Plasma-Activated Water

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Plasma-activated water (PAW) is generated by treating water with cold atmospheric plasma (CAP) using controllable parameters such as plasma-forming voltage, carrier gas, temperature, pulses, or frequency as required. PAW is reported to have lower pH, higher conductivity, and higher oxidation-reduction potential when compared with untreated water due to the presence of reactive species. PAW has received significant attention from researchers over the last decade due to its non-thermal and non-toxic mode of action, especially for bacterial inactivation. This review summarizes the properties of PAW, the effect of various treatment parameters on its efficiency in bacterial inactivation along with its usage as a standalone technology as well as a hurdle approach with mild thermal treatments.

Keywords: cold atmospheric plasma ; microbes ; disinfection

1. Introduction

Food spoilage is defined as a change in any food product that leads to a significant reduction in its sensory qualities, such as color, texture, and overall smell, due to physical damage or chemical changes (e.g., oxidation), and thus rendering it unacceptable by the consumer [1]. These changes are mainly the result of microbial growth and metabolism in the food, which may lead to the production of enzymes that facilitate to reactions resulting in deleterious by-products affecting the food. These by-products vary in different types of food and can lead to adverse sensory properties, including the presence of slime, off-odors, and off-flavors. Bacterial strains associated with spoilage include *Pectobacterium carotovorum* [2][3], *Brochothrix thermosphacta* [4], *Clostridium perfringens* [5], *Bacillus spp.* [6][7], *Pseudomonas fragi* [8], *Pseudomonas fluorescens* [9], *Shewanella putrefaciens* [10], *Serratia liquefaciens* [11] and *Hafnia alvei* [12]. Food spoilage is a primary concern for food industries due to susceptible loss of shelf life and hence the economic losses followed by a long-term impact on consumer preferences. Nevertheless, food spoilage is also a threat to the environment as it leads to excessive wastage that end ups in the landfill, which does not contribute to sustainable living. This is supported by a survey conducted in 2018, which indicated that 30–50% of the total food produced exclusively by one country in a year ends up in the landfill, with the contribution from households, processing industries, food services, primary production sector and retails being 53%, 19%, 12%, 11%, 5%, respectively [13]. Minimizing food spoilage by employing multiple interventions might help not only the food industries but also the environment.

Another concern for the food processing industries and the regulatory authorities is food poisoning due to bacterial growth in food. Some bacterial strains are capable of producing toxins under certain conditions either in the food itself or inside the human body once live bacterial cells are ingested, while others are enteropathogenic and entero-invasive pathogens [14]. Few examples of foodborne pathogens include *Clostridium botulinum* [15][16], *B. cereus* [17], *Staphylococcus aureus* [18][19], *Listeria monocytogenes* [20], *Salmonella enterica* serovar typhimurium [21], *Salmonella* spp. [22], *E. coli* O157:H7 [23], *C. perfringens* [24], *Shigella* spp. [25], *Yersina* spp. [26] and *Campylobacter jejuni* [27]. Food poisoning has been a major public health concern, particularly regarding outbreaks affecting immunocompromised individuals and infants and thus may lead to adverse social and economic effects. Although many food-poisoning cases go under-reported due to quick recovery and almost minimum effect on healthy individuals, some still have adverse effects on immunocompromised individuals and infants [28]. Alternate disinfection technologies that do not employ thermal treatments or harmful chemicals could be valuable options for minimizing contamination and growth of microbial contaminants leading to either spoilage or food poisoning. Such sustainable technologies are of great importance to the vast ever-growing population with increasing demand of food across the globe.

Recently, the application of advanced oxidation processes (AOPs) for the decontamination of fruit and vegetables has been widely investigated. These technologies include electrolyzed water [29][30], gaseous ozone [3][31], UV light [32][33], and cold plasma [33][34]. One of these oxidation technologies is plasma-activated water (PAW), which is generated by treating water with cold atmospheric plasma (CAP) using controllable parameters such as plasma-forming voltage, carrier gas, temperature, pulses, or frequency as required. Plasma has been recognized as the fourth state of matter. It is the ionized gas usually produced when gas molecules are exposed to the electric field, forming reactive species and ions [35]. PAW has received significant attention from researchers over the last decade due to its non-thermal and non-toxic mode of action, which is mainly due to the reactive species that could react with the bacterial structural components and later organelles, leading to death [36].

2. Systems for PAW Generation

The fundamental method of generation of PAW involves operating a plasma generator inside the water to generate the ions, which lead to reactive species for bacterial inactivation. There are various combinations and models in the literature leading to difference in the final outputs; these are outlined in Table 1.

