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Potential of Electrolyzed Water as an Alternative Disinfectant Agent in the Fresh-Cut Industry

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Abstract Water disinfection is one of the most critical processing steps in fresh-cut vegetable production. Technologies capable for the efficient disinfection of process water and recycled water would allow reducing wastewater and have less impact on the environment. Among the chemical disinfectants, hypochlorite solutions are still the most widely used. Electrochemical disinfection of the wash water has been demonstrated to be effective in eliminating a wide spectrum of pathogens in process water. Both hypochlorite solutions and electrochemically produced chlorine compounds, in particular hypochlorous acid, are effective disinfectants when adequate doses are used. A new electrochemical process using borondoped diamond electrodes can generate additional reactive oxidant species than chlorine and further enhance the disinfecting capacity. However, there are pros and cons on the use of one or other disinfectant agents. In this review, the technological advantages and the limitations of electrolyzed water, particularly regarding the organic matter content, are discussed and compared to the use of hypochlorite.

Keywords Water disinfection · Sanitizers · Non-thermal water treatment · Process water · Microbiological control · Chlorinated by-products

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Introduction

The fresh-cut industry started in the United States in the early 1980s and grew rapidly, supplying fast-food restaurant chains and providing consumers with convenient products. The fresh-cut produce market has grown exponentially from bagged salads to other items such as carrots, onions, and spinach, and it is consolidated as one of the most important food sectors (Gorny 2005; James 2006; Floristán 2009).

Fresh-cut fruits and vegetables are unique foods because they are processed with minimal preparation. Produce can become contaminated with disease-causing pathogenic bacteria, viruses, and protozoan parasites in different steps throughout the supply chain (Beuchat 1996; EFSA 2013; Gil et al. 2015). To date, effective intervention strategies have been developed, but they cannot completely eliminate microbial food safety hazards associated with consumption of this uncooked produce. Preventing contamination of fresh fruits and vegetables by microbial pathogens is, therefore, the most effective strategy to assure that these foods are healthy and safe for human consumption (USFDA 2001).

Recently, the role of water as a contributing factor to foodborne disease has been recognized as an important and potentially impacting component during food production and processing (FAO/WHO 2008; EFSA 2013). This is particularly relevant for the fresh-cut produce industry. The fresh-cut industry requires a washing step, which usually represents a sanitizing step, to remove dirt, debris, and microorganisms responsible for quality loss and decay as well as to precool and remove cell exudates from the cut product (USFDA 2001). The process water should be of such good quality that it does not contaminate the produce, even with little water replenishment. To maintain the quality of the water, an effective disinfection agent or system should be used to destroy microorganisms of public health concern and other spoilage microorganisms, without adverse effect on the quality and safety of the product (FDA 1998). This review covers the problem of maintaining the microbiological quality of process water that comes into contact with produce in the washing tank and the water reconditioning of this process water to be reused in the fresh-cut production (Fig. 1).

A large number of review articles have been published recently concerning the efficacy of water disinfection treatments in reducing microbial population on the produce and in the water (Sapers 2001; Parish et al. 2003; Gil et al. 2009; Goodburn and Wallace 2013). Most of them agree that water disinfection treatment in the washing tank may not guarantee produce safety but it is capable of eliminating microbial population in the wash water, avoiding cross-contamination between produce tissues. Water disinfectants must not only be effective but they must also be compatible with processing practices and technical capabilities (Sapers 2003). In addition, water treatments must be affordable, safe to use, have no effect on product quality, and with approval from applicable regulatory agencies (Ölmez and Kretzschmar 2009). As mentioned before, disinfection treatments can be also used for water reconditioning outside the washing tank (Van Haute et al. 2013).

Problem Scope

Among the different industries, the food industry ranks third in water consumption and wastewater discharge rates coming after the chemical and refinery industries. In particular, in the fresh-cut industry, large volumes of water are commonly used during processing of fruits and vegetables (IFPA 2001). Basic production steps include washing raw materials, removing inedible portions, and cutting and washing the processed product. Process water is water resulting from the contact with raw materials, after washing and rinsing. The process water may contain organic compounds, pesticides, and bacteriological contaminations. Usually, large washing tanks (1–5 m³) are

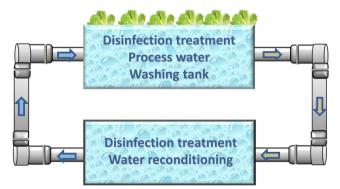


Fig. 1 Schematic representation of the water disinfection treatments applied in a fresh-cut processing line to treat process water within the washing tank, which is in contact with fresh produce, and for water reconditioning of process water that can be reused in the washing tank

used to wash and rinse fresh-cut products, and in most cases, the washing tank is refilled with fresh potable water to reduce cross-contamination. However, accumulation of organic matter and microorganisms in the washing tank cannot be completely avoided. Thus, the process water within the washing tank is characterized by high loads of both organic matter and microorganisms, unless a proper disinfection agent or system is in place. As defined by Suslow (1997), disinfection treatment of process water is to inactivate or destroy pathogenic bacteria, fungi, viruses, cysts, and other microorganisms in order to maintain the quality of process water in the washing tank and to reduce the risks associated with the reuse of process water after water reconditioning.

