LWT - Food Science and Technology 64 (2015) 337–343

Contents lists available at ScienceDirect

LWT - Food Science and Technology

journal homepage: www.elsevier.com/locate/lwt

Coagulation of turbidity and organic matter from leafy-vegetable wash-water using chitosan to improve water disinfectant stability

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ARTICLE INFO

Article history:
Received 14 October 2014
Received in revised form
29 May 2015
Accepted 31 May 2015
Available online 10 June 2015

Keywords:
Fresh produce
Chemical oxygen demand
Disinfection by-products

ABSTRACT

The use of chitosan for coagulation of artificial fresh-cut lettuce wash-water, made from butterhead lettuce and tap water, was assessed for reducing particulate and dissolved matter in order to improve the physicochemical quality, as such lowering the disinfectant demand and allowing longer reuse of post-harvest wash-water. Chitosan was used to coagulate industrial leafy-vegetable wash-water from a processing company. Lowering the pH improved the reduction efficiency. Although the turbidity reduction was very high (>90%), virtually no dissolved organic matter was removed. Rapid sand filtration subsequent to coagulation resulted in more than 2 log reduction of aerobic psychrotrophic counts and total coliforms. Coagulation lowered the disinfectant demand (of free chlorine and peracetic acid) to some degree but had no positive impact on the trihalomethanes formation. As such, coagulation does not seem to be a viable option to lower the disinfection by-products, the disinfectant dose or the water use by itself. However, due to the efficient particle reduction by chitosan coagulation it shows potential to be used as a pretreatment for a subsequent biological treatment in a more developed water treatment system to allow water reuse in the fresh produce companies.

1. Introduction

Cross-contamination via water during fresh produce washing at the processing plant plays a role in the spread of pathogens and spoilage microorganisms among produce (Holvoet, Jacxsens, Sampers, & Uyttendaele, 2012; Lopez-Galvez et al., 2012). In order to avoid cross-contamination, water disinfection is often applied in the washing bath. During the industrial washing of fresh produce, (in)organic particulate and dissolved matter is transferred to the wash-water. In the case of fresh-cut produce, exudates, leaking from cut surfaces, increase the organic matter content of the wash-water. As the water is recirculated during these processes to conserve water, a build-up of (in)organic matter in the water occurs (Selma, Allende, Lopez-Galvez, Conesa, & Gil, 2008a, 2008b; Van Haute, Sampers, Holvoet, & Uyttendaele, 2013; Van Haute, Sampers, Jacxsens, & Uyttendaele, 2015). As such, a higher consumption of water disinfectant, and (if relevant) a higher production of disinfection by-products (DBPs) take place (Van Haute et al., 2015). Besides the use of disinfectants that do not produce harmful DBPs, strategies to lower the organic matter content could be applied to allow a longer reuse of water in this industry, as well as to lower the disinfectant consumption.

Coagulation-flocculation is widely applied in water and wastewater treatment to destabilize colloids and particles, resulting in particle aggregation, allowing reduction by gravity (sedimentation, flotation) and/or filtration. The most frequently used coagulants are mineral additives such as alum and polyaluminum chloride, and synthetic polymers based on polyacrylamide. However, aluminum coagulants are often overdosed to ensure coagulation efficiency, which results in water or sludge with high aluminum content that contributes to the accumulation of aluminum in the environment. Also, the spread of the neurotoxic and probable human carcinogen acrylamide mono- and oligomers might be hazardous (Ahmad, Sumathi, & Hameed, 2006; Chen & Chung, 2011; Renault, Sancey, Badot, & Crini, 2009; Vinci, Mestdagh, & De Meulenaer, 2012). There is increased interest in biopolymers with coagulating properties because they are biodegradable, usually nontoxic, and eliminate the problems related to dealing with potentially toxic sludge,

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http://dx.doi.org/10.1016/j.lwt.2015.05.063
0023-6438/

even allowing recycling of sludge from food processing waste water (Bina, Mehdinejad, Nikaeen, & Attar, 2009; Chi & Cheng, 2006; Mishra & Bajpai, 2005). In this study chitosan was used as coagulant. Chitosan has been applied for coagulation-flocculation treatment of various effluents, including wastewaters from several food processing operations (Renault et al., 2009; Shahidi, Arachchi, & Jeon, 1999). One of the main uses in food processing operations is based on the ability of chitosan to recuperate lipids and proteins from food wastewaters by flocculation (Ahmad et al., 2006; Boeris, Micheletto, Lionzo, da Silveira, & Pico, 2011; Chi & Cheng, 2006; Fernandez & Fox, 1997).

Implementation of coagulation-flocculation in produce washing processes could be a method to prolong the water use during operation. The efficiency of the method depends on the capability of the coagulation-flocculation process to remove sufficient amounts of organic matter from the wash-water, translating into a lower disinfectant demand, and lower DBPs (a. o. trihalomethanes (THMs)) production in the wash-water during chlorination, which were assessed in this study.

