Use of ozone in the food industry

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Received 28 October 2002; accepted 8 October 2003

Abstract

Ozone is a strong oxidant and potent disinfecting agent. Even though it is new for the US, it has been utilized in European countries for a long time. Ultraviolet radiation (188 nm wavelength) and corona discharge methods can be used to generate ozone. The bactericidal effects of ozone have been documented on a wide variety of organisms, including Gram positive and Gram negative bacteria as well as spores and vegetative cells. In this review, chemical and physical properties of ozone, its generation, and antimicrobial power of ozone with two suggested mechanisms were explained as well as many advantages of ozone use in the food industry. There are numerous application areas of ozone in the industry such as food surface hygiene, sanitation of food plant equipment, reuse of waste water, treatment and lowering biological oxygen demand (BOD) and chemical oxygen demand (COD) of food plant waste. Treating fruits and vegetables with ozone has been found to increase shelf-life of the products. Notably, when ozone is applied to food, it leaves no residues since it decomposes quickly. In this review, use of ozone in food industry was discussed.

Keywords: Ozone; Food; Sanitation; Antimicrobial; Food waste; Disinfectant

1. Introduction

Ozonation has been used for years to disinfect water for drinking purposes in Europe. A number of other commercial uses have been found for ozone including disinfection of bottled water, swimming pools, prevention of fouling of cooling towers, and wastewater treatment (Rice, Robson, Miller, & Hill, 1981; Legeron, 1982; Schneider, 1982; Echols & Mayne, 1990; Costerton, 1994; Videla, Viera, & Guiamet, 1995; Strittmatter, Yang, & Johnson, 1996). In the US ozone application in the food industry has not been widely used; however, the United States Food and Drug Administration granted generally recognized as safe (GRAS) status for use of ozone in bottled water in 1982. Ozone use was approved by the US Department of Agriculture for reconditioning recycled poultry chilling water in 1997. After a year of reviewing the worldwide database on ozone, an expert panel in 1997 decreed that ozone was a GRAS substance for use as a disinfectant or sanitizer for foods when used in accordance with good manufacturing practices. Since the US Food and Drug Administration did not object to the expert panel’s findings, ozone has now been approved for use as a disinfectant or sanitizer in foods and food processing in the United States (USDA, 1997).

Ozone is a powerful antimicrobial substance due to the its potential oxidizing capacity. Ozone use may have many advantages in the food industry. There are suggested applications of ozone in the food industry such as food surface hygiene, sanitation of food plant equipment, reuse of waste water, lowering biological oxygen demand (BOD) and chemical oxygen demand (COD) of food plant waste (Rice, Farguhar, & Bollyky, 1982; Guzel-Seydim, 1996; Majchrowicz, 1998; Dosti, 1998). Multifunctionality of ozone application makes ozone a promising agent. Although ozone has not been commonly used in the dairy and food industry, it has found limited applications in a few areas such as conversion of green tea to black tea (Graham, Struder, & Gurkin, 1969), cleansing of shellfish (Anonymous, 1972), and disinfection of poultry carcasses and chill water in the poultry industry (Yang & Chen, 1979; Sheldon & Brown, 1986; Chang & Sheldon, 1989). In this review, mainly its chemical properties, generation,
antimicrobial properties, application on food surfaces, application on food plant equipment as an alternative sanitizer.

2. Chemical and physical properties of ozone

Ozone was first discovered by the European researcher C.F. Schönbein in 1839. It was first used commercially in 1907 in municipal water supply treatment in Nice and in 1910 in St. Petersburg (Kogelschatz, 1988). Major physical properties of pure ozone were given in Table 1. Ozone is the second most powerful common oxidizing agent (Table 2).

Ozone is formed in the stratosphere, in photochemical smog and by UV sterilization lamps, high voltage electric arcs, and gamma radiation plants (Mustafa, 1990). At room temperature, ozone decomposes rapidly and, thus, does not accumulate substantially without continual ozone generation (Peleg, 1976; Miller et al., 1978). At room temperature, ozone is a nearly colorless gas. Ozone has a pungent, characteristic odor described as similar to “fresh air after a thunderstorm” (Coke, 1993). It is readily detectable at 0.01–0.05 ppm level (Miller et al., 1978; Mustafa, 1990; Mehlman & Borek, 1987). It is found in low concentration in nature. Ozone has a longer half-life in the gaseous state than in aqueous solution (Rice, 1986). Ozone in pure water rather quickly degrades to oxygen, and even more rapidly in impure solutions (Hill and Rice, 1982). Ozone solubility in water is 13 times that of oxygen at 0–30°C and it is progressively more soluble in colder water (Rice, 1986) (Table 3). Ozone decomposition is faster at higher water temperatures (Rice et al., 1981).