Currently, water reconditioning rate of process water to be reused in the fresh-cut industry is low because of the cost of the recycling water when water has high organic and bacteriological pollution. However, water reconditioning is still an option to reduce water and energy consumption, depending on the available disinfection treatment. Economic considerations and environmental concerns, including wastewater discharge regulations, make water reuse a valuable practice for the freshcut industry. In fact, some companies that are competing for increasingly scarce water supplies have found that it is more cost-effective to treat and reuse their process water than to locate new supplies (Wouters 2001; ILSI 2008). Among the regulatory requirements, the Codex guidelines provide the requirements for the water to be reused (Codex Alimentarius Commission 2004). Thus, in most cases, process water that is recycled or reused needs to be treated to improve its quality and to avoid any impact on the safety of the product. In general, a typical industrial process water treatment consists of a combination of physical, biological, and chemical processes to remove solids, organic matter, and microorganisms.

Disinfectant Requirements

The first step to choose a disinfectant for process water is to ensure that all necessary regulatory approvals are in place. In the United States, the legislation for process water disinfectant is complex as it may be regulated by the US Food and Drug Administration (FDA) and/or the USA Environmental Protection Agency (EPA) (IFPA 2001). The regulatory status of the disinfectants may depend on the type of product to be washed (i.e., processed vs. raw agricultural commodity) and the location where the disinfectant is to be used (e.g., field vs. processing facility). For fresh-cut produce, sanitizers are regulated by the FDA as a secondary direct food additive. For raw commodities that are washed in the fields, sanitizers are "pesticides" that are regulated by the EPA. In Europe, however, it is the biocides products Regulation (EU Biocides Regulation No 528/2012 2012) which covers a very diverse group of products, including disinfectants, pest control products, and preservatives.

Secondly, disinfectant agents or systems should be able to guarantee the innocuousness of the process water during production to avoid the risk of cross-contamination during washing. Wash water in tanks, re-circulated water, or water that is reused in a spray wash system can become contaminated with pathogens if contaminated product coming in from the field is washed in that water (IFPA 2001). Pathogens can survive for a relatively long period of time in water or in plant residues entrapped in processing line equipment and can subsequently contaminate clean product that passes through that water (Beuchat 1996; López-Gálvez et al. 2009). Disinfectants can be used to treat the water that contacts produce to prevent crosscontamination. However, not all the commercially available sanitizers are effective reducing the cross-contamination in the washing tanks. Table 1 shows a classification of different commercially available disinfection treatments based on their ability to inhibit cross-contamination in a washing tank.

The amount of water consumed to guarantee the safety of the washing system is also a great concern. The adoption of lower water consumption systems is needed to improve water management for the fresh-cut industry. In fact, the competitiveness of the whole agri-food industry and the sustainability of food production depend on many factors one of them being the use of water. Current food production and processing systems, especially for the small and medium-sized enterprises (SMEs), need to be revised and optimized with the aim of achieving a significant reduction in water usage. The development of new technologies, which enable the reduction of water and disinfectant, is essential.

The operating cost of the different disinfectant agents or systems is also another concern. Details of the operating cost for the most commonly used wash water disinfection systems used in produce operations were presented in the review on intervention strategies to reduce or eliminate pathogen contamination in the production of safe fresh-cut products (Parish et al. 2003). Based on this, the annual cost of chlorine from sodium hypochlorite (NaOCl), considering the capital and reagent costs, to treat 23,000 m³ was about 2700€, while the cost using chlorine gas was less than half (1125 \in). This data was determined on the basis of several assumptions, which might have varied over time. Nowadays, the annual operating cost of chlorine from NaOCl to treat the same amount of water $(23,000 \text{ m}^3)$ in a medium size processing plant is $4800 \in$. Therefore, the fresh-cut industry needs to find alternative technologies that can ensure produce safety but do not increase the operating cost.

