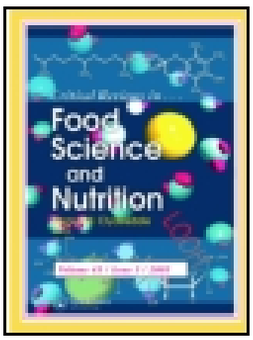

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REVIEW

A review on recent advances in plasma-activated water for food safety: current applications and future trends

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ABSTRACT

Plasma-activated water (PAW), the water or solutions treated with atmospheric cold plasma, is an eco-friendly technique with minimal changes in food products, making it a befitting alternative to traditional disinfection methods. Due to its potential microbicidal properties, PAW has been receiving increasing attention for applications in the food, agricultural, and biomedical fields. In this article, we aimed at presenting an overview of recent studies on the generation methods, physicochemical properties, and antimicrobial activity of PAW, as well as its application in the food industry. Specific areas were well discussed including microbial decontamination of food products, reduction of pesticide residues, meat curing, sprouts production, and disinfection of food contact materials. In addition, the factors influencing PAW efficiency were also well illustrated in detail, such as discharge parameters, types and amounts of microorganisms, characteristics of the liquid solution and food products, and treatment time. Moreover, the strategies to improve the efficacy of PAW were also presented in combination with other technologies. Furthermore, the salient drawbacks of this technology were discussed and the important areas for future research were also highlighted. Overall, the present review provides important insights for the application of PAW in the food industry.

KEYWORDS

Plasma-activated water; decontamination; food; reactive species; mechanism

Introduction

Microbial contamination of foods, an increasingly serious threat to global public health, can occur at all stages of food processing. Spoilage microorganisms can adversely affect the nutritional values, color, texture, and edibility of foods, causing excessive economic loss (Amit et al. 2017). Moreover, the consumption of foods or beverages contaminated with bacteria or other pathogens can result in foodborne disease outbreaks. As estimated by the U.S. Centers for Disease Control and Prevention (CDC), foodborne pathogen infections cause approximately 48 million illnesses, 128,000 hospitalizations, and 3000 deaths each year in the United States, mainly caused by *Campylobacter* spp., *Clostridium perfringens*, *Salmonella* spp., *Listeria monocytogenes*, and norovirus. The total economic cost of foodborne illnesses is $15.5 billion each year in the United States (Hoffmann et al. 2015). Therefore, suitable processing methods are necessary to ensure the microbiological safety of food.

With growing consumer demand for fresher, safer, and more nutritious foods, non-thermal food processing technologies have been extensively studied in recent years, such as high hydrostatic pressure, pulsed electric field, ultrasound, and cold plasma (Augusto 2020; Hernandez-Hernandez et al. 2019). Plasma, known as the fourth state of matter, is a partially ionized gas comprising electrons, ions, uncharged neutral particles (such as atoms, molecules, and radicals), and ultraviolet photons (Thirumdas et al. 2018). In recent years, cold plasma has attracted a lot of attention in the food and agricultural industries, mainly for applications in food sterilization and preservation (Hernandez-Hernandez et al. 2019). However, the highly irregular surface topography of food products offers numerous hidden places for microorganisms, thus increasing their resistance against the cold plasma treatment. To solve this problem, plasma-activated water (PAW) has been developed. Using PAW, the water exposed to plasma discharge, has been widely acknowledged as an alternative method for microbial disinfection of food products (Thirumdas et al. 2018). As an environment friendly and cost-effective disinfectant, PAW exhibits outstanding and broad antibacterial activity, offering new application possibilities in the food, agricultural, and biomedical fields (Kaushik et al. 2018; Thirumdas et al. 2018). In addition, other plasma-activated liquids (PALs), such as phosphate-buffered saline (PBS), saline, and medium, also exhibit excellent antimicrobial activities (Kaushik et al. 2018).

This review focused mainly on the application of PAW for the microbiological safety and quality of foods, on its preparation methods, and its physicochemical properties. Moreover, the factors influencing the efficiency of PAW and the combination of PAW with other technologies were well

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reviewed in detail. Finally, the limitations and future research suggestions were also identified and discussed in this article. Therefore, this review may potentially provide a solid basis for the development of PAW-based technologies in the food industry.

Generation of PAW

Generally, PAW is mainly produced based on atmospheric cold plasma (ACP) discharge in three categories: direct discharge within the liquids (Figure 1a–c); discharges in the gas phase over the liquid surface (Figure 1d–f); and multiphase discharges, such as discharges in bubbles inside liquids (Figure 1g and h) or contacting liquid sprays or foams (Figure 1i). Plasma jet, gliding arc discharge, dielectric barrier discharge (DBD), and surface micro-discharge (SMD) are the most common plasma sources used to produce PAW.

Various pilot units have been recently developed for PAW production. Andrasch et al. (2017) used the plasma source PLexc^2 to obtain PAW with a yield of 1 L/min (Schnabel et al. 2020). In 2017, a pilot unit for PAW production was developed by Pemen et al. (2017) with a capacity of 0.5 L per batch. Overall, the devices for PAW production do not meet the requirements of food processing, and the manufacturing cost of PAW is higher (one euro per 30 L of PAW) than chemical disinfectants, which hinders the practical applications of PAW in the food and agriculture industry (Schnabel et al. 2020).

Physicochemical properties of PAW

When plasma interacts with liquids, various complex chemical reactions occur at the interface region between the two media, resulting in the generation of reactive species and dramatic changes in the physicochemical properties of the treated solutions, including their pH, oxidation-reduction potential (ORP), and electrical conductivity.

Reactive species

During plasma discharge, various species are generated in the gas, such as nitric oxide radical (•NO), hydroxyl radical (•OH), superoxide anion radical ($\bullet O_2^-$), atomic oxygen (O), singlet oxygen ($^1O_2$), nitrogen ions (N_2^+), and excited nitrogen molecules (N_2^*). When these reactive species come into

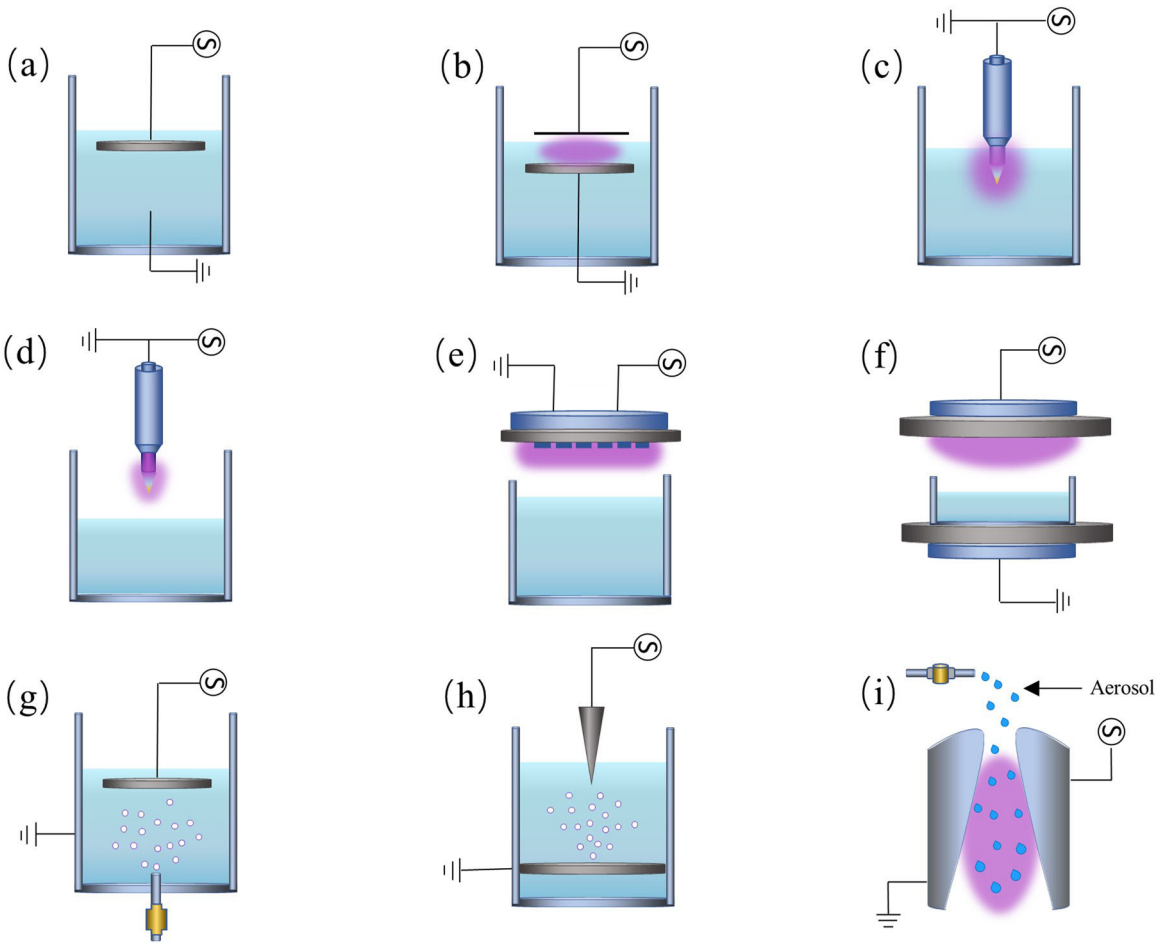

Figure 1. Schematic of different discharges used for the preparation of plasma-activated liquids. (a, b) Direct discharge within the liquids, adapted with permission from Locke et al. (2006), Copyright (2020) American Chemical Society. (c) Plasma jet direct contact with liquid (Kondeti et al. 2018). (d) Gas phase discharge over the liquids (Kondeti et al. 2018). (e, f) Dielectric-barrier discharge over the liquids, adapted with permission from Park et al. (2017), Copyright (2020) American Chemical Society. (g) Corona discharge in bubbles (Vanraes and Bogaerts 2018). (h) Plasma streamer bubble discharge in water. (i) Gliding arc discharge with aerosol (Perinban, Orsat, and Raghavan 2019).

contact the liquids, numerous long-lived reaction products are formed, e.g., hydrogen peroxide (H_2O_2), nitrate (NO_2^-), nitrite (NO_3^-), ozone (O_3), peroxynitrite anion ($ONOO^-$), and peroxynitrous acid (ONOOH). The type and concentration of the reactive species present in the PALs are significantly affected by the plasma device, the working gas, and the liquids used.