Table 1. The effect of different generation conditions and the characteristics of PAW**.**

Gas and Additional Features	Gap (Between Water Surface and the Upper Electrode)	AC Voltage and Frequency	Quantitative Changes after Generation Time	References
Grounded copper electrode (diameter 0.5 mM) on top and a capillary tube to generate bubbles	10 mM	3–6 kV, 3–10 kHz	pH changed from 6.75 to 3.77 and NO_2^- concentration changed from 3.77 μM to 8.686 μM in 15 min of activation	[41]
Plasma jet unit coaxial tungsten electrode and a quartz tube (diameter of 700 μM)	nil	6–10 kV, 7.0 kHz	Not determined	[42]
Plasma jet with RD1004 rotating nozzle	8.1-cM	voltage (295 V), air pressure (1990 mBar), and frequency (22.5 kHz)	pH changed from 6.5 to 3.1 and Oxidation-reduction potential (ORP) increased from 376.54 to 534.52 RmV in 5 mins	[43]
Atmospheric pressure plasma jet (patent atmospheric pressure plasma jet (APPJ))	0 mM	3.0 kV and 16 kHz	pH reduced from 7 to 3.2 in 20 mins and the ORP increased from 310 to 510 mV.	[40]
1. DC-driven streamer corona. 2. Transient spark discharge	10 mM	(~10 mA) with a 5–20 kHz repetition rate, 10 kV	The pH changed reduced by 4 units.	[44]
Air plasma generator with copper electrodes and quartz dielectric	2 cM	20 kHz, high voltage (not specified)	The pH changed from 6.8 to 2.3, ORP changed from 250 to 540 mV.	[45]
Atmospheric cold plasma jet	7·5 cM	20 kHz, 30 kV	The pH changed from 5.88 to 2.85, ORP changed from 406.1 to 565.40 mV.	[46]

Most studies have indicated an immediate drop in pH and an increase in electrical conductivity and the ORP as a result of the formation of reactive species in the PAW samples (Table 1). However, the increase of change in these properties cannot be directly correlated with a single factor or reason. When PAW is produced, the gaseous species from either the working or the atmospheric gas enters the liquid-gas interface and as a result there are complex reactions leading to the non-equilibrium, hence generation of the ionic moieties [44][47][48]. This process is highly influenced by the electric field and also using bubble implosions which hence the movement as well as dispersion of the phenomenon across the interface [49]. A recent review suggests that the electrical breakdown in water can occur without a phase change such as evaporating liquid and condensing or dissolving vapor [48]. The factors affecting the changes in PAW during activation may depend on multiple factors. For example, increase in discharge power, which is a direct function of applied voltage, would affect the increase in electric conductivity of the PAW [47]. On the other hand, in another study Vlad et al. showed that increase in treatment time would increase bacterial inactivation by PAW [50]. Although these studies reported above (Table 1) have used different set ups for the PAW generation, it could still be concluded that the efficiency can be a combined effect of two or more factors such as PAW activation time, temperature, power used and the aeration or bubbling to improve the formation of reactive oxygen species (ROS) [51].

Physicochemical Properties of PAW

PAW shows lower pH, higher conductivity and higher oxygen reduction potential when compared with untreated water [44][52]. The reduction in pH is due to the formation of acidic chemical species, which result in a steep decrease from pH 7 to pH 3 within 5–10 min of activation, but with little change thereafter [37][53]. Oxidation-reduction potential (ORP) can be defined as the ability of any solution to acquire or loose electrons to an electrode, and this property of PAW is much more prominent as compared with non-activated water. ORP of PAW depends on the strength of activation, which further depends on the applied voltage, carrier gas and other parameters leading to an increase of up to 63% [54]. Conductivity is the ability of any solution to allow current to pass through it and is reported to significantly increase due to plasma activation, primarily because of the generation of ions [45]. With a plasma jet that was operated from a 10 kHz sinusoidal high-voltage power source with 18 kV peak-to-peak AC voltage using pre-mixed oxygen and argon, the pH reduced from 7 to 3, ORP increased from 250 to 550 mV, conductivity rose from 0 to 410 μS/cm and temperature increased from 25 to 30 °C after 15 min of activation [55]. With a similar plasma source, when PAW was produced using 0.40–0.42 kV AC voltage, the conductivity increased from 5 to 20 mS/cm, the ORP value increased from 180 to 250 mV, pH decreased from 7.0 to 6.0, and the temperature increased from 20 to 40 °C [56]. Hence, the change in the reactive species of PAW are measurable as ORP and pH, and these changes directly show their effect on the potential to attack and disrupt the bacterial membranes during inactivation [40].

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