Chlorine Supremacy

Chlorine is the most widely used water disinfectant because it is highly effective, inexpensive, and available to control foodborne disease. To date, no other sanitizing agent has competed with chlorine in all areas needed for safe food

 Table 1
 Classification of commercially available disinfectant agents based on their ability to inhibit cross-contamination in process water from the fresh-cut industry

Disinfectant	Process water (COD mg/L)	Classification	References
Sodium hypochlorite	Tap water	Good	Luo et al. 2011, 2012
(≥ 5 ppm)			Tomás-Callejas et al. 2012
			Van Haute et al. 2013
	COD=500	Good	Van Haute et al. 2013
			Gómez-López et al. 2014
	COD=750	Good	López-Gálvez et al. 2010b
	COD=1000	Good	Van Haute et al. 2013
Chlorine dioxide	Tap water	Good	López-Gálvez et al. 2010b
(≥ 3 ppm)			Pao et al. 2007
			Tomás-Callejas et al. 2012
Electrolized water	COD=3-14	Failure	Ongeng et al. 2006
Electrolized water	COD=500	Failure	Gómez-López et al. 2015
(<1 ppm free chlorine) pH=6.5			
Electrolized water + 0.5 % salt	COD=500	Good	Gómez-López et al. 2015
$(\geq 5 \text{ ppm free chlorine}) \text{ pH}=6.5$			
Peroxyacetic acid	500-700	Good	López-Gálvez et al. 2009
(≥ 60 ppm) pH=4.3			
Lactic acid	500-700	Failure	López-Gálvez et al. 2009
(20,000 ppm) pH=2.2			

production (McLaren 2000; Gómez-López et al. 2014). Chlorine is also used extensively as a disinfectant in wash, spray, and flume waters in the raw fruit and vegetable industry (Suslow 2001).

The three forms of chlorine commonly used for fresh fruits and vegetables are chlorine gas, calcium hypochlorite (Ca(ClO)₂), and NaOCl. Chlorine gas is generally restricted to use in very large operations and requires automated injection systems with in-line pH monitoring. It is highly effective in situations in which soil, plant debris, and decayed fruit or vegetables may enter in the early stages of washing and grading. The other two hypochlorite solutions are the most common sources of chlorine used for disinfection of produce and process water. Another commonly used sanitizer is called chlorine or chlorinated water that consists of a mixture of chlorine gas (Cl₂), hypochlorous acid (HOCl), and hypochlorite (OCl-). The antimicrobial activity of chlorinated water depends largely on the amount of HOCl present in the water but also on the generation of Cl₂ and OCl⁻ which depends on the pH (White 1992; Suslow 1997). Some details of chlorine chemistry have been discussed in Waters et al. (2012).

The terms free chlorine (FC), reactive chlorine, residual chlorine, and available chlorine are used to describe the amount of chlorine in any form available for oxidative reaction and disinfection. The choice of chlorine source involves technical and practical aspects such as stability of active substances and concentrations of active substances for longer periods (Suslow 2001). The amount of chlorine needed for disinfection of water depends not only on the pH but also on the amount and kinds of inorganic (particularly ammonia, nitrites, iron, and manganese) and organic (particularly amino acids and simple proteins) substances present in water (Suslow 1997). When organic matter is present in the process water, pathogen inactivation efficacy is significantly dependent on the FC, which directly relates to both the initial FC concentration and the organic load (Shen et al. 2013). This phenomenon is common to all types of chlorine-based sanitizers. Ayebah et al. (2006) found bactericidal activity of electrolyzed water (EW) decreased with increasing organic matter (chicken serum). Composition of the process water is an important factor in FC loss. Protein and phenolic compounds cause significant chlorine loss of chlorinated water, whereas carbohydrates, fat, and mineral have no significant effect on chlorine loss (Waters and Hung 2014). Prevention of pathogen survival in chlorinated process water can be achieved by maintaining sufficient FC concentration at all times. However, maintaining a relatively consistent level of FC during commercial fresh-cut wash operations is a technical challenge in practice because of the quick reaction of FC with organic materials in the produce wash tank (Suslow 2013).

In addition, chlorine is a disinfectant that has certain health and safety limitations. Chlorine oxidizes certain types of organic matter to produce undesirable disinfection by-products (DBPs) in process water, such as chloroform (CHCl₃) or other trihalomethanes (THM), that have known or suspected carcinogenic potential (Richardson 2003). The THMs of concern in water disinfection are chloroform, bromodichloromethane, dibrochloromethane, and bromoform; the first two have been classified by the WHO's International Agency for Research on Cancer (IARC 1999a, 1999b) as possibly carcinogenic to humans. Studies carried out by Gómez-López et al. (2013a) to determine the potential THM formation during washing of fresh-cut leafy greens demonstrated that after 1 h of using low doses of NaOCl, as a disinfectant agent in process water with high organic matter (>500 mg O_2/L of chemical oxygen demand, COD), the total THM values (194.0±29.6) in the process water were over the authorized limit fixed for drinking water (EU 98/83 1998; USEPA 2009).