Metal ions

Electrode erosion during in-liquid discharges has also been extensively studied, which might result in the release of metal ions into PAW (Corella Puertas, Dzafic, and Coulombe 2020). According to Chen et al. (2018), the levels of copper and zinc ions in PAW increased as a function of plasma exposure time, contributing to PAW-induced microbial inactivation.

pH

In most cases, the different plasma treatments lead to an acidification of the treated water or solutions, as indicated by the pH, which is inversely proportional to the plasma activation time (Ercan et al. 2013; Shen et al. 2016; Tian et al. 2015). After 3 min of DBD plasma treatment, the pH of deionized water (pH = 6.80), PBS (pH = 6.94), and 5 mmol/L N-acetylcysteine (NAC) solution (pH = 6.24) decreased to 2.00, 2.58, and 2.35, respectively (Ercan et al. 2013). Such a decrease of the pH value is hypothesized to be mainly caused by the formation of nitric and nitrous acids as well as ONOOH, generated from the NO, NO_2, and NOx formed in the plasma phase (Oehmigen et al. 2010). In addition, the generation of acidic hydronium ions (H_3O^+) by the reaction of the water molecules with H_2O_2 generated in air or liquid might also contribute to the decrease in the pH value (Oehmigen et al. 2010). Therefore, acidic pH is assumed to play a critical role in the microbial inactivation reduced by PAW (Naïtali et al. 2010).

ORP

ORP reflects the oxidizing or reducing ability of a solution and is considered as a primary factor affecting microbial inactivation by destroying cell membrane integrity and cellular defense mechanisms (Liao, Chen, and Xiao 2007). The ORP values of PAW displayed a significant increase, mainly depending on the plasma activation time (Ma et al. 2015). As observed by Tian et al. (2015), PAW generated beneath the water surface exhibited a higher ORP value compared to that produced above the water surface. Furthermore, H_2O_2 supplementation could increase the ORP of plasma-activated distilled water (Wu et al. 2017). The high ORP of PAW is mainly due to the formation of various reactive chemical species, such as H_2O_2, O_3, NO_3^-, NO_2^-, and ONOOH (Kaushik et al. 2018).

Electrical conductivity

Electrical conductivity, a measure of the ability of an aqueous solution to conduct electricity, depends on the types of ions, their concentrations, and the solution temperature. The electrical conductivity of PAW increased dramatically with the activation time (Ma et al. 2015; Wu et al. 2017; Zhang et al. 2016), suggesting the generation of certain active ions during the plasma-liquid interactions. Wu et al. (2017) revealed that the conductivity of plasma-activated H_2O_2 solutions simultaneously increased with the H_2O_2 concentration. Vlad and Anghel (2017) demonstrated that the electrical conductivity of PAW prepared with air-discharge plasma was higher than those with He or Ar. With increasing solution conductivity of 100–500 μS/cm, both the production rate and the saturation level of H_2O_2 decreased by the corona discharge in water (Lukes et al. 2008).

Temperature

Plasma activation does not result in significant increases in solution temperatures (Ercan et al. 2013; Qian et al. 2019; Tian et al. 2015). According to Ercan et al. (2013), DBD plasma-treated fluid (deionized water, PBS, and 5 mM NAC solution) temperatures were maintained within the ranges of 23–26 °C, even when the plasma treatment lasted 3 min. The temperatures of the PAW samples activated by a plasma microjet above or beneath the sterile distilled water surface for 20 min reached 31.5 °C and 38.1 °C, respectively (Tian et al. 2015). Thus, PAW is suitable for the disinfection of heat-sensitive food materials. Moreover, the thermal effect of the plasma discharge during PAW preparation could be prevented by using a circulating water jacket (Kamgang-Youbi et al. 2007).

In vitro antimicrobial activity and mechanisms of PAW

Bacteria

Table 1 shows an overview of investigations related to the in vitro antimicrobial activity of PAW and other PALs against planktonic bacteria. As presented in Table 1, PAW and PALs could effectively inactivate various food spoilage bacteria (such as *Aeromonas hydrophila*, *Shewanella putrefaciens*, and *Pseudomonas fluorescens*) and pathogenic bacteria (e.g., *Staphylococcus aureus*, *Escherichia* coli, and *Listeria innocua*).

Biofilms are densely packed communities of microorganisms that are adherently attached to each other and/or a surface, representing a significant threat to food safety (Xu et al. 2020). PAW also exhibits antibiofilm activity and can disrupt pre-formed biofilms (Chen, Su, and Liang 2016; Ercan et al. 2014; Handorf et al. 2020; Smet et al. 2019). As reported by Park et al. (2017), a 10-min PAW treatment caused a 1.9 log CFU/cm^2 inactivation of *E. coli* DH5α cells in biofilms formed on stainless steel pieces. The long-lived reactive species (such as H_2O_2, NO_3^-, NO_2^-, and O_3) are determined to play key roles in PAW-mediated inactivation

Table 1. *In vitro* inactivation efficiency of PAW and PALs against planktonic bacteria.

Strain	Preparation parameters	Treatment condition	Log reduction	Reference
H. alvei	Gliding arc discharge, compressed air at 550 L/h, distance of 13 cm, 200 mL sterile NaCl solution (0.15 mol/L), 2 to 5 min	H. alvei suspension (12 mL) was incubated with 188 mL of PAW for 5, 10, 15, or 20 min	>5 log after 15 min treatment	Kamgang-Youbi et al. (2007)
S. epidermidis, L. mesenteroides, and H. alvei	Gliding arc discharge, compressed air at 550 L/h, distance of 13 cm, 20 mL of SDW, 5 min	Microbial suspensions (0.1 mL) were incubated with 9.9 mL of PAW for 5 to 30 min	S. epidermidis: about 7 log after 15 min; L. mesenteroides and H. alvei: >5 log after 20 min	Kamgang-Youbi et al. (2009)
H. alvei	Gliding arc discharge, 20 mL of SDW, 5 min	H. alvei suspension (0.1 mL) was incubated with 9.9 mL of PAW for 5, 10, 20, or 30 min	1.1, 2.2, 5.4, and >5.9 log after 5, 10, 20, and 30 min exposure	Naïtali et al. (2010)
E. coli K12	Surface micro-discharge plasma, 5 kV, 10 kHz, 0.288 W/cm2, distance of 42 mm, 10 mL of DW, 20 min	E. coli pellet was resuspended in 1 mL of PAW for 15 min	5.6 log	Traylor et al. (2011)
E. coli	DBD plasma, 1 mL of DW, 1, 2, and 3 min	E. coli suspension (50 μL) was incubated with 50 μL of PAW for 15 min	$PAW_{1\,min}$: 0.39 log; $PAW_{2\,min}$: 1.05 log; PAW_{3min}: >7.08 log	Kojtari et al. (2013)
S. aureus, S. mutans, and *E. coli*	Plasma jet, 0.42 kV, 30 mA, 98% Ar + 2% O_2 at 5 L/min, 20 mL of DW, 20 min	Bacterial suspension (0.1 mL) was incubated with 20 mL of PAW for 5, 10, 15, or 20 min	S. mutans: about 5 log after 5 min exposure; S. aureus: 3.6 log after 15 min exposure; E. coli: about 3.5 log after 15 min exposure	Ye et al. (2013)
S. aureus	Plasma jet, 98% Ar + 2% O_2 at 5 L/min, 10-mm distance, 20 mL SDW, 20 min	S. aureus suspension (0.1 mL) was incubated with 20 mL of PAW for 10, 20, 30, or 40 min	6 log after 10 min exposure	Zhang et al. (2013)
E. coli K12	Surface dielectric barrier discharge, 17 kV, 6 kHz, 5 mL of NaCl solution (0.85%), 1, 3, 5, and 7 min	E. coli suspension (0.1 mL) were incubated with 4.9 mL of PAL for 15 min	6 log after 5 min exposure to PAW 3 min	Hänsch et al. (2015)
S. aureus and 38 strains of coagulase-positive Staphylococci	Glid Arc reactor	Bacterial suspension (10 mL) was incubated with 90 mL of PAW for 3, 5, 7, or 10 min	0 to 7.38 with an average of 5 log after 3 min treatment; 4.99 to 10.11 with an average of 7.61 log after 10 min exposure	Lipovan et al. (2015)
S. aureus	Plasma microjet, 0.40–0.42 kV, 30 mA, 98% Ar + 2% O_2 at 5 SLM, 10-mm distance, 20 mL of SDW, above (PAW-B) or beneath (PAW-B) the water surface, 5, 10, 15, and 20 min	S. aureus suspension (0.1 mL) was incubated with 20 mL of PAW for 20 min	PAW-A: 0.4 to 1.2 log; PAW-B: 3.4 to 5.0 log	Tian et al. (2015)
S. aureus	Plasma microjet, 20 kHz, air at 260 L/h, 2 cm distance,15 mL of SDW, 5 or 10 min.	S. aureus suspension (0.1 mL) was incubated with 9.9 mL of PAW for 10 min	$PAW_{5\,min}$: about 7 log; $PAW_{10\,min}$: about 7 log	Zhang et al. (2016)
S. aureus	Plasma microjet, 100 kV, 0.35 A, 20 kHz, air at 260 L/h, 15 mL of DW, 5 min	S. aureus suspension (0.1 mL) was incubated with 9.9 mL of PAW for 3, 5, or 10 min	2 log after 3 min exposure, about 2.5 log after 5 min treatment	Wu et al. (2017)
S. aureus	Plasma microjet, 100 kV, 0.35 A, 20 kHz, air at 260 L/h, 15 mL of H_2O_2 solutions (1, 10, and 100 mM), 5 min	S. aureus suspension (0.1 mL) was incubated with 9.9 mL of PAL for 3, 5, or 10 min.	3 log for 3-min exposure to PAL with 10 mM H_2O_2, 4.6 log for 3-min exposure to PAL with 100 mM H_2O_2,	Wu et al. (2017)
E. aerogenes	Plasma jet, 295 V, 22.5 kHz, air pressure of 1990 mBar, 8.1 cm distance, 200 mL SDW, 5 min	Bacterial suspension (0.1 mL) was reacted with 1 mL of PAW for 10 min.	1.92 log	Joshi et al. (2018)
S. putrefaciens	Microplasma jet, 6–12kV, 7 kHz, air at 1.0 SLM, 40 mL of DW, 0–30 min	Bacterial suspension (20 μL) was incubated with 300 μL of PAW for 5 min	0.13 to 2.0 log	Qi et al. (2018)
E. coli	Pinhole discharge reactor, 8 kV, 20 kHz, NaCl solution (500 μS/cm) at 72 mL/min, air at 12.5 mL/min	E. coli suspension (20 μL) was incubated with 1.98 mL of PAW for 0.5, 1, 1.5, 2, 3, or 4 min	About 1.5 log after 4-min exposure to PAW	Suganuma & Yasuoka (2018)
E. coli	Pinhole discharge reactor, 8 kV, 20 kHz, sodium citrate solution (500 μS/cm) at 72 mL/min, air at 12.5 mL/min	E. coli suspension (20 μL) was incubated with 1.98 mL of PAL for 0.5, 1, 1.5, 2, 3, or 4 min	About 4 log for 1-min exposure to PAL	Suganuma & Yasuoka (2018)

(*continued*)

Table 1. Continued.