2. Materials & methods

2.1. Experimental setup

Experiments were performed in both standardized process water (SPW) and industrial process water (IPW) from a leafy-vegetables processing plant. Coagulation-flocculation-sedimentation experiments were performed with a jar test apparatus. Efficiency was assessed based on turbidity and chemical oxygen demand (COD) reduction. The reduction of aerobic psychrotrophic plate count (APC), total coliforms, and inoculated *Escherichia coli* (*E. coli*) O157 strains after coagulation-flocculation-sedimentation-rapid sand filtration was assessed in SPW.

2.2. Standardized process water

Butterhead lettuce (*Lactuca sativa* L.) was purchased from a local market in Belgium and transported at <4 °C to the lab for further handling. After discarding the outer leaves, 67 g of lettuce was put in a stomacher bag (0.5 mm pore size), 200 mL of tap water was added, and the mixture was homogenized for 2 min. The COD of this suspension was determined, and subsequently the suspension was diluted with tap water to obtain SPW containing about 800 mg O_2/L COD.

2.3. Wash-water from a fresh-cut leafy-vegetables processing company

IPW was collected during the leafy-vegetables washing operation. Water was collected in sterile recipients and transported to the lab at <4 °C, where it was stored at 4 °C till execution of the experiments. In the company, ground water was used as water source during washing of butterhead lettuce, iceberg lettuce, endive and radicchio (ca. 250 kg leafy-vegetables/h; 450 L washing bath volume). Water from the same processing line, processing the same leafy-vegetables, was sampled on two different days and process duration: after 1 h of vegetable processing (IPW Low), after 4 h of vegetable processing (IPW High).

2.4. Coagulants

Chitosan of lower molecular weight (LMW; ~0.2 Pa·s for 10 g chitosan/L acetic acid solution (10 mL/L acetic acid) at 25 °C), and higher molecular weight (HMW, ~0.8 Pa·s for 10 g chitosan/L acetic acid solution (10 mL/L acetic acid) at 25 °C) were purchased from Sigma-Aldrich (Diegem, Belgium). Chitosan solutions were made by dissolving 5 g chitosan/L hydrochloric acid solution (0.1 mol/L hydrochloric acid).

2.5. Determination of degree of deacetylation and molecular weight of chitosan

The degree of deacetylation (DD) was determined with the two abrupt change potential titration method as described by Wang et al. (2006). The apparent pKa values were estimated from the titration curves by locating the half-equivalence point. The molecular weight was determined with an Ubbelohde viscosimeter (PM Tamson Instruments, Bleiswijk, The Netherlands) as described by Wang et al. (2006).

2.6. Coagulation-flocculation-sedimentation experiments

Different concentrations of chitosan were added to the SPW at pH 5 and unchanged pH (approximately 7.5). To change the pH of the water, hydrochloric acid (1 mol/L) was added. Chitosan was also tested at pH 9, to assess the reduction efficiency in the absence of any protonated amine-groups in the chitosan molecule. The pH was controlled at pH 9 by adding NaOH (1 mol/L) to compensate for the pH drop caused by the addition of acidified chitosan. In the case of pH 5, the pH drop was insignificant (up till 35 mg/L chitosan) and in the case of unaltered pH (7.5) the alkalinity of the tap water functioned as a buffer which resulted in non-significant pH drops (up till 50 mg/L).

Jars were filled with 1 L SPW and the water temperature controlled at 7–8 °C to simulate the water temperature at the processing plant. Coagulants were added during stirring at 200 rpm with a conventional jar test apparatus (RER, IKA Labortechnik, Staufen, Germany). The SPW was stirred for 2 min at 200 rpm and subsequently for 28 min at 30 rpm, after which the formed flocs were allowed to settle for 30 min. The upper 400 mL were withdrawn from the jar using a pipette and analyzed for turbidity and/or COD. The same set of experiments that were executed in SPW, were repeated in IPW.

The optimal dose of coagulant, i.e. the lowest dose that provides the greatest turbidity reduction after sedimentation and obtained in the screening process, was used to repeat the experiment in order to assess the microbial reduction but this time with subsequent filtration over 70 cm of filter sand (effective size = 0.52 mm; uniformity coefficient = 1.5) in a borosilicate glass column (2.5 cm internal diameter) and at a filtration speed of 10 m/h, using a peristaltic pump (Sci-Q 323, Watson-Marlow, Gent, Belgium). For this purpose, phosphate buffer (containing *E. coli* O157 cells) was added to the SPW before execution of the jar test experiment under mixing of the SPW to obtain ca. 6 log CFU/mL. Experiments on *E. coli* O157 were executed separately from those on the naturally present microorganisms (total coliforms and APC), to make sure *E. coli* O157 did not contribute to the APC counts. *E. coli* O157, total coliforms and APC in the SPW were enumerated at the start, after sedimentation and after filtration.

2.7. Assessing the impact of coagulation on disinfectant decay and THMs production

IPW High (both coagulated and untreated) was exposed to 100 mg/L free chlorine (28.4 g/L NaOCl, La Croix, Brussel, Belgium) or 5 mg/L peracetic acid (Chriox 5, Christeyns Chemicals, Gent, Belgium) at 4 °C and, while being mixed continuously, periodically sampled to measure the disinfectant concentration. IPW High (both coagulated and untreated) was exposed to a dose of 500 mg/L free chlorine and stored, while continuously being mixed, at 4 °C. This