There is no current legal limit for THM concentrations in fresh-cut washing tanks. Considering the legislation for water intended for human consumption, the limit fixed by the European legislation of total THMs is 100 µg/L (EU 98/83 1998) and 80 µg/L for the American legislation (USEPA 2012). Even though much concern has been raised over the presence of THMs when chlorine-based sanitizers are used for washing fresh-cut produce, the evidence accumulated so far indicates that their concentration in commercial salads poses no risks to human health (COT 2006). Recent studies have also demonstrated that the presence of THMs in process water does not affect the wholesomeness of the end product (López-Gálvez et al. 2010a; Gómez-López et al. 2013a; Van Haute et al. 2013). At laboratory level, even though THMs can be present at high levels in the process water (>120 μ g/L), their concentration falls to undetectable level ($<5 \mu g/L$) in the fresh produce after rinsing (Gómez-López et al. 2013a; Van Haute et al. 2013), unless very extreme conditions are applied such as the use of 700 mg/L sodium hypochlorite combined with the presence of very high organic load (1800 mg O₂/L chemical oxygen demand) (López-Gálvez et al. 2010a).

At high pH, chlorine reacts with organic nitrogen-based materials to produce chloramines. Waters and Hung (2014) measured the THM reaction products from chlorinated water at different pH with resorcinol. They found chloroform accounts for the largest percentage of THMs detected in all samples (>99 %), indicating that it is the main THM product when chlorine reacts with organic materials. The data indicates that the pH 6 and 9.3 chlorinated water when reacting with resorcinol produces approximately 10 times more chloroform (9.54 and 12.4 mg/L, respectively) than chlorinated water at pH 2.5 (1.1 mg/L). They also found pH of chlorinated water affects the rate of chlorine by-product formation, as well as the type of chlorine by-product formed. Increases in pH lead to the formation of chloroform as a chief THM product. The formation of chloramines during washing of fresh-cut produce has not been fully addressed up to now. Another group of DBPs of concern in water are the haloacetic acids, composed by monochloroacetic acid, monobromoacetic acid, tribromoacetic acid, dichloroacetic acid, and dibromoacetic acid; the latter three have been classified as possibly carcinogenic to humans (IARC 2013, 2014). As for THMS, there is no current legal limit for concentrations in fresh-cut washing tanks, but for water intended for human consumption, a limit has been set by the US-EPA at 60 μ g/L for five haloacetic acids (USEPA 2012).

Chlorate is another by-product of using chlorine or chlorine dioxide for the disinfection of water (Kaufmann-Horlacher et al. 2014). A detailed presentation of the problem of chlorate residues and the analytical technique can be found in their article. Kaufmann-Horlacher et al. (2014) analyzed fruit, vegetable, and cereal samples for the presence of chlorate in a special state wide monitoring program. Among the 1087 samples analyzed, 24.5 % have been found to contain chlorate residues in a range of 0.01 to 2.7 mg/kg considered "unfit for human consumption" and thereby judged as "unsafe food" in accordance with Article 14 of Regulation (EC) 178 (EU 2002), in which the general principles and requirements of food law in Europe were established.

Partially due to the possible generation of DBPs, the use of chlorine in fresh-cut produce washing is prohibited in some European Union countries such as Germany, Switzerland, the Netherlands, Denmark, and Belgium (Van Haute et al. 2013).

Electrolyzed Water

The main disadvantages regarding the use of chlorine are the risks associated with the storage, shipping, and handling of chlorine, which require increased safety regulations. Guidelines covering recommendations about the handling and use of chlorine have been published by US-EPA (USEPA 1999) and the Canadian Food Inspection Agency (CFIA, 2014). In contrast with the problems of hypochlorite of skin and membrane irritation and toxicity, EW is not corrosive to skin, mucus membrane, or organic material (Huang et al. 2008). EW or electrochemically activated water (ECA or ECAW) is a solution generated by passing a dilute salt solution (sodium chloride, NaCl, and potassium chloride, KCl, are commonly used) through an electrolytic cell (Fig. 2). The anode side of an electrolytic cell, from which acidic EW is obtained, produces various chlorine compounds and ions such as HOCl, OCl⁻, and Cl₂ gas. It can be produced on-site at the concentration ready to use and hence avoid many of the risks associated with chlorine mentioned above.

There are many different types of EW generators to produce EW with different water properties (e.g., pH, chlorine concentration, ORP, chloride ion). Waters et al. (2012) gave a comprehensive review on EW generation methods and their respective water properties. In a conventional electrolysis