Strain	Preparation parameters	Treatment condition	Log reduction	Reference
P. deceptionensis CM2	Plasma jet, 750 W, compressed air (0.18 MPa), 200 Ml of SDW, 30, 60, or 90 s	Bacterial suspension (0.1 mL) was incubated with 0.9 mL of PAW for 6 min	1.54 to 5.30 log	Xiang et al. (2018)
E. coli	Plasma jet, about 0.3 kWh/L PAW, 50 mL of SDW, 5, 10, 20, or 30 min	E. coli suspension (0.1 mL) was reacted with 9.9 mL of PAW for 10 min	1.03 to 4.40 log	Zhou et al. (2018)
E. coli (O1:K1:H7)	NEAPR source, DW, Ar + O_2+N_2 at 1.4 SLM, 21-mm distance, 100 mL of DW, 10 min	E. coli suspension (0.3 mL) was incubated with 2.7 mL of PAW at 250 rpm and 30 °C for 24 h	About 7 log after 24 h treatment	Iwata et al. (2019)
E. coli K12 and *S. aureus*	Plasma jet, 6.8 kV, 1.5 kHz, 0.5 cm distance, 20 mL of deionized water, Ar at 3 SLM, 6.5min	Bacterial suspension was incubated with PAW for 1 h	E. coli K12: 7.14 log; S. aureus: 3.10 log	Royintarat et al. (2019)
E. coli K12 and *S. aureus*	Cylindrical DBD plasma, 15 kV, 20 kHz, 0.5 cm distance, 20 mL SDW, 99% Ar +1%O2 at 4 SLM, 11.5min	Bacterial suspension was incubated with PAW for 1 h	E. coli K12: 0.45 log; S. aureus: 2.45 log	Royintarat et al. (2019)
E. coli O157:H7	Plasma jet, 5 kV, 40 kHz, 750 W, compressed air (0.18 MPa, 30 L/min), 200 mL of SDW, 60 s	E. coli suspension (0.1 mL) was incubated with 0.9 mL of PAW at 25, 40, 50, or 60 °C for 4 min	0.77 log to undetectable level	Xiang, Kang, et al. (2019)
E. coli DH5α	Plasma array, 18 kV, 60 mA, 5 kHz, air, 1.5 cm distance, 5 mL of DW, 5 min	Bacteria pellets were incubated with 1 mL of PAW for 15 min	About 6 log	Ma et al. (2020)
E. coli, *L. innocua*, *S. aureus*, *A. hydrophila*, *P. fluorescens*, and *S. putrefaciens*	Plasma jet, 15, 22, or 30 kV, 20 kHz, air at 11 L/min, 7.5 cm distance, 30 mL of SDW, 5 min	Bacterial suspension (1 mL) was incubated with 9 mL of PAW at 4 °C for 0.5, 1, 3, 5 or 24 h	2.99 log for E. coli, 2.50 log for L. innocua, 0.49 log for S. aureus, 5.61 log for A. hydrophila, >5.58 for P. fluorescens, and undetectable level for S. putrefaciens after 0.5-h exposure to PAW prepared at 30 kV	Zhao, Ojha, et al. (2020a)

DW, distilled water; SDW, sterile distilled water; SLM, standard liters per minute.

of bacterial biofilms (Xu et al. 2020). However, further investigation would still be necessary to clarify the inactivation mechanism of PAW-induced biofilms, such as the influence of PAW on the production of quorum-sensing signal molecules.

Bacterial spores are a threat to food safety due to their increased resistance to extreme environmental stresses compared to that of vegetative cells. Bai et al. (2020) discovered that *Bacillus. cereus* spores in 10^6 CFU/mL were reduced by 1.62 to 2.96 $\log_{10}$ CFU/mL after PAW treatment at 55 °C for 5–60 min. The synergistic interaction of PAW and mild heat against bacterial spores has also been observed in rice. *B. cereus* spores in rice were inactivated by 1.54 and 2.12 log reductions after 60 min of exposure to PAW at 40 and 55 °C, respectively (Liao, Bai, et al. 2020).

Fungus

PAW also exhibits antifungal activity against yeasts (Kamgang-Youbi et al. 2009; Tian et al. 2017; Ye et al. 2013), molds (Ma et al. 2016), and fungal spores (Hojnik et al. 2019). Kamgang-Youbi et al. (2009) observed approximately 3 log reduction in *Saccharomyces cerevisiae* culture after 30 min of exposure to PAW. The antifungal effect of PAW could also be found in real foods, especially in plant foods such as fruits and vegetables (Guo et al. 2017; Ma et al. 2016; Xiang, Liu, et al. 2019). Besides, PAW could also inactivate fungal spores to a certain extent. Following the treatment of air-PAW and oxygen-PAW for 30 min, *C. gloeosporioides* spores populations were diminished by 96% and 55%, respectively (Wu et al. 2019). Following PAW treatments of 2 or 24 h at 15 °C, Los et al. (2020) also found out that the population of *Aspergillus flavus* spores only decreased by 0.2 and 0.6 log CFU/mL, respectively, while their metabolic activities maximally decreased by 42.2% and 55.2%, respectively. Compared with bacteria, yeast and mold cells show higher PAW resistance, which might be due to their distinctive structure (Kamgang-Youbi et al. 2009). Therefore, the antifungal effect of PAW might be significantly improved by combining PAW with other methods such as ultrasound and mild heat (Royintarat et al. 2020; Zhang et al. 2020).

Viruses

Viruses, such as noroviruses, hepatitis A and E viruses, rotaviruses, astroviruses, and adenoviruses, are the major cause of food poisoning (Hirneisen et al. 2010). Guo et al. (2018) reported that PAW efficiently inactivated bacteriophages T4, Φ174, and MS2 in a time-dependent manner. PAW and PALs (generated with 0.9% NaCl solution and 0.3% H_2O_2 solution) could completely inactivate Newcastle disease virus

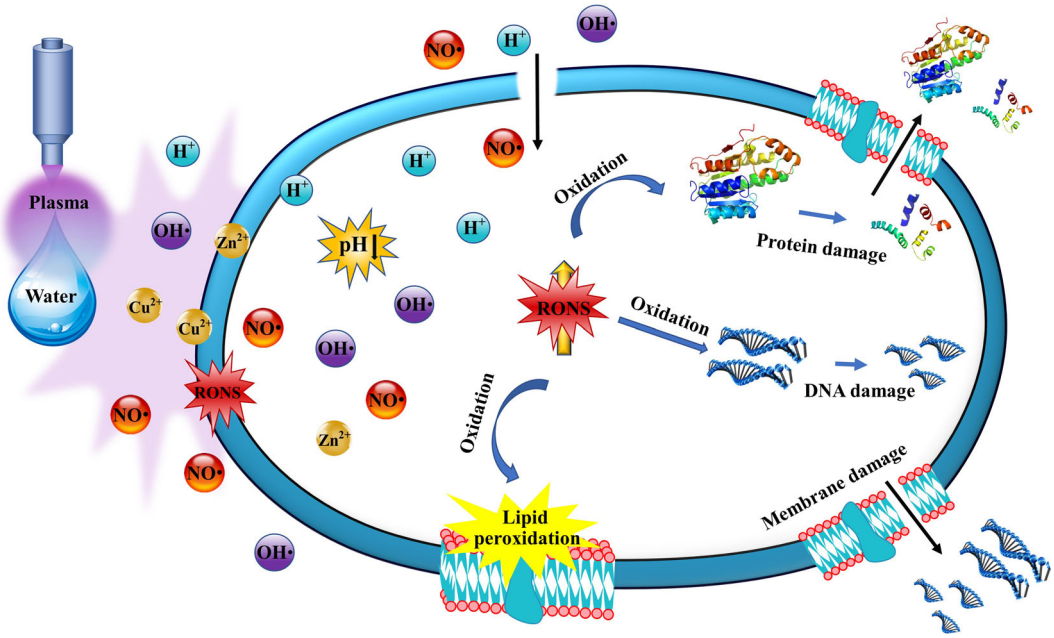

Figure 2. Schematic diagram of the PAW-induced inactivation of microbial cells.

following a 30 min exposure (Su et al. 2018). Therefore, PAW, as an environmental friendly method, is a promising alternative strategy to combat the foodborne and environmental viruses.

