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Review on the Development of Plasma Discharge in Liquid Solution

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Abstract. Review papers on the basic principles and applications of plasma discharge in liquid have been reported. However, the development of plasma discharge in liquid is not widely reviewed. The aim of this paper is to review on the elements involved in developing plasma discharge in liquid. An overview will be given of the various types of existing power source, working gas, dielectric barrier, electrode material used to produce plasma, and reactive species production inside plasma discharge.

INTRODUCTION

Plasma discharge inside liquid solution garnered much interest nowadays because it leads to interesting scientific problem and can be applied in a wide range of areas. Process involved in plasma discharge in liquid is slightly different compared to the atmospheric plasma discharge. There are three main factors that differentiate plasma discharge in liquid and atmospheric plasma discharge namely density of charge particles, dielectric strength, presence of ions and their mobility process in aqueous solution. Atmospheric plasma production is reported to use less energy compared to plasma discharge in liquid. It is because plasma discharges in liquid need greater electric field which is higher voltage input in order to breakdown the gas bubbles produce inside the plasma treated liquid. Although, plasma discharge in liquid need higher voltage, this treatment had been proved to be used in several important applications namely water treatment, chemical synthesis, plasma medicine, wound healing, plasma sterilisations, disinfection, surface treatment, material synthesis and also synthesis of nanoparticles [1]–[5].

There are two main theory of plasma discharge in liquid which are electronic theories and bubble breakdown theory [6], [7]. Electronic theory can be described as the breakdown of water by electrons that accelerate after the electric field is applied. During the reactions, the electrons collide with each other and become ionize, thus producing more free electrons. This reaction caused to the breakdown of water. Meanwhile, bubble breakdown can be described as formation of bubbles inside plasma discharge liquid[8]. During the discharge process, high field region produce by power source inside plasma will lead to heating process, and thus lead to liquid vaporization. During the vaporization of liquid, breakdown of gas will occur within each bubble. This process will lead to the production of reactive species inside the solution [9].

Hence, this paper intends to describe the fundamental theories to develop plasma discharge in the liquid. The main elements that will be discussed in this paper are the types of power input source and gas supply used to develop plasma, dielectric barrier, reactive species production inside plasma discharge, and lastly is the properties and durability of electrode material.

POWER INPUT, DIELECTRIC BARRIER AND WORKING GAS SOURCES

The common voltage power sources used in plasma device are alternating current (AC), direct Current (DC), pulsed, microwave, and radio frequency (RF) [6]. This power sources was connected to the electrode of the device in order to generate plasma inside or above the liquid solution. Different electrode locations lead to the

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different value for the initial power sources. It is because plasma inside the liquid solution needs more energy compared to the plasma above the liquid solution. During the treatment, plasma emits UV radiation and intense shock waves. As the high-voltage pulse ends, the plasma channel cools and transfers its thermal energy to the surrounding water, resulting in the formation of steam bubbles. The breakdown of bubbles inside the liquid solution required a higher number of electric field as reported by Bruce R. Locke [10]. Continuous measurement of voltage and current flow at the plasma discharge area can be detected by using current probe (Tektronix P6021) and a voltage probe (Tektronix P6015A) had been reported by other research [11].

Dielectric barrier discharge (DBD) is commonly known as low-temperature discharge and it is operating in atmospheric pressure. It was obtained between two parallel electrodes separated by some millimetres gap and it is excited by alternating current (ac) voltage with frequency in the range of 1–20 kHz. The advantages of this type of plasma is it can be used in surface processing and in plasma chemistry [12], [13]. There are two functions of dielectric barrier discharge namely it limits the amounts of charge transported by a single microdischarge and distributes the microdischarge over the entire electrode surface [14]. To achieve the desired advantages, the dielectric barrier material also is one of the crucial needs in the development of plasma device. It is because this barrier act as a shield used in order to limit the electrical current flows within the discharge channel and also to prevent the developing of arc discharge [15]. Dielectric material that are commonly used by previous researches are glass, quartz, polymer and ceramic material [6], [16]–[18]. These materials have low dielectric loss but high breakdown strength[12]. This dielectric material is located between two electrodes [13], [19].