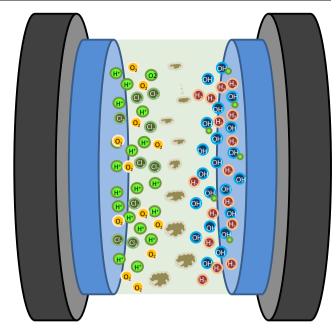


Fig. 2 Schematic representation of a electrolytic cell where a dilute salt solution containing organic matter pass through the boron-doped electrodes generating oxidative species which will degrade the organic matter

process, a dilute salt solution is electrolyzed with a membrane partition, resulting in the production of acidic EW at the anode and alkaline EW at the cathode (Umimoto et al. 2013). The membrane acts to separate anolyte and catholyte, and it stops the displacement of the ions and feed the current by electrons. The anolyte is charged in oxidative species and protons H providing acidity and the catholyte is charged in reductive species and OH⁻ providing alkalinity. Acidic EW has been shown to be effective in killing foodborne pathogens under in vitro conditions and in reducing microbial counts and pathogens in vegetables (Graça et al. 2011). Depending on its pH, EW is commonly classified into acidic EW (pH 2.2 to 2.7), slightly acidic EW (pH 5.0 to 6.5), and neutral EW (pH 6.5 to 7.5). The production of slightly acidic to neutral EW with conventional systems containing a membrane usually requires electrolysis of hydrochloric acid itself, use of hydrochloric acid as a pH adjuster, or an additional process, such as the mixing of the two types of EW in different amounts (Umimoto et al. 2013). Recently, the development of new electrolysis systems with and without the use of membranes has been shown to be efficient to generate acidic to neutral EW (Gómez-López et al. 2013b; Umimoto et al. 2013). In the new developed systems without the use of membranes, the species (oxidative species and protons H^+ as well as reductive species and OH) are generated and automatically mixed so the pH is not strongly influenced, but it will depend on the mixture process. Therefore, several types of EW can be generated based on the pH of the final solution. Experimental studies included in this review article refer to the different types of EW including acidic, slightly acidic, and

neutral, and whenever possible, the pH of the solution is indicated.

In the fresh-cut industry, electrolyzed water (EW) is the sanitizer created by adding small amounts of NaCl to the washing water subjected to electrolysis. Under these conditions, hypochlorous acid (HOCl) is generated (Izumi 1999). Thus, the use of EW as a sanitizing agent for produce is considered a special case of chlorination. For this reason, the use of EW will only be allowed in countries where the use of chlorine is allowed such as the United States, where it can fall within the frame regulation 21 CFR 173.315, chemicals used in washing or to assist in the peeling of fruits and vegetables (FDA 2013), and some European countries such as Spain and England. The use of acidic EW as a food additive has been approved in Japan since 2002 (Yoshida et al. 2004).

Several studies have shown the potential of EW for the disinfection and improvement of physicochemical quality of water, including drinking water (Martínez-Huitle and Brillas 2008) and wastewater from industrial and domestic activities (Chen 2004; Ongeng et al. 2006; Anglada et al. 2009; Schmalz et al. 2009; Poyatos et al. 2010). Regarding the fresh-cut industry, since 1999, many reports have demonstrated the potential use of EW as a chlorine substitute for washing fresh fruits and vegetables (Izumi 1999; Koseki and Isobe 2007; Hricova et al. 2008; Pangloli et al. 2009; Hung et al. 2010; Pangloli and Hung 2011, 2013a; Kim and Hung 2012; Olaimat and Holley 2012). Recent studies carried out by our group have evidenced that the use of this technology in the fresh-cut processing line could represent a good alternative to NaOCl (López-Gálvez et al. 2012; Gómez-López et al. 2013a, 2013b). The best way to demonstrate the potential use of EW as a disinfection treatment for process water is determining its potential to avoid cross-contamination in the washing tank. Dynamic systems that allow the continuous addition of organic matter and disinfectants are recommended to test disinfectant treatments (Fig. 3).

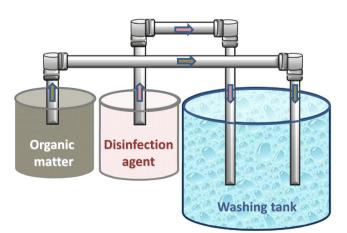


Fig. 3 Schematic representation of dynamic system used for disinfection treatments simulating a washing tank from the fresh-cut industry

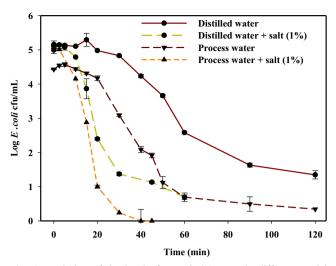


Fig. 4 Evolution of the level of *E. coli* O157:H7 in different model waters during treatment with the electrochemical disinfection. Model waters treated were distilled water, distilled water supplemented with salt (1 %), process water containing 500 mg/L of chemical oxygen demand (COD), and process water (COD=500 mg/L) supplemented with salt (1 %). The values are shown as *symbols* connected by a *solid line* and *vertical bars* represent the standard deviation

Regarding this, the efficacy of electrochemical disinfection to inactivate *Escherichia coli* O157:H7 in process wash water containing organic matter has been demonstrated (López-Gálvez et al. 2012). Figure 4 shows experimental results obtained when electrolyzed water was generated using boron-doped diamond (BDD) electrodes with and without the addition of salt. The effect of different operating conditions including current density, recirculation flow rate, and electrode doping level to inactivate microorganisms and decrease COD has been determined for lettuce process water under dynamic conditions (Gómez-López et al. 2013b, 2015).