Antimicrobial mechanisms of PAW

The mechanisms associated with the PAW-induced inactivation of microorganisms have been addressed in several studies (Figure 2). As described by Naïtali et al. (2010), the acidified chemical mixture of H_2O_2, NO_3^-, and NO_2^-, resulted in a 2.5 log reduction of *Hafnia alvei* after 20 min, which explained 75% of the logarithmic reduction achieved by PAW and 99.99% of the number of dead bacteria. These data suggested that the combined effects of long-lived reactive chemical species and acidic conditions are mainly responsible for the microbial inactivation induced by PAW. After exposure to PAW, remarkable structural changes were be observed in the microbial cells, such as cell shrinkage, holes on the surface of bacterial cells (Xiang et al. 2018; Shen et al. 2016), and deformation of external viral shape (Su et al. 2018). PAW could also disrupt the microbial cell membrane integrity and membrane potential, resulting in the leakage of intracellular components such as proteins and nucleic acids (Xiang et al. 2018; Zhang et al. 2016). A significant decrease was observed in the intracellular pH and an increase in the intracellular RONS levels in microbial cells upon PAW exposure, which might take part in the inactivation of microorganisms by affecting their cellular metabolism and physiological functions (Zhang et al. 2016; Zhang et al. 2020). It was demonstrated that PAW caused a remarkable alteration in the DNA structure and chemical bonds of *S. aureus* cells (Zhang et al. 2016). Furthermore, the release of metal ions from the electrodes plasma reactor might also play an important role in the PAW-induced microbial inactivation (Chen et al. 2018).

Overall, the recent investigations of the antimicrobial mechanisms of PAW mainly focus on the cellular structure, metabolism, and physiological function of the tested strains. However, the influences of PAW on microbial gene expression and protein synthesis have not yet been fully understood. Therefore, a better understanding of the PAW treatment-induced signaling events on microbial cells is required for applying PAW in the improvement of microbiological food safety.

Application of PAW in the food industries

The application of PAW in the food industries has been well-investigated over the past few years, such as preservation of fruits and vegetables, decontamination of meat and shell eggs, pesticide reduction, and the curing of meat products (Figure 3).

Decontamination of agricultural products

The application of PAW for the decontamination of agricultural products is outlined in Table 2. As demonstrated by previous studies, PAW could effectively inactivate aerobic bacteria, yeasts, and molds on fruits (such as strawberries, Chinese bayberry, and grapes), vegetables (e.g., mung bean sprouts, spinach, and lettuces), and edible mushrooms such as button mushrooms (Table 2), thereby extending the shelf life of foodstuff and reducing the generation of food waste. PAW is also used for the preservation of fresh-cut fruits, such as pears (Chen et al. 2019), kiwifruits (Zhao et al. 2019), and apples (Liu et al. 2020).

The application of PAW has been recently tested at the Leibniz Institute for Plasma Science and Technology for the decontamination of fresh-cut lettuces, one of the most popular substrates for testing PAW efficacy (Fröhling et al. 2018; Schnabel et al. 2015; Schnabel et al. 2019; Schnabel et al.

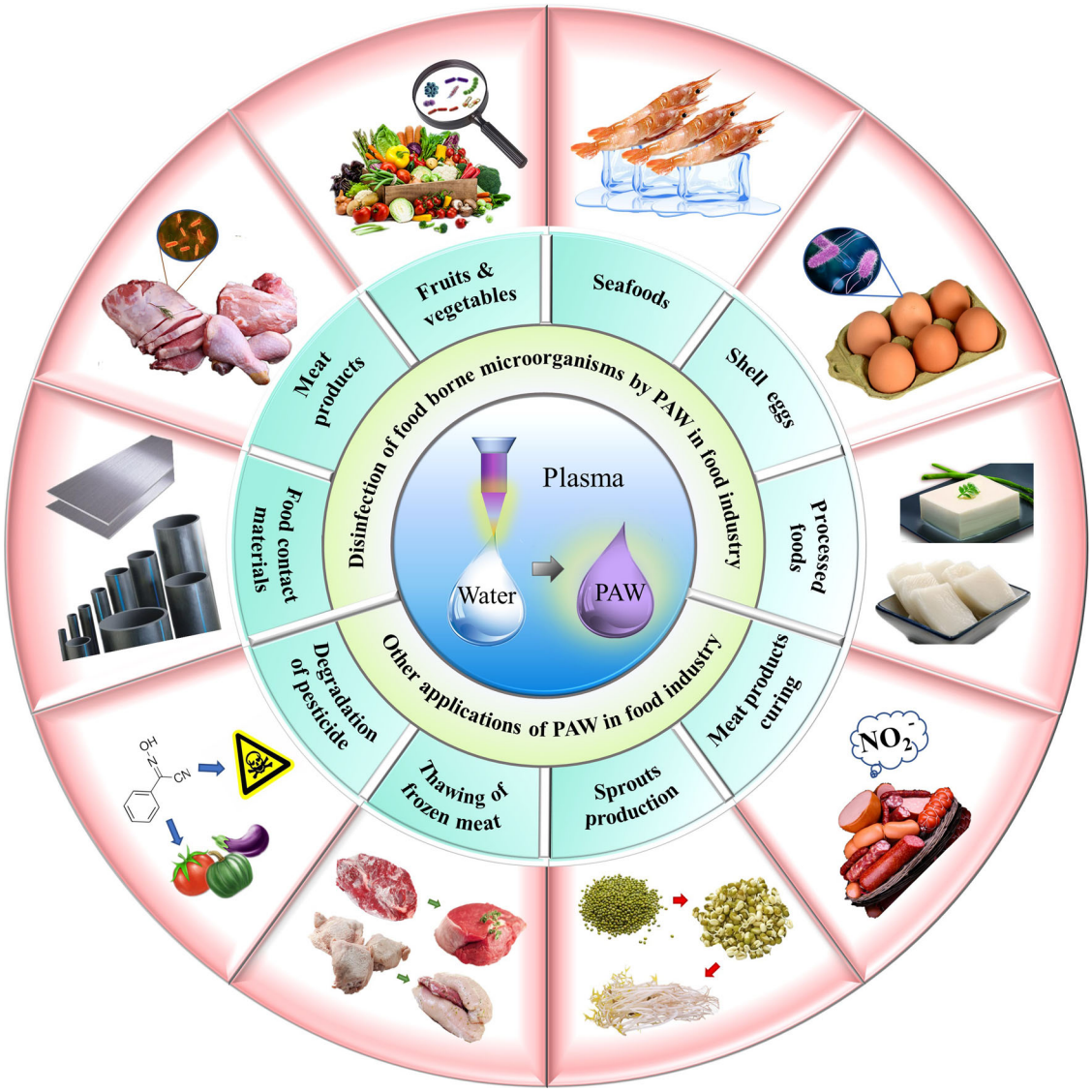

Figure 3. Schematic representation of PAW application in the food industries.

2020). The study showed that the maximal 5 log reduction of total aerobic counts was observed on fresh-cut lettuces after PAW washing at the lab-scale, while an approximate reduction of 2 log reduction was found at the pilot-scale (Schnabel et al. 2020). Moreover, the PAW-washing treatment resulted in no adverse effect on the color, texture, surface structure, and lettuce tissue cells or ganelles. As reported by Xiang, Liu, et al. (2019), the use of a PAW-soaking treatment up to 30 min caused no significant changes in the sensory characteristics of mung bean sprouts, including their appearance, color, flavor, texture, and overall acceptance.

Pesticide reduction in fresh produce

The continuous and non-judicious application of agrochemicals leads to varying detrimental effects on humans, the environment, and biodiversity (EFSA 2019). As reported by Zheng et al. (2019), a maximum phoxim residue reduction of 73.60% was observed on grapes following a PAW dip-treatment for 10min, which was significantly higher than those of following treatments with deionized water and commercial cleaning agent almawin. Furthermore, two degradation intermediates are identified to be 2-Hydroxyimino-2-phenylacetonitrile, O-diethyl O-(alpha-cyano benzylideneamino) phosphonate (Zheng et al. 2019). Similarly, Ranjitha Gracy, Vidhi, and Mahendran (2019) observed a maximum chlorpyrifos reduction of 51.97% in tomatoes following a PAW treatment, which was higher than that after distilled water immersion (0.12%–0.64%). The degradation of pesticides is primarily due to the reactive oxygen and nitrogen species present in PAW (Zheng et al. 2019). Thus, PAW treatment might be a feasible and effective approach to reduce pesticide residues in fresh fruits and vegetables.

Decontamination and curing of meat products

The efficiency of PAW to inactivate microorganisms in different meats and meat products has also been evaluated

Table 2. Effects of PAW and PALs on microbial inactivation in fruits and vegetables.