Meanwhile, the common working gas used in plasma device are helium, neon, argon, nitrogen, oxygen air and the mixture of these gases [20]. Noble gas is frequently used as a working gas because they have low ignition voltage, has high gas thermal conductivity, has long lifetime of meta-stable excited atoms and highest electron energy level among all material and lastly because of the properties of noble gas which is inert. Inert gas prevents it from mixing with other monomers[15]. Besides, helium gas has the highest ionization energy compared to other noble gas which is 24.5eV [21]. Meanwhile, air plasma also can be used to replace inert gas as a working gas. The main advantages of air plasma compare to inert gas are it is cheaper that inert gas and also the system process are simpler. Unfortunately, air plasma only can be used in certain application only such as sterilization because it leads to acidification of plasma treated liquid [22]. Air plasma was reported to produce more reactive nitrogen species during the plasma process. On the contrary, different working gases yield different concentration of reactive species. O Galmiz et. al. proved this by identifying the effect of plasma working gas to the production of excited species [23]. Types of working gas used in this research are air, argon, nitrogen and oxygen. Meanwhile, the compositions of the reactive species are determined by using optical emission spectroscopy (OES). Results shows that oxygen and argon produce the same radical species during plasma process which are hydroxyl radical (OH), oxygen radical (O) and hydrogen radical (H). Although, the same radical species are produce, the different in the concentration for both gases are recorded. It was reported that hydrogen radical recorded the highest inside argon gas, and concentration of oxygen and singlet oxygen radical was the highest during oxygen plasma discharge [6], [24].

TYPE OF ELECTRODE

Electrode plays one of the important roles in the development of plasma discharge in liquid because it influences the ionic discharge formation during the treatment. Previous research shows that electrode affect the production of chemical species during the plasma treatment. M. J. Kirkpatrick in his research shows that nickel-chromium electrode is able to produce higher net rate production of hydrogen peroxide, and oxygen compared to platinum high voltage needle electrode. Meanwhile, stainless steel lead to certain negative chemical effect in liquid solution after treatment [25]. Besides, other research also state that platinum shows low production of chemical species but it was commonly used as electrode because it has the ability to generate hydroxyl radical (OH) without reaching mineralization state [26]. Meanwhile, T. Kaneko et. al. introduced new types of electrode in his research which is ionic solution that act as non-metal liquid electrode. Ionic liquid is used as a solution to produce static and stable plasma production. It is a type of solution that consists of positive and negative ions that has an extremely low vapor pressure and high heat capacity[1].

For plasma discharge in liquid device, the electrode will be located inside or above the liquid solution. Hence, there are a few electrode properties that need to be considered while choosing the electrode which are corrosion resistance, conductivity, hardness, electrode form, size, current load and toxicity [27]. Firstly, study about electrode lifetime need to be conducted before developing any plasma discharge in liquid in order to identify the corrosion resistance of the electrode. It is very important because, every electrode had its certain limit operating time. Performance of electrode needs to be investigated to ensure there is no particles release from the electrode while treatment was conducted. The factor affecting the erosion of electrodes by varying the solution conductivity value was investigated by P.Lukes et.at [28]. This study investigates the effect of liquid

conductivity to the erosion process of several types of electrodes which are platinum, tungsten and stainless steel. The electrode erosion was measured by using two methods which are measure the mass loss before and after the treatment and the second method was by analysed the surface morphology of the electrode by using scanning electron microscopy (SEM). There are two types of solution conductivities used which are 100 and 500uS/cm. Result shows that the electrode that recorded the highest amount of mass loss is tungsten while the least erosion material was platinum. Besides, it also shows that the solution conductivity affects the erosion process of electrode. The increment of solution conductivity leads to the higher value of electrode erosion.

Electrode can be divided into two groups of material which are inert electrodes and reactive electrode. Inert electrodes consist of graphite (carbon), platinum, gold, titanium and rhodium. Meanwhile, reactive electrode consists of copper, zinc, lead, silver and platinum wire. Type of materials that commonly used as electrode is copper, graphite, titanium, brass, silver, platinum, tungsten and nickel. Table 1 shows the advantages and the disadvantages of the electrode [27].

