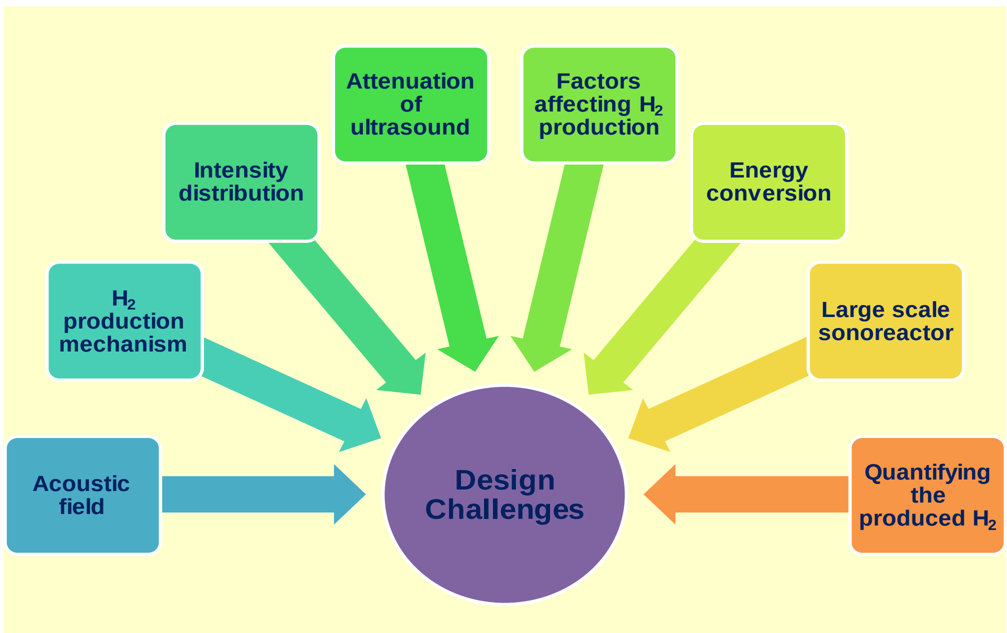


Figure 21: Challenges associated with sonoreactors

11. Conclusions

In this article, a comprehensive review is provided on the background and principles associated with *Sono-Hydro-Gen*. This approach is an interesting and expanding field as it is considered one of the benefits from utilizing the power of ultrasound using specially designed sono-reactor.

To obtain the optimum design of the sonoreactor at a range of ultrasonic frequencies, comprehensive numerical and experimental investigations should be performed to address lab-scale and industrial concerns to achieve the optimum design of the *Sono-Hydro-Gen* reactor. Some insightful lab-scale results are reported on the effect of some important factors influences the hydrogen production rates from the *Sono-Hydro-Gen* reactor. These factors are crucial to initiate a strong database to establish an industrial sono-reactor.

Promising results are obtained to date from both lab-scale experiments and numerical tests, however, it worth noting that the *Sono-Hydro-Gen* sonoreactor are still in the stage of testing, development and application commercialization. The key challenges, which need to be addressed for the development and application for a durable reactor for industrial application, include:

- Characterizing the flow field, the acoustic field of the acoustic cavitation bubble
- Characterizing the cavitational energy intensity distribution inside the sono-reactor
- Enhancing and characterizing the *Sono-Hydro-Gen* process
- Developing a low cost but effective sono-reactor geometry
- Optimizing the operating modes of the sono-reactor

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