				Results		
Fresh produce	Microorganism	Preparation parameters	Treatment condition	Log reduction	Quality attributes	Reference
Strawberries	*S. aureus*	Plasma jet, 18 kV, 10 kHz, 2 cm distance, 98% Ar + 2% O_2 (5 L/min), 80 mL of SDW, 10 or 20 min.	Samples were immersed in PAW for 5, 10, and 15 min at 300 rpm, then stored at 20 °C for 4 d.	1.6 to 2.3 log CFU/g at day 0; 1.7 to 3.4 log CFU/g at day 4	No significant change in color, firmness, and pH value	Ma et al. (2015)
Fresh-cut iceberg lettuce (2 × 2 cm)	*E. coli* K12, *P. fluorescens*, *P. marginalis*, *P. carotovorum*, and *L. innocua*	Microwave discharge plasma, 2.45 GHz, 1.1 kW, air at 18 SLM, DW, 5, 15 or 50 s.	Specimens were immersed in PAW for 1, 3, or 5 min.	1.8 to 6.1 log	After 8 d of storage, little influences on the texture and appearance of the tested samples	Schnabel et al. (2015)
Mung bean sprouts	*E. coli* K12, *P. fluorescens*, *P. marginalis*, *P. carotovorum*, and *L. innocua*	Microwave discharge plasma, 2.45 GHz, 1.1 kW, air at 18 SLM, DW, 5, 15 or 50 s.	Specimens were immersed in PAW for 1, 3, and 5 min.	2.5 to 3.5 log	After 8 d of storage, little influences on the texture and appearance of the tested samples	Schnabel et al. (2015)
Chinese bayberry	Total aerobic bacteria and fungi	Plasma jet, 20 kHz, 3 cm distance, air (260 L/h), 1600 mL of SDW, 25 min.	Samples were soaked in PAW for 0.5, 2, or 5 min and then stored at 3 °C for 8 d.	Day 0: 0.8 log for bacteria, 0.4 log for fungi; day 8: about 1.1 log for bacteria and fungi	Decay incidence decreased, firmness and total soluble solids were well maintained, the CIRG was increased	Ma et al. (2016)
Button mushrooms	Total aerobic bacteria and fungi	Plasma jet, 18 kV, 10 kHz, 1 cm distance, 98% Ar + 2% O_2 (5 L/min), 500 mL SDW, 15 min.	Samples were immersed in PAW for 5, 10, and 15 min, then stored at 20 °C for 7 d.	1.5 log for bacteria and 0.5 log for fungi at day 7	No significant change in the color, pH, or antioxidant properties; the softening was delayed	Xu et al. (2016)
Grapes	*S. cerevisiae*	Plasma jet, 8.2 kV, 1.1 to 1.3 mA, 1.5-mm distance, air at 1.2 L/min, 20 mL SDW, 30 or 60 min.	Grapes were soaked in PAW for 30 min.	$PAW_{30\,min}$: 0.38 log; $PAW_{60\,min}$: 0.53 log	No significant change in surface color and total anthocyanin content	Guo et al. (2017)
Fresh-cut endive lettuce (5 × 5 cm)	Total viable count	PLexc[2] plasma source	Lettuces were washed with or without PAW at different processing steps, then stored at 2 °C for 7 d.	Day 0: 0.64 to 1.6 log CFU/g; day 7: 0.27 to 0.95 log CFU/g	n.d.	Fröhling et al. (2018)
Grape tomatos, limes, and spiny gourds	*E. aerogenes* B 199 A	295 V, 22.5 kHz, 8.1 cm distance, air (1.990 Bar), 200 mL of SDW, 5 min	Samples were washed with PAW for 3 min at 50 rpm	1.98 log for grape tomatos, 1.77 log for limes, 1.03 log for spiny gourds	n.d.	Joshi et al. (2018)
Grape tomatos, limes, and spiny gourds	*E. aerogenes* B 199 A	295 V, 22.5 kHz, 8.1 cm distance, air (1.990 Bar), 200 mL of citrate-phosphate buffer, 5 min	Samples were washed with plasma-activated buffer for 3 min at 50 rpm	2.00 log for grape tomatos, 1.97 log for limes, and 1.62 log for spiny gourds	n.d.	Joshi et al. (2018)
Fresh-cut pears (2 cm-thick)	Total aerobic bacteria, yeasts, and molds	Microplasma array device, 6, 8, or 10 kV; 9.0 kHz, air at 1.0 SLM, DW	Samples were immersed in PAW for 5 min, then were stored at 4 °C for 12 d.	12th day: 0.11 to 0.65 log for total aerobic bacteria, 0.84 to 1.04 log for yeasts and 0.31 to 0.77 log for molds	The quality and antioxidant properties were well maintained	Chen et al. (2019)
Fresh-cut iceberg lettuce and red leaf lettuce (3 × 3cm)	*S. typhimurium*	Glow plasma, O_2 or air	Lettuce pieces were washed with PAW for 1 or 3 min.	3.0 log for iceberg lettuce; 2.6 log for red leaf lettuce	No significant change in color parameters	Khan & Kim (2019)
Fresh-cut iceberg lettuce (5 × 5 cm)	*P. fluorescens* and *L. innocua*			*P. fluorescens*: reduced below detection limit	n.d.	Patange et al. (2019)

		Submerged DBD plasma, 20 kV, 7.45W, 25.8kHz, air, 700 mL of SDW	Samples were washed in the reactor for 0 to 10 min	within 3 min treatment; *L. innocua*: about 2.4 log after 5 min treatment		
Fresh-cut endive lettuce (2 × 2 cm)	Total aerobic bacteria	PLexc^2 plasma source, 1.3 kW and 12 SLM for the first stage, 1.7 kW and 80 SLM for the second stage, DW or tap water	Fresh-cut lettuces were washed with PAW with different scenario.	Pilot-scale: <2 log; lab-scale: >6.0 log	No adverse influences on the color and texture.	Schnabel et al. (2019)
Fresh-cut spinach leaves	Total aerobic bacteria	Surface barrier discharge, 36 W, 11 kV, 12 kHz, 44.8-mm distance, air, DW, 20 min	Samples (5 g) were rinsed with 100 mL of PAW for 2 min at 120 rpm.	about 1 log after storage at 4 °C for8 d	No adverse impacts on a^* and b^* values; higher L^* value.	Vaka et al. (2019)
Mung bean sprouts	Total aerobic bacteria, yeasts, and molds	Plasma jet, 5 kV, 40 kHz, compressed air (0.18 MPa, 30 L/min), 0.5-mm distance, 200 mL of SDW, 30 s	Sprouts (10 g) were immersed in 200 mL of PAW at 130 rpm for 30 min.	2.32 log for total aerobic bacteria and 2.84 log for yeasts and molds	No significant changes in the antioxidant capacities, total phenolic and flavonoid contents, and sensory characteristics.	Xiang, Kang, et al. (2019)
Fresh-cut kiwifruit (7 × 4 × 0.4 cm)	*S. aureus*	Microplasma jet, 10 kV, 8 kHz, air at 1.0 SLM, 40 mL of DW, 30 min	Samples were sprayed with 1 mL of PAW and then stored at 4 °C for 8 d.	1.8 log CFU/g on the 8th day	The firmness and b^* value of kiwifruit slices were well maintained	Zhao et al. (2019)
Fresh-cut apple (1 × 1 × 1 cm)	Total aerobic bacteria, molds, yeasts, and coliforms	Microplasma array device, 8 kV, 7.0 kHz, air at 1.0 SLM, DW, 10 min	Samples were immersed in PAW for 5 min and then were stored at 4 °C for 12 d	1.05 log for aerobic bacteria, 0.64 log for molds, 1.04 log for yeasts, and 0.86 log for coliforms	No influences on the firmness, titratable acidity, antioxidant contents and activities. The browning was inhibited	Liu et al. (2020)
Fresh-cut lettuce	Total plate count	PLexc^2 plasma source, DW for lab-scale experiment, tap water for up-scaled experiment	Fresh-cut lettuces were Washed with PAW with different scenario	Up to 5 log	No adverse impacts on the color, texture, surface structure, and the lettuce tissue cell organelles.	Schnabel et al. (2020)
Sliced potatoes and white radishes	GFP-*P. carotovorum* 10057	Micro-plasma jet, 1 cm distance, N_2, sterile PBS, 5 min	Samples were smeared with 100 μL of PAL, and 100 μL of GFP-*P. carotovorum* culture was spread on top	NPB exerted strong antibacterial effect on GFP-*P. carotovorum*	No influences on color, aroma, hardness, thickness, and tissue of sliced potatoes and white radishes.	Seo et al. (2020)
Grapes	*S. cerevisiae*	Plasma jet, 5 kV, 750 W, 40 kHz, compressed air (0.18MPa, 30L/min), 300 mL SDW, 90 s	Grapes were immersed in PAW at 25 °C for 30 min	0.39 log	No adverse impacts on the firmness, color, total soluble solids, total phenolics, vitamin C, and antioxidant properties.	Xiang et al. (2020)

DW, distilled water; n.d.: not determined; SDW, sterile distilled water; SLM, standard liters per minute.

Table 3. Effects of PAW and PALs on the microbiological safety and quality of meat products.

Meat type	Microorganism	Preparation parameters	Treatment condition	Results		Reference
				Log reduction	Quality attributes	
Amulsion-type sausage	Total aerobic bacteria	surface dielectric barrier discharge, 3.14 W, 15 kHz, air, 100 mL of DW containing 1% sodium pyrophosphate, 120 min.	Ground meat was mixed with PAW (9%). The sausages were storaged at 4 °C for 28 d.	PAS showed similar antimicrobial ability with $NaNO_2$	No noticeable effects on the total aerobic bacterial counts, color, and peroxide values of sausagesduring storage; less residual nitrite.	Jung et al. (2015a)
Loin ham	Total aerobic bacteria	SDBD, 15 kHz, 500 mL of DW containing 1% sodium pyrophosphate, 2 h	PAL was added into brine solutions at 20% (w/w).	0.33 log CFU/g	Higher a^* and lower residual nitrite than those of $NaNO_2$-cured samples.	Yong et al. (2018)
Chicken breast (2 × 2 × 1 cm, 8 g)	*P. deceptionensis* CM2	Plasma jet, 5 kV, 40 kHz, 750W, compressed air (0.18 MPa), 5-mm distance, 200 mL of SDW, 30, 60, or 90 s.	Samples were soaked in PAW for 3, 6, 9, and 12 min.	1.05 log CFU/g	Low pH, decreased a^* and b^*, mainted hardness and springiness, decreased cohesiveness and gumminess.	Kang et al. (2019)
Fresh beef (5 × 5 cm, 10 g)	*S.* Enteritidis	Plasma jet, 9.2kV, 0.024 mA, 0.46W, 10-mm distance, air at 22.5L/min, 300 mL of lactic acid solution (0.05%–0.20%), 80 s.	Beef samples were immersed in PAL for 20s	1.24 to 3.52 log CFU/g	No significant changes in color, pH, lipid oxidation, odor and protein	Qian et al. (2019)
Beef jerky (10 × 4 × 0.5 cm, 10 g)	*L. innocua* M300	Plasma jet, 9.2kV, 0.024 mA, 300W, 20 kHz, air or N_2 (1 L/min), 100 mL of brine solution, 10 min	Beef slices were cured in PAL for 18–20 h at 4 °C.	0.5 log CFU/mL in brines and 0.85 log CFU/g in jerky	No significant changes in L^*, b^*, TBARS, shear force, and cutting strength, while a^* value increased significantly.	Inguglia et al. (2020)
Chicken muscle, rough skin, and smooth skin (diameter:1 mm; thickness: 1–4 mm)	*E. coli* K12 and *S. aureus*	Plasma jet, 6.8 kV, 1.5 kHz, Ar at 3.0 SLM, 20 mL of deionized water, 6.5 min	Samples were soaked in PAW at 4, 25, and 40 °C for 30, 45, and 60 min, respectively.	0.35 to 0.56 log for *E. coli*, 0.08 to 0.43 log for *S. aureus*	The color (L^*, a^*, and b^*), hardness, protein and lipid contents were well maintained.	Royintarat et al. (2020)
Fresh beef (3 × 3 × 1 cm, 10 g)	Total bacteria	Microplasma jet, 10kV, 8 kHz, air at 1.0 SLM, 40 mL deionized water, 30 min	PAW (1 mL) was sprayed on each piece of beef.	3.1 log CFU/g, extended shelf life by 4 to 6 d	The pH, TVB-N, lipid oxidation, cooking loss, color, and texture of beef were improved.	Zhao, Chen, et al. (2020)

DW, distilled water; SDW, sterile distilled water; SLM, standard liter per minute; TBARS, thiobarbituric acid reactive substances; TVB-N, total volatile basic nitrogen.

over the last decades. As shown in Table 3, PAW could effectively inactivate various spoilage and foodborne pathogenic bacteria present in raw meat and meat products. Besides, PAW could also effectively improve the quality attributes of meat products, including their nutritive value, pH, texture, color, and other sensory properties (Table 3).