TABLE 1: Advantage and disadvantages of electrode material [27].

Electrode material	Findings
Copper	Have better strength and better electrical conductivity than silver. Copper are usually used as the electrode and also for electrical contact application.
Graphite and carbon	It can be applied as an electrochemical electrode because it has corrosion resistant compare to several metal materials. Advantages of graphite are, it is easily mould and low in cost.
Titanium	It is a non-ferrous metal which contains good corrosion resistance and also good fatigue properties. It also commonly used in electrochemical processes.
Brass	It is a combination of copper and zinc. It has lower conductivity compared to copper.
Platinum	It has lower contact resistance and also has high erosion compared to other material.

REACTIVE SPECIES.

Plasma discharge in liquid are able to generate an abundant of reactive species in liquid solution namely hydrogen peroxide, hydrogen, oxygen, hydrogen radical, superoxide ozone, hydrogen peroxide, hydroxyl radicals, nitric oxide, and nitrogen dioxide and it has been reported by previous researches [6], [7], [28]–[31]. Each radical species leads to different effect to the treated sample. Furthermore, some of these radical also contain higher number of oxidizing agent which has the ability to inactivate microorganism and also destruct organic compound inside water solution [32]–[36]. There are several factors that lead to the production of reactive species namely type of discharge, polarity of high voltage electrode, pH, conductivity, type of input gas, flow rate of input gas and humidity [37], [38]. Conductivity of a liquid plays significant role in plasma process because it effects the production of the reactive species [39]. Higher conductivity leads to bigger production of UV light which is increase the current inside the liquid[29]. Unfortunately, it decreases the value of formation of reactive species[30]. Besides, previous research stated that increment of conductivity of liquid lead to the increment of liquid temperature during plasma treatment. Hence, it is suggested to add external cooling circuit to prevent the temperature of liquid above 24 ◦C [40].

Reactive species can be classified into two group namely Reactive Oxygen Species (ROS) and Reactive Nitrogen Species (RNS)[41]. Examples of ROS are hydroxyl radical, atomic oxygen, ozone, and hydrogen peroxide. Meanwhile, the examples of RNS are nitrites, nitrates and peroxynitrites. The production of these reactive species caused to chemical effects during the treatment process namely direct and indirect reactions. Direct reaction defined as the process involved between the effect of radical species to the changes of chemical and biological of treated sample. In this process, the radical produced during plasma reaction will attack the target changes. Meanwhile, the indirect reaction can be defined as the changes occur to the pH and conductivity of the treated liquid sample during the plasma process. This change was reported to continue even after the plasma treatment was done, but it had certain lifetime depends on the plasma process. Hence, it is very important to identify the behaviour and sources of the reactive species in order to develop a high potential plasma discharge in liquid. To achieve this purpose, there are various techniques that can be used to detect the reactive species inside the plasma treated liquid namely measurement chemical probe [42], reactive species scavenger [43], [44], spin trapping reagent [45], UV-VIS spectrometer [24], [46], laser-induced fluorescence techniques [47], light emission spectroscopy [48], electron paramagnetic resonance spectroscopy [49], optical emissions spectroscopy (OES), Fourier transform infrared spectroscopy (FTIR) [46], electron spin resonance (ESR), Fluorescence spectrometer [24] and ion chromatography.

Chemical probe and reactive species scavenger are the easiest method that can be used to detect reactive species inside the aqueous solution[50]. For chemical probe, each type of reagent will give different effect to the different types of radical formed. Meanwhile, scavenger is a substance that usually used in mixtures in order to