Mechanisms of Action

There is a long-standing controversy about which is the primary factor of EW causing microbial inactivation. Many authors agree that the electrochemical treatment has two mechanisms of action: direct oxidation at the anode surface and indirect oxidation in the bulk solution by oxidants produced from the substances present in the water (Anglada et al. 2009). Between the two mechanisms of action, indirect oxidation seems to be predominant and many researchers believe that the existence of chlorine species is the main factor related to microbial inactivation by EW (Nakagawara et al. 1998). Although redox potential (ORP) has been considered as the main factor affecting the EW efficacy according to Kim et al. (2000a, b) and Liao et al. (2007), most of the studies correlated EW efficacy with the formation of chlorine species (Nakagawara et al. 1998). However, it must not be forgotten that other antimicrobial substances are generated during water electrolysis, and even chloride-free water can possess antimicrobial properties due to generation of reactive oxygen species such as hydroxyl radicals and ozone (O_3) (Jeong et al. 2006).

Several studies have addressed the mechanism of microbial inactivation by EW. These include the increase of membrane permeability together with leakage of cellular components and the decrease of activity of several key enzymes in bacteria such as E. coli and Staphylococcus aureus (Liao et al. 2007; Zeng et al. 2010) as well as the yeast Candida albicans (Zeng et al. 2011). Degradation of chromosomal DNA has been also observed in Pseudomonas aeruginosa (Kiura et al. 2002) and cause aggregation of essential bacterial proteins (Winter et al. 2008) or interfering bacteria cell protein composition (Cloete et al. 2009). Tang et al. (2011) studied the disinfection of Bacillus subtilis var. niger and found EW decreased dehydrogenase activity, intensified membrane permeability, elevated suspension conductivity, and caused leakage of intracellular K^+ , protein, and DNA, indicating a damage of cell walls and membranes.

Inactivation of Foodborne Pathogens

Regarding foodborne pathogens, many studies have described the efficacy of EW in killing pathogenic microorganisms in vitro as well as inoculated onto vegetable surfaces; however, EW has shown the same limitations as other disinfectants on the inactivation of microorganisms in whole and fresh-cut vegetables (Gómez-López et al. 2008a). Several studies have attributed EW a higher decontaminant effect than chlorinated water at the same FC concentration. For example, EW with 50 mg/L FC was as effective as chlorinated water with 120 mg/L FC in reducing Salmonella, Listeria monocytogenes, and E. coli O157:H7 counts on lettuce surface (Abadias et al. 2008). Likewise, Hung et al. (2010) observed that EW was more than or as effective as chlorinated water in reducing E. coli O157:H7 populations on strawberries and broccoli, with >1 log cfu/g reductions when testing at 4 °C and 1 or 5 min soaking time, while Park et al. (2001) observed that EW was as effective bactericide as chlorinated water in reducing E. coli O157:H7 and L. monocytogenes on lettuce. On cut apple, EW was equally or more effective than HOCl in decontamination of fresh-cut apple against E. coli O157:H7, Listeria innocua, and Salmonella choleraesuis (Graça et al. 2011). On the other hand, studies carried out in our groups showed that fresh-cut spinach treated with equal FC concentrations using NaOCl or EW showed similar reductions (about 1 log cfu/g) of psychrophilic populations (Gómez-López et al. 2013a).

Technological Advantages

Great differences have been found in the efficiency of FC production between different electrode materials at low

chloride concentrations. There are efficient electrode materials such as titanium electrodes with mixed oxide coatings based on iridium and/or ruthenium oxide and doped diamond electrodes (Kraft 2008). In particular, new electrode materials such as boron-doped diamond (BDD) make electrochemical disinfection a more promising process (Polcaro et al. 2007). Diamond electrodes are known to achieve high efficiency due to the in situ electrogeneration of electrochemical oxidants from water and dissolved substances. Such oxidants are shortlived free radical species (Jeong et al. 2006) and more stable substances such as chlorine species, O₃, and hydrogen peroxide (H₂O₂) (Furuta et al. 2005). Several studies have attributed the effectiveness of the BDD electrodes to the simultaneous presence of reactive oxygen species (ROS) which could give synergetic oxidant effects sufficient to perform an effective disinfecting process without the mediation of active chlorine products (Cho et al. 2004). However, Schmalz et al. (2009) demonstrated that the direct bacterial inactivation of chloridecontaining wastewater by the ROS (mainly •OH) radicals generated during electrochemical disinfection using BDD electrodes was negligible. On the other hand, it should be taken into account that electrochemical disinfection is largely dependent on cell configuration as well as the flow rate and the current density (Gómez-López et al. 2013b). The inactivation rate accelerates with increasing current density caused by a faster generation of electrochemical oxidants.