Nitrite (NO_2^-) is commonly used in the curing of meat to develop cured color and flavor and to inhibit microorganism-related lipid oxidation and spoilage. PAW could be considered as an effective and innovative substitute for synthetic nitrite in cured meat manufacturing (Jung et al. 2017). Jung et al. (2015a) reported that PAW supplementation caused no noticeable influences on the total aerobic bacterial counts, color, and peroxide values of sausages compared with those of celery powder and sodium nitrite throughout 28 d of storage at 4 °C. Moreover, PAW-supplemented sausage had the lowest concentrations of residual nitrite and reached higher sensory scores than those of sausage with celery powder (Jung et al. 2015a). Similar results were also observed in PAW-treated meat products, such as beef jerky (Inguglia et al. 2020), meat batters (Jung et al. 2015b), and loin ham (Yong et al. 2018). The influence of PAW on loin ham surface color is shown in Figure 4b.

Eggshell disinfection

Foodborne *Salmonella* spp. is the main eggshell-associated pathogenic microorganism, causing complex public health

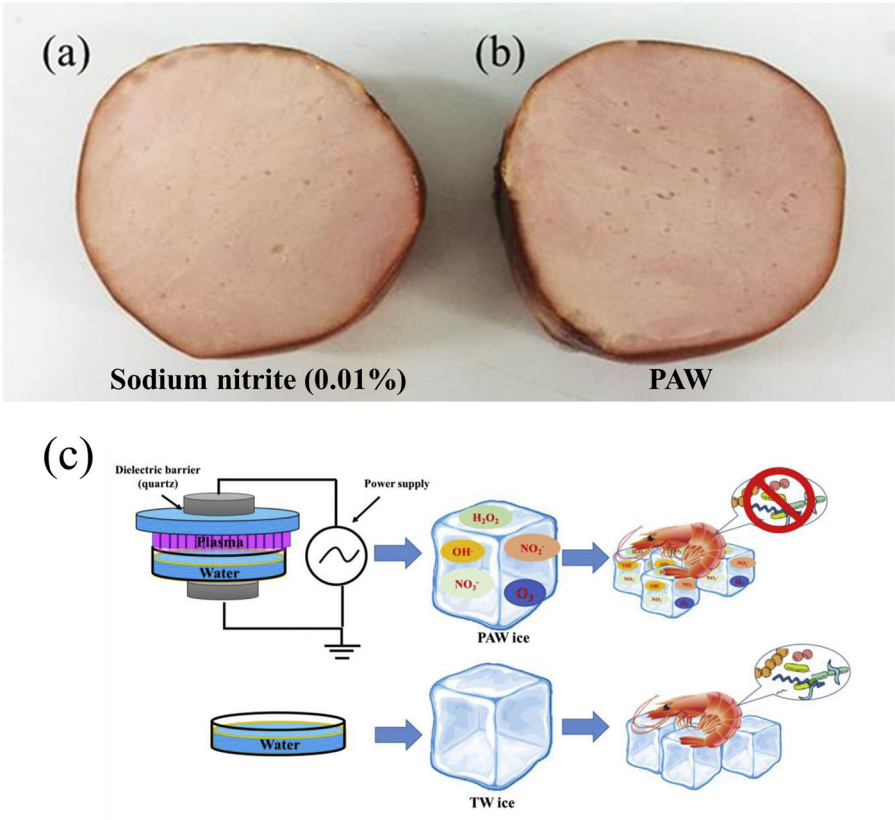

Figure 4. The visual appearance of loin ham cured with sodium nitrite (a) or PAW (b) (Yong et al. 2018) and (c) application of PAW-ice for preservation of shrimps (Liao et al. 2018).

concerns and economic problems worldwide. Lin et al. (2019) revealed that the 60-s treatment of PAW prepared with reserve osmotic water at 60 W for 20 min resulted in 4.40 $\log_{10}$ CFU/egg reduction of *Salmonella* Enteritidis, which was significantly higher than the 0.77 $\log_{10}$ CFU/egg reduction of sterile water treatment for 60 s. Furthermore, the freshness indices of the PAW-treated eggs were better than those of the chlorine-treated ones. These data indicate that PAW might be a better and more effective alternative to commercial methods for eggshell cleaning and disinfection.

Thawing media for enhancing microbiological safety

Freezing is one of the most widely used methods for meat, poultry, fish, and seafood preservation. Technical thawing for the above-mentioned products is necessary prior to cooking or further processing. There is a considerable opportunity for microbial growth in frozen foods during the slow defrosting process, which might pose an increased risk to human health (Leygonie, Britz, and Hoffman 2012). Liao, Xiang, et al. (2020) recently evaluated the application of PAW in the thawing of frozen beef. The authors found that the total aerobic bacteria, mold, and yeast contents of PAW-thawed beef were all significantly lower than those of air-, water-, microwave-, or slightly acidic electrolyzed water-thawed samples. Furthermore, PAW-supplemented thawing did not result in adverse effects on beef texture, pH, and color compared with other conventional methods. Therefore, PAW might be used as an active thawing media to maintain microbial safety and quality attributes of frozen products.

Seafood decontamination

Various kinds of seafood, such as oysters, salmon, and shrimp, are the most common cause of foodborne illness in the world. Liao et al. (2018) used PAW-ice for the preservation of fresh *Metapenaeus ensis* (Figure 4c). The total viable count of shrimps treated with PAW-ice increased from an initial 3.9 log CFU/g to 6.5 log CFU/g after storage at 5 °C for 9 d, while that of the tap water ice-treated samples increased to 8.6 log CFU/g by day 9. Moreover, PAW-ice also delayed the progress of melanosis in shrimps without any adverse effects on their quality.

PAW-ice has been used for the preservation of fresh salmon strips as well (Jiao et al. 2019). The final level of *L. monocytogenes* of salmon strips treated by PAW-ice was 3.6 log CFU/mL after storage at 4 °C for 5 d, significantly lower than the 5.1 log CFU/mL value of the control samples treated with sterile water-ice. PAW-ice could also effectively slow down the increase of volatile basic nitrogen (TVB-N) levels and the pH of fresh salmon strips. In a more recent study, Zhao, Ojha, et al. (2020b) demonstrated that the PAW treatment resulted in a 0.4-log reduction of *P. fluorescens* incubated on fresh mackerel fillets. Thus, PAW and PAW-ice could be used for seafood decontamination and preservation (Kulawik and Tiwari 2019).

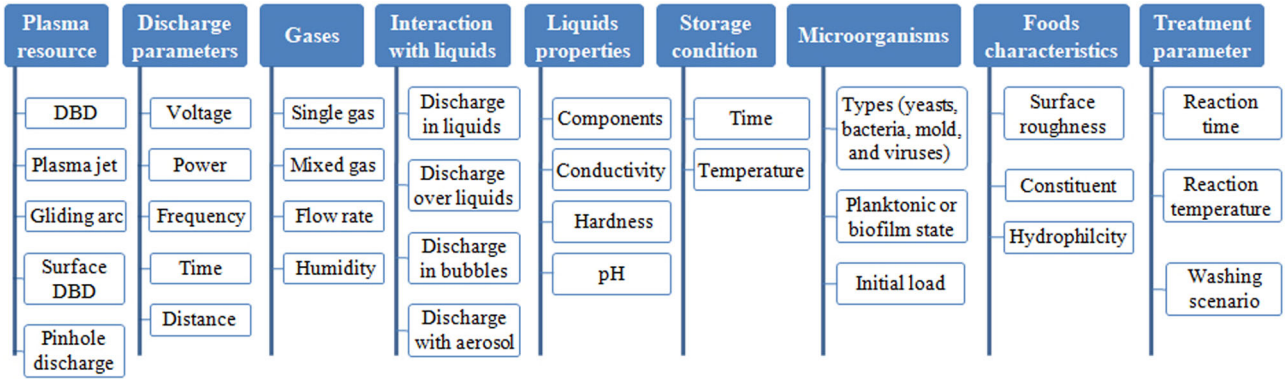

Figure 5. Factors influencing the antimicrobial efficiency of PAW.

Application of PAW in processed foods

PAW has also been successfully used for the decontamination and preservation of processed foods and ready-to-eat products, such as salted kimchi cabbage (Choi et al. 2019), tofu (Frias et al. 2020), Korean rice cakes (Han et al. 2020), and thin sheets of bean curd (Zhai et al. 2019). Choi et al. (2019) assessed the effects of PAW washing on the microbiological quality of shredded salted Chinese cabbages. After PAW washing, the maximum reduction was 2.0 log CFU/g for mesophilic aerobic bacteria, 2.2 log CFU/g for lactic acid bacteria, 1.8 log CFU/g for yeasts and molds, 0.9 log CFU/g for coliforms, 1.5 log CFU/g for *L. monocytogenes*, and 1.3 log CFU/g for *S. aureus*, respectively. PAW washing caused negligible changes in the moisture content, reducing sugar content, instrumental hardness, and color values of salted Chinese cabbage.