remove or inactivate impurities. The effect of these measurements will be shown in term of changes of the colour reagent and changes in aromatic ring structure before and after the treatment. Among the reactive oxygen species, hydroxyl radical (OH) have the highest oxidation value with redox potential 2.7 V [51]. It plays an important role in decontamination process and chemical oxidation of organic molecules. So, it is very important to detect the production of this species inside the plasma treated liquid. Unfortunately, this species has short lifetime due to its high reactivity. It is reported that reactivity of species affect the lifetime of the species [2]. Besides, this radical species also are difficult to measure in aqueous solution because it has the ability to convert to the other reactive species such as hydrogen peroxide through recombination or other reaction. Example of common chemical probe used to detect hydroxyl radical is Terephtalic acid. Terephtalic acid (TA) is known as chemical probe and also OH scavenger [60]. It is because it has the ability to not react with other radical species namely oxygen (O_2) and hydrogen peroxide (H_2O_2) and it also reported as the best sensitive OH detection [42], [47], [52]. The OH radical can be detecting by the addition of OH at the aromatic ring of terephtalic acid. Unfortunately, chemical probe and reactive species scavenger cannot provide the exact concentration value of reactive species produce inside the plasma treated liquid. In order to quantify the concentration of reactive species produced inside the plasma treated liquid, the measurements are usually conducted by using UV-VIS spectrometer, fluorescence spectrometer or spin trapping resonance [45]. The overall examples of other chemical reagent that are commonly used for reactive species detection are summarized in table 2.

In addition, reactive species were also reported to cause changes in pH of the liquid solution after the treatment. However, the effect of reactive species radical on the pH is not widely studied [50]. Some research reported that plasma discharge in liquid cause to the acidic treated liquid after the treatment [53]. Wahyudiono et. al. reported the pH changes in solution after plasma treatment. His research was conducted in order to investigate the decolouration of Orange G, Orange II, Congo Red, and Naphthol Blue Black by using plasma. Through the investigation, result shows that the pH solution decreases when the input voltage was increased during the decomposition of dye molecule into organic acids[29].

TABLE 2: Types of chemical detection for reactive species detection

Types of chemical reagent	Type of reactive species.	Method of detection.
Amplex Red reagent	Hydrogen peroxide (H_2O_2)	It will produce red Fluorescent oxidation product if reacted with H_2O_2 after the treatment[45].
Titanium reagent	Hydrogen peroxide (H_2O_2)	The formation of hydrogen peroxide inside the liquid solution will caused the changes in reagent colour to yellow [9], [54], [55].
Griess reagent	Nitrate/ nitrite	This reagent was added into the water to detect the nitrate/ nitrite [45].
Terephtalic acid [Sigma Aldrich]	Hydroxyl radical (OH)	Converts terephtalic acid into 2- hydroxylterephtalic acid (HTA) through the addition of OH at the aromatic ring [24], [38], [52], [56]–[59]
Methylene blue	Hydroxyl radical (OH)	Methylene blue will turn colourless after plasma discharge if there is hydroxyl radical inside the solution [55], [60], [61]
Solutions of singlet oxygen Sensor Green [Invitrogen]	Singlet oxygen	The changes can be measured by using fluorescence spectrometer at the excitation and emission of 504 nm and 525 nm [24].
Nitrite and nitrate test kit [HACH]	To detect nitrite and nitrate	The formation of nitrite and nitrate inside the solution can be measured at the absorbance value 515 nm and 330 nm respectively [24].

SUMMARY

Plasma discharge in liquid is gaining interest from researchers because it can be widely applied in various areas. However, to the best of authors' knowledge, the study about the main elements involved in the development of plasma discharge in liquid is not widely studied. This impedes the development of plasma discharge in liquid because researchers need to perform preliminary tests prior the process of developing the plasma. There are two major challenges in developing plasma discharge in liquid namely process that lead the production of reactive species and also the electrode material and configuration. In reactive species case, the process that cause to the production of this species inside liquid solution is still inconclusive. It is reported that the bubbles breakdown in liquid lead to the production of reactive species. The higher the rate of gas bubbles,

the higher the concentration of reactive species inside plasma treated liquid. However, the factors that ignite the production of the bubbles inside the liquid solution and the effect of this bubbles to the production of reactive species is not fully understood. Meanwhile,, the investigation about electrode lifetime is very important. It is because for plasma discharge in liquid, the electrode will be located inside the liquid solution. Hence, the material with higher erosion resistance is needed for this device. It is very important because to avoid any particles release from the electrode while the plasma treatment is conducted. Hence, this review intends to describe the main fundamental element that involved in developing plasma discharge and further investigation about the problem listed will be conducted.

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