Limitations

The main factors affecting EW efficacy against microorganisms are the organic matter content of the water, pH, temperature, agitation, and water hardness. In the case of EW, the organic matter decreases its efficacy (Oomori et al. 2000; Park et al. 2009; López-Gálvez et al. 2012) because it reacts with FC reducing chlorine availability for disinfection. The efficacy of EW depends on pH since its antimicrobial action relies mainly on chlorine, whose most active species is HOCl, with pH determining the most important species: Cl₂ below pH 3 (typical of acidic EW), and HOCl and ClO⁻ above pH 4 (typical of NEW), with the highest antibacterial effect at pH 4 (Nakagawara et al. 1998; Len et al. 2000; Park et al. 2004). It has been reported that the bactericidal efficacy of EW increases with temperature in vitro (Rahman et al. 2010) and in vivo (Koseki et al. 2004). However, the results should be interpreted with precaution because of the range of temperatures applied during tests. While in the "in vitro" test no significant differences were observed between 4 and 23 °C for common pathogenic bacteria, differences were detected between 35 and 50 °C, which are uncommon temperatures in fresh-cut processing lines (Rahman et al. 2010). Similar results have been reported for fresh-cut lettuce inoculated with E. coli O157:H7 in which no significant differences were found for the efficacy of the electrochemical disinfection

between 4 and 20 °C (Koseki et al. 2004). Agitation facilitates inactivation of microorganisms on surfaces, probably due to microbial detachment and better contact between microorganisms and sanitizer (Park et al. 2002). While agitation is common in the fresh-cut washing industry due to the constant flow of water, it should be taken into account that it promotes chlorine losses probably because it accelerates the interface mass transfer of chlorine gas (Len et al. 2002). The chlorine losses can be expected to be similar to those of chlorinated water at a regular pH (6.5), but higher at the pH of acidic EW, which moves the chlorine species equilibrium towards chlorine gas. The effects of hardness and pH of water used to prepare EW solutions on the bactericidal activity have also been examined (Pangloli and Hung 2013b). Results indicated that free chlorine levels of EW significantly increased with water hardness. The increase of water hardness might have increased the concentration of electrolytes and conductivity or electrical current in the solutions and hence more chlorine production (Pangloli and Hung 2013b). These authors reported that increasing water hardness from 0 to 50 mg/L significantly increased the reduction of E. coli O157:H7 from 5.8 to 6.4 log cfu/mL.

As a chlorine-based disinfectant treatment based on FC, EW generates disinfection by-products originated from ClO⁻ and HOCI when combined with organic matter. Therefore, it could be expected that all the aforementioned disadvantages associated with the use of hypochlorite regarding the accumulation of disinfection by-products when treating process water containing high organic load can also be applicable to EW. However, there are only few studies evaluating the formation of disinfection by-products in process water when treated with EW (Table 2). Studies carried out by our group (Gómez-López et al. 2013a) have evidenced that total formation of THM was similar in process water obtained from a baby spinach processing line when treated with sodium hypochlorite and EW combined with 1 % of NaCl. However, preliminary data obtained in process

water obtained in the washing tank of fresh-cut iceberg lettuce showed that the formation of THM in process water containing around 1500–2000 mg/L of COD and treated with EW was 10 times higher than in process water treated with sodium hypochlorite. Therefore, depending on the disinfection treatment, the formation of disinfection by-products could vary depending on the organic matter released from the product exudates.

Impact of EW on Fresh Produce Quality

A diverse range of chlorine concentrations have been applied for produce washing with 50–100 mg/L hypochlorite remaining the most commonly used dose (Erkmen 2010). As a special case of chlorination, microbial reductions reported in fresh produce washed with EW are similar to those described for other chlorine-based sanitizers such as NaOCl within the range of 0.5–2.5 log cfu/g, depending on the working conditions. In general, all the studies carried out to determine the impact of EW on the quality of fresh produce agreed that EW can be used for a wide range of fresh and fresh-cut products such as carrots, bell peppers, radish, potatoes, lettuce, tomatoes, strawberries, cucumbers, spinach, broccoli, and kalian-hybrid broccoli (Izumi 1999; Koseki et al. 2001, 2004; Bari et al. 2003; Gómez-López et al. 2013a; Martínez-Hernández et al. 2013; Navarro-Rico et al. 2014).