Recently, Han et al. (2020) reported that PAW treatment for 20 min reduced the number of total aerobes, *Candida albicans*, and *Penicillium chrysogenum* on Korean rice cakes by 2.78, 1.00, and 1.97 log CFU/g, respectively, which was statistically higher than the values obtained in the case of the distilled water-treated samples. Moreover, PAW effectively reduced the number of foodborne pathogens incubated on Korean rice cakes, including *E. coli* O157: H7, *S.* Typhimurium, and *L. monocytogenes*. The authors also showed that the color values, pH, and firmness of Korean rice cake were not changed significantly following the PAW treatment.

Soy-based foods, such as tofu and thin sheets of bean curd, are the most famous and popular foods in East Asian countries. However, tofu is a favorable medium for the growth of microorganisms due to its near-neutral pH, as well as high protein content and water content. Frias et al. (2020) confirmed that the microbial loads on PAW-treated tofu samples were reduced by 0.5–0.8 log CFU/g after 24 h of storage. The fresh-like texture and color properties of the PAW-treated tofu samples could be effectively maintained. Similar findings were also observed in PAW-treated thin sheets of bean curd, a traditional bean product in Southeast Asia (Zhai et al. 2019).

Sprout production

Sprouts, a kind of nutritious vegetable, have been widely consumed for centuries. Due to its high levels of reactive species and strong antimicrobial property, PAW exhibits great potential for sprout production practices (Fan et al. 2020; Lee et al. 2018; Lo Porto et al. 2018; Zhou et al. 2019). Lee et al. (2018) discovered that PAW could effectively promote the germination rate of soybean seeds and significantly increase the amount of ascorbate, asparagine, and γ-aminobutyric acid in sprouts. These results were in good agreement with the findings of Fan et al. (2020) and Zhou et al. (2019). Warm and humid conditions are needed for the sprouting of seeds, providing an ideal environment for the survival and rapid growth of foodborne pathogens, including *S.* Enteritidis, *E. coli* O157:H7, *B. cereus*, *L. monocytogenes*, and *S. aureus* (Yang et al. 2013). According to previous research, PAW watering could also effectively reduce the microbial loads on sprouts. For the sprouts treated by Air, O_2, He, and N_2-PAW, the microbial counts decreased by 5.17, 4.29, 2.80, and 2.04 log, respectively (Zhou et al. 2019). Therefore, as stated above, PAW might potentially contribute to the hygienic and economic production of sprouts.

Decontamination of food contact materials

Kamgang-Youbi et al. (2009) investigated the effect of PAW on the biofouling of microorganisms on 304 stainless steel and high density polyethylene (HPDE) surfaces, which are widely-used materials in the food industry. They discovered that PAW could inactivate the adherent bacterial cells in a time-dependent manner. The counts of *S. cerevisiae*, *H. alvei*, *Leuconostoc mesenteroides*, and *Staphylococcus epidermidis* adhered to stainless steel decreased by 3.07, 5.36, 4.69, and 6.09 log, respectively, upon PAW exposure for 30 min. Similar trends were also observed in the case of HPDE surfaces-adherent bacteria. Obvious morphological damage was observed in the cells of the four tested strains adhering to stainless steel and HPDE surfaces (Kamgang-Youbi et al. 2018).

In summary, the application of PAW and other PALs in the food industry has been extensively investigated in the past few years. Future research efforts shoud focus on the influences of PAW on consumer perception and the sensory attributes of foods (Gambaro 2018; Rocha et al. 2020).

Factors influencing PAW efficiency

Generally, PAW efficacy depends on multiple factors, such as the plasma exposure dose, types of target microorganisms,

treatment conditions, characteristics of food products, and storage duration and temperature (Figure 5).

Cold plasma discharge parameters

PAW is generated with certain types of gas discharges at atmospheric pressure. Therefore, the discharge parameters of the ACP treatment have a significant influence on the inactivation efficiency of PAW, including the types of electrical discharge, device geometries, electrode configurations, voltage, frequency, input power, and activation time.

Generally, the inactivation efficiency of PAW against microorganisms on food products increases simultaneously with the plasma exposure time (Guo et al. 2017; Lin et al. 2019; Schnabel et al. 2015; Xiang, Liu, et al. 2019). Longer activation times might promote the generation of reactive chemical species in PAW, which are mainly responsible for its antimicrobial activity (Ma et al. 2015; Sano et al. 2014). As demonstrated by Xiang et al. (2018), the concentrations of H_2O_2, NO_3^-, and NO_2^- in PAW increased significantly with increasing exposure time of water-to-plasma. In addition, the conductivity and ORP of PAW were also increased, while the pH value decreased simultaneously with the increasing exposure time of plasma (Xu et al. 2016).

Zhao, Ojha, et al. (2020a) discovered that the viable counts of *E. coli* cells were reduced by 0.1, 1.2, and 5.67 log after 1-hour exposure to PAW activated at 15, 22, and 30 kV, respectively. Similar findings were also reported by Qi et al. (2018) and Suganuma and Yasuoka (2018). For example, the populations of *S. putrefaciens* cells were reduced by 0.13, 0.38, and 2.0 log after exposure to PAW activated at 6, 7, and 8 kV for 30 min, respectively, while 1.75- and 2.0- log reduction were obtained in the case of PAW prepared at 10 and 12 kV for 25 min, respectively (Qi et al. 2018). Higher discharge voltage might result in higher levels of reactive chemical species generated in plasma and PAW (Qi et al. 2018). However, a higher discharge voltage might lead to reduced generation or increased degradation of certain reactive species, resulting in a decrease in the antimicrobial activity of PAW (Chen et al. 2019; Liu et al. 2020). As reported by Chen et al. (2019), the total aerobic bacteria on fresh-cut pears decreased only by 0.11 log after the treatment of PAW prepared at 10.0 kV and storage at 4 °C for 12 d, which was significantly lower than the 0.65 log decrease in the case of PAW activated at 8.0 kV. The mechanisms underlying the influence of discharge voltage on the inactivation efficiency of PAW should thus be further investigated in future. The antimicrobial activity of PAW could also be affected by the input powers for plasma activation (Tian et al. 2017). As revealed by Lin et al. (2019), the PAW prepared with higher power displayed stronger inactivation capacity. The pH values of PAW decreased, while the ORP values and the concentrations of H_2O_2, NO_3^-, and NO_2^- increased steadily with increasing discharge power (Lin et al. 2019; Suganuma and Yasuoka 2018).

Properties of working gas

The efficiency of PAW is significantly affected by the characteristics of the working and environment gas (e.g., the gas composition, humidity, and flow rate). Smet et al. (2019) discovered that the PAW generated using He + 1% O_2 exhibited greater antibacterial and antibiofilm activities than that generated using He. Similarly, PAW prepared with artificial air (21% O_2 + 79% N_2) showed stronger antifungal activity against *Colletotrichum gloeosporioides* spores than that prepared with air (Wu et al. 2019). The composition of the working gas might affect the electron-impact reactions in the main discharge and post-discharge regions, resulting in remarkable changes in the physicochemical properties and antimicrobial efficacy of PAW (Lai et al. 2020; Wu et al. 2019). For example, the air-PAW had a lower pH value but higher concentrations of ozone, and nitrite than that of oxygen-PAW (Wu et al. 2019).

Plasma interaction with liquids

The efficiency of PAW is also affected by the plasma discharge categories, as shown in Figure 1. As observed by Tian et al. (2015), PAW generated beneath the water surface exhibited stronger disinfection efficiency against *S. aureus* than that produced above the water surface, which was associated with ORP and conductivity.

Properties of target liquid solution

According to previous studies, the antimicrobial properties of PAW are affected by the characteristics of liquids, including liquid components, pH, hardness, and conductivity. The addition of exogenous substances might affect the physicochemical characteristics and efficacy of PAW. As shown by Ercan et al. (2013), the plasma-activated N-acetylcysteine exhibited stronger antimicrobial properties against *E. coli* than the plasma-activated deionized water or phosphate buffer saline (PBS). Moreover, the supplementation of methionine (Ercan et al. 2014), lactic acid (Qian et al. 2019), and H_2O_2 (Wu et al. 2017) could also effectively improve the antimicrobial activity of PAW.

The acidification of the plasma-activated solution plays an important role in PAW-induced microbial inactivation (Naïtali et al. 2010). Therefore, the efficacy of PAW is remarkably affected by the pH value and the buffering capacity of the solutions. Ma et al. (2020) reported that the *E. coli* count decreased from an initial 10^7 CFU/mL to levels below the detection limit after a 5-min PAW exposure, while the plasma-activated PBS only eliminated *E. coli* cells by 1.8 log under the same conditions. After the 5-min-lasting plasma treatment, the pH value of the plasma-activated deionized water decreased to 2.8, while that of the plasma-activated PBS slowly decreased to approximately 6 (Ma et al. 2020). This might be attributed to the buffering capacity of PBS and the phosphate buffer (PB) against the acidic chemicals (such as nitric acid, nitrous acid, and peroxynitrous acid) generated during plasma activation, thereby slowing

down the decrease in the pH value of PAL and significantly affecting the aqueous-phase chemistry of the reactive chemical species in the liquid. In contrast, the PALs generated with citrate-phosphate buffer or lactic acid with an initial pH of 3.1 exhibited higher inactivation efficacy against microbial cells than PAW, which might be due to the interactive effect of the reactive species and low pH (Joshi et al. 2018; Qian et al. 2019).

Water hardness and conductivity are important factors affecting the inactivation efficacy of PAW. As suggested by Lin et al. (2019), lower-hardness water is more suitable for PAW production. The PAW prepared using hard water showed low ORP and high pH values, which might weaken the inactivation efficacy of PAW. Lukes et al. (2008) reported that the production rate and the saturation level of H_2O_2 by the corona discharge in water significantly decreased with increasing solution conductivity (in the range of 100–500 μS/cm by the addition of dilute sulfuric acid to deionized water). Similar findings were also revealed in other studies, indicating that the hydroxyl radical intensity and the H_2O_2 generation rate decreased with increasing solution conductivity during pulsed electrical discharge (Shih and Locke 2011).

Storage conditions of PAW

Generally, the antimicrobial potential of PAW substantially decreases during storage. Shen et al. (2016) showed that fresh PAW resulted in an approximately 5 log reduction in the case of *S. aureus*. After being treated with PAW stored at 25 °C for 1 or 30 d, *S. aureus* decreased by 1.8 and approximately 1 log reduction, respectively. Similar results were also obtained by other researchers (Iwata et al. 2019; Lin et al. 2019; Traylor et al. 2011). The antimicrobial activity of PAW could be well maintained at lower storage temperatures. Shen et al. (2016) revealed that the bactericidal efficiency of PAW significantly decreased with increasing storage temperature (25, 4, −20, and −80 °C) for up to 30 d, which might be related to the changes in the concentrations of the long-lived reactive species (including H_2O_2, NO_3^-, and NO_3^-) and ORP.

Characteristics of microorganisms

Different types of microorganisms, such as bacteria, yeasts, and molds exhibit different PAW sensitivity patterns. Microorganisms could be placed in the following order on their decreasing resistance to PAW: Yeasts > Gram-negative bacteria > Gram-positive bacteria (Kamgang-Youbi et al. 2009; Ye et al. 2013), which is mainly due to the differences in the chemical composition and structure of their cell walls. The variations in bacterial isolates and phenotypes also have an impact on the inactivation efficacy of PAW. Lipovan et al. (2015) investigated the antibacterial effect of PAW against 38 isolates of coagulase-positive *Staphylococci* (27 were *S. aureus* and 11 were non-*S. aureus* strains) and *S. aureus* ATCC 25923. The reduction values of the 39 tested strains ranged from 0 to 7.38 with an average of 5 log after a 3-min treatment of PAW. Rodriguez et al. (2020) compared the PAW sensitivity of *E. coli* K12 cells with different resistance to H_2O_2. They discovered that the *E. coli* K12 cells with evolved resistance to H_2O_2 showed less sensitivity to the bactericidal effects of PAW compared to the wild-type strain.

Microorganisms can exist both in planktonic and biofilm state, which also significantly affects the antimicrobial efficacy of PAW. The bacteria within biofilms are less susceptible to PAW than their free-living counterparts (Hozák et al. 2018; Smet et al. 2019). As reported by Smet et al. (2019), the populations of planktonic *L. monocytogenes* and *S.* Typhimurium decreased by 5.3 and 5.8 log, respectively. Under the same conditions, the *L. monocytogenes* and *S.* Typhimurium cells within biofilms were only inactivated by 3.2 and 3.9 log reductions, respectively. The initial microbial load strongly affects the antimicrobial efficacy of PAW. Kamgang-Youbi et al. (2008) revealed that the inactivation rate k_{max} of PAW against *H. alvei* cells decreased when the initial viable population increased from 2×10^4 to 8×10^6 CFU/mL. Similar results were obtained by Bai et al. (2020) in their experiments showing that the inactivation efficacy of PAW against *B. cereus* spores was related to the initial spore concentration.

Food-related factors

PAW shows less inactivation activity against microbes on food products than against in vitro model, which might due to the influence of food constituents, such as proteins, lipids, and polysaccharides (Kang et al. 2019; Xiang et al. 2018; Zhao, Ojha, et al. 2020b). Zhang et al. (2016) and Bai et al. (2020) demonstrated that the addition of bovine serum albumin (BSA) into PAW could decrease the disinfection efficacy of PAW against *S. aureus* and *B. cereus* spores, which might be mainly attributed to the physical barrier effect and reactive species scavenging ability of BSA. Xiang, Kang, et al. (2019) also revealed that the inactivation efficiency of PAW decreased after being mixed with beef extract or peptone at a final concentration of 0 to 5.0 g/L at room temperature for 15 min. The authors further described that the beef extract and peptone supplementation significantly decreased the oxidation-reduction potential and NO_2^- concentration, while increased the pH values of PAW, which might contribute to the changes in the antimicrobial capacity of PAW. Thus, more attention should be paid to the effect of interfering substances on the disinfection efficacy of PAW before its practical application.

Surface roughness is an important factor influencing the inactivation efficacy of PAW. Microorganisms tend to attach to or be entrapped in the grooves or cavities of fruits, protecting the cells against washing or disinfection treatments (Wang et al. 2009). Royintarat et al. (2020) compared the antibacterial efficacy of PAW toward *E. coli* and *S. aureus* on rough and smooth chicken skin. Following the PAW treatment at 40 °C for 60 min, the *E. coli* K12 and *S. aureus* cells on the rough skin (at a thickness of 4 mm) decreased by 0.56 and 0.43 log, respectively. At an optimal PAW

treatment condition for smooth skin (40 °C, 60 min, and 1-mm thickness), the *E. coli* K12 and *S. aureus* populations were decreased by 0.35 and 0.08 log CFU/mL, respectively. Joshi et al. (2018) also reported consistent findings when assessing the inactivation effect of PAW against *Enterobacter aerogenes* incubated on grape tomatoes, limes, and spiny gourds.

Treatment conditions

Generally, the antimicrobial efficacy of PAW gradually increases with an increasing contact time (Bai et al. 2020; Xiang et al. 2018; Zhao, Ojha, et al. 2020a; Zhou et al. 2018). Similar results were also obtained when PAW was used for the decontamination and preservation of food products (Kang et al. 2019; Ma et al. 2015; Xiang, Liu, et al. 2019; Xu et al. 2016). In addition, the in vitro antimicrobial efficacy of PAW is improved with increasing exposure temperatures (Bai et al. 2020; Xiang, Wang, et al. 2019; Zhang et al. 2020). Similarly, PAW treatment at higher temperatures also exhibited stronger inactivation efficacy against microbes on food products, such as salted kimchi cabbage (Choi et al. 2019), rice (Liao et al. 2020), and grapes (Xiang et al. 2020).

The order of using PAW in the different steps of the washing process affects its antimicrobial activity (Andrasch et al. 2017; Fröhling et al. 2018; Schnabel et al. 2020). The most recent work of Schnabel et al. (2020) assessed the influence of different PAW-washing scenarios on the microbial inactivation in fresh-cut lettuce. Scenario 3 was the best, which consisted of 5 washing steps, pre-bathing with PAW, pre-rinsing with tap water, pre-washing with tap water, main washing with PAW, and post-rinsing with tap water. After the washing based on the above-described scenario, the total plate count on fresh-cut lettuce decreased by 4 to 5 log reduction in a lab-scale experiment, while the reduction was less than 2 log in the pilot-scaled experiment.

Combination of PAW with other technologies

PAW shows lower inactivation activity against microorganisms in food products than that in pure culture, which might be due to the interfering effects of food constituents (Zhao, Ojha, et al. 2020b). Therefore, PAW has been used in combination with other technologies, such as mild heat and ultrasound to enhance microbial inactivation by additive or synergistic effects (Royintarat et al. 2020; Zhang et al. 2020).

PAW combined with mild heat

Mild heat treatment, a short heat treatment at 40 to 60°C, is the most commonly used combined with other non-thermal technologies to improve microbial inactivation. According to previous reports, the inactivation efficiency of PAW against bacteria, bacterial spores, and yeasts was remarkably enhanced in combination with mild heat treatment (Bai et al. 2020; Liao, Bai, et al. 2020; Zhang et al. 2020). Xiang, Wang, et al. (2019) confirmed that a 7.28-log reduction in *E. coli* O157:H7 cells was achieved following PAW treatment at 50 °C for 4min, which was significantly higher than the 0.77 log CFU/mL after PAW treatment alone at 25°C for 4 min and the 0.75 log CFU/mL after mild heating at 50°C for 4 min. A similar conclusion was also drawn by Zhang et al. (2020) that PAW treatment at 50 °C exhibited markedly improved inactivation capacity against *S. cerevisiae*. Moreover, the enhanced microbial inactivation efficiency of PAW combined with mild heat was also observed in food products, such as salted kimchi cabbage (Choi et al. 2019), rice (Liao, Bai et al. 2020), and grapes (Xiang et al. 2020). For example, Xiang et al. (2020) observed that the synergistic treatment of PAW and mild heating (50 to 55 °C) could effectively inactivate *S. cerevisiae* incubated on grapes without any observable adverse effect on the firmness, color, total soluble solids, total phenolics, vitamin C, and antioxidant properties of samples. In another study, Choi et al. (2019) also observed that the sequential treatment of PAW and mild heating showed synergistic antimicrobial activity against bacteria, yeasts, and molds on salted Chinese cabbage.

PAW combined with ultrasound

As an environmental friendly and cost-effective technique, ultrasound has shown multiple applications in the food industry over the past few years (Gallo, Ferrara, and Naviglio 2018). A recent study published by Royintarat et al. (2020) examined the efficacy of PAW and ultrasound in the inactivation of foodborne pathogens in chicken meat and skin. The *E. coli* K12 and *S. aureus* populations on chicken breast decreased by 1.33 and 0.83 CFU/mL, respectively, which were higher than those of the PAW treatment alone under the same conditions (0.46 log CFU/mL for *E. coli* K12 and 0.46 log CFU/mL for *S. aureus*). The synergistic use of PAW and ultrasound resulted in slight changes in the color, hardness, protein and lipid contents, and the sensory score of the chicken samples.

Conclusions and future perspectives

The antimicrobial activity of PAW on a variety of foodborne microorganisms has been extensively investigated in recent years. The data collected so far suggest that PAW could be used for ensuring the microbiological safety and quality of food products. However, further research is still needed for the practical applications of PAW in food preservation and processing. First, the comprehensive safety assessment of PAW must be performed before its practical applications in the different fields of the food industry, and the regulatory guidelines should be established. Though various PAW-generation devices have been already developed over the years, the low yield and high production cost seriously hinder the application of PAW to improve the safety and quality of food products. Therefore, intense research efforts should urgently focus on the development and up-scaling of lower-cost PAW production approaches for its commercial applications. The generation mechanism of reactive species

in PAW is considered as one of the most important future research directions, which would potentially help to improve the antimicrobial capacity of PAW by optimizing the plasma process parameters for PAW production. In addition, the antimicrobial activity mechanisms of PAW should be further elucidated in more detail using multi-omics technologies, such as transcriptomics, proteomics, and metabolomics. Overall, as a promising environmental friendly and effective antimicrobial agent, PAW provides potential solutions to improve the safety and quality of food products.

Disclosure statement

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