Based on results from different authors, no generalization can be established for the effect of EW on the respiration rate of produce because of the high variability among studies. For example, EW, which has been reported to increase the respiration rate (RR) of iceberg lettuce (Koseki and Itoh 2002), had no effect (Vandekinderen et al. 2009a) or decreased RR (Rico et al. 2008) in other studies. Very different results have also been reported for white cabbage after washing with the treatment, where an increase (Koseki and Itoh 2002), no change (Gómez-López et al. 2007), or even a decrease in RR (Vandekinderen et al. 2009b) have been observed. These

Table 2 THM formation previously reported in process water after treatment with different chlorine-based disinfectant agents

Disinfectant	Process water (COD mg/L)	THM formation (µg/L)	Reference
Sodium hypochlorite (1–2 ppm free chlorine) pH=6.5	Tap water COD=500	<6.3 27.8±5.4	Van Haute et al. 2013
	COD=1000	124.5±13.4	
Sodium hypochlorite (2–4 ppm free chlorine) pH=6.5	COD=500	194.0 ± 29.6	Gómez-López et al. 2013a
Sodium hypochlorite (100 pm free chlorine) pH=6.5	COD=700	217.0 ± 38.0	López-Gálvez et al. 2010a
Sodium hypochlorite (700 pm free chlorine) pH=6.5	COD=1800	$3618.0 {\pm} 633.0$	López-Gálvez et al. 2010a
Sodium hypochlorite (40 ppm free chlorine) pH=2.5	Tap water	$0.9 {\pm} 0.5$	Waters and Hung 2014
Sodium hypochlorite (40 ppm free chlorine) pH=6.0	Tap water	$0.8 {\pm} 0.6$	Waters and Hung 2014
Electrolyzed water (2-4 ppm free chlorine) pH=6.5	COD=500	50.2±2.1	Gómez-López et al. 2013a
Electrolyzed water+1 % salt (2–4 ppm free chlorine) pH=6.5	COD=500	125.9 ± 15.4	Gómez-López et al. 2013a
Chlorine dioxide (3.7 ppm chlorine dioxide) pH=7.1	COD=700	<5.0	López-Gálvez et al. 2010a

controversial results could be due to different conditions applied during washing (e.g., FC concentration, contact time, temperature, and pH) or to the fresh produce (e.g., variety, maturity stage).

The desirable effect of disinfectants on the sensory properties of fresh-cut vegetables is to preserve quality and slow down deterioration. EW (50 mg/L FC) did not significantly affect the quality characteristics such as color and general appearance as well as visual quality (Izumi 1999; Park et al. 2001; Abadias et al. 2008; Gómez-López et al. 2008b) of fresh-cut lettuce and carrots. However, when EW was carried out at higher FC concentrations (240 mg/L), it caused detrimental effects on fresh-cut lettuce resembling leaf burn although showing a significantly higher reduction of *E. coli* 0157:H7 (Koseki et al. 2004). In baby spinach, in general, no significant changes were found in the overall quality between NaOC1- and EW-treated product although quality decreased during storage regardless of the sanitizing treatments (Gómez-López et al. 2013a).

The impact of electrochemical disinfection on the content of nutrients and phytochemicals of fresh produce has been addressed only in few studies. The results demonstrated an irrelevant effect. For example, total phenols and antioxidant capacity were not affected after washing fresh-cut carrots and lettuce with electrochemical disinfection and NaOCl containing 4, 20, and 30 mg/L of FC in process water (Vandekinderen et al. 2008, 2009a, b). Washing fresh-cut pineapple in acidic EW for 2 min followed by 2 % NaCl for 1 min did not cause any detrimental effect on vitamin C content (Raiputta et al. 2013). According to this, Gómez-López et al. (2013a) reported no significant differences on the vitamin C content of baby spinach leaves washed in process water with electrochemical disinfection and NaOCl. However, when the FC concentration increased to 4.4 using EW, reductions of vitamin C were reported after washing (Gómez-López et al. 2013a). In general, most of the available literature agrees that the antioxidant content of leafy greens is not greatly affected after washing in chlorine-based sanitizer when relatively low FC concentrations (\leq 50 mg/L) are used. In fact, in many studies, the vitamin C content remained relatively stable after a decontamination treatment. This may be attributed to the activation of an antioxidative system that promotes the biosynthesis of vitamin C from the carbohydrate pool (Pérez et al. 1999; Vandekinderen 2009).

Conclusions

Based on the available data, the disinfection capacity of EW for process water is clear. EW has been described as an ecofriendly technology due to the use of simple and nonhazardous raw materials such as water and NaCl. Due to the instability and highly reactive nature of ClO⁻ and HOCI, it will disappear rapidly when entering the environment. Oxidized molecules have been recognized as the main byproducts of ClO⁻ and HOCI use. However, chlorinated species are considered as the most important to assess from an environmental point of view. Additionally, mineralization of organic matter occurring during electrolysis can decrease pollution caused by wastewater discharges.

Therefore, within chlorine-based disinfectant treatments, EW can be considered as a chlorine substitute that could be used in those countries where chlorinated water is allowed.

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