Ozone is a blue gas at ordinary temperature, but at concentrations which it is normally produced the color is not noticeable. At −112°C, ozone condenses to a dark blue liquid. Liquid ozone is easily exploded if greater than 20% ozone to oxygen mixtures occur. Explosions may be detonated by electrical sparks or by sudden changes in temperature or pressure. However, in practical usage explosions of ozone are extremely rare. The three atoms of oxygen in the ozone molecule are arranged at an obtuse angle whereby a central oxygen atom is attached to two equidistant oxygen atoms; the included angle is approximately 116°49′ and the bond length is 1.278 Å. Four structures of ozone are shown in Fig. 1 (Oehlschlaeger, 1978).

Although in low concentrations ozone is not an extremely toxic gas, at high concentration ozone may be fatal to humans. After 1–2 h exposure to ozone (0.65 ppm) dogs exhibited rapid breathing whereas long-term (4–6 weeks) ozone exposure (0.2 ppm) to young rats exhibited lung distension (Barlett, Faulkner, & Cook, 1974). It was found that 0.2 ppm and higher concentrations of ozone can cause varying degrees of damage to the respiratory tract, depending on exposure length (Schwartz, Dungworth, Tarkington, & Tyler, 1976). Damage of pulmonary system involves the trachea (Schwartz, et al., 1976), bronchi (Castleman, Dungworth, & Tyler, 1973), and alveoli (Schwartz et al., 1976).

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Major physical properties of pure ozone (Manley and Niegowski, 1967)</th>
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<tbody>
<tr>
<td>Boiling point</td>
<td>−111.9 ± 0.3°C</td>
</tr>
<tr>
<td>Melting point</td>
<td>−192.5 ± 0.4°C</td>
</tr>
<tr>
<td>Critical temperature</td>
<td>−12.1°C</td>
</tr>
<tr>
<td>Critical pressure</td>
<td>54.6 atm</td>
</tr>
</tbody>
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<tr>
<th>Table 2</th>
<th>Oxidizing agents and their oxidation potential (Manley and Niegowski, 1967)</th>
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</thead>
<tbody>
<tr>
<td>Oxidizing agent</td>
<td>Oxidation potential (mV)</td>
</tr>
<tr>
<td>Fluorine</td>
<td>3.06</td>
</tr>
<tr>
<td>Ozone</td>
<td>2.07</td>
</tr>
<tr>
<td>Permanganate</td>
<td>1.67</td>
</tr>
<tr>
<td>Chlorine dioxide</td>
<td>1.50</td>
</tr>
<tr>
<td>Hypochlorous acid</td>
<td>1.49</td>
</tr>
<tr>
<td>Chlorine gas</td>
<td>1.36</td>
</tr>
</tbody>
</table>

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<tr>
<th>Table 3</th>
<th>Temperature and solubility relationship of ozone in water (Rice et al., 1981)</th>
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<tbody>
<tr>
<td>Temperature (°C)</td>
<td>Solubility (liter ozone/liter water)</td>
</tr>
<tr>
<td>0</td>
<td>0.640</td>
</tr>
<tr>
<td>15</td>
<td>0.456</td>
</tr>
<tr>
<td>27</td>
<td>0.270</td>
</tr>
<tr>
<td>40</td>
<td>0.112</td>
</tr>
<tr>
<td>60</td>
<td>0.000</td>
</tr>
</tbody>
</table>

Fig. 1. Resonance structures of ozone molecule (Oehlschlaeger, 1978).
3. Ozone generation

Rice et al. (1981) explained the general ozone generation. In order to generate ozone, a diatomic oxygen molecule must first be split. The resulting free radical oxygen is thereby free to react with another diatomic oxygen to form the triatomic ozone molecule. However, in order to break the O–O bond a great deal of energy is required. Ultraviolet radiation (188 nm wavelength) and corona discharge methods can be used to initiate free radical oxygen formation and, thereby, generate ozone. In order to generate commercial levels of ozone, the corona discharge method is usually used (Fig. 2).

There are two electrodes in corona discharge, one of which is the high tension electrode and the other is the low tension electrode (ground electrode). Those are separated by a ceramic dielectric medium and narrow discharge gap is provided (Fig. 2). When the electrons have sufficient kinetic energy (around 6–7 ev) to dissociate the oxygen molecule, a certain fraction of these collisions occur and a molecule of ozone can be formed from each oxygen atom. If air is passed through the generator as a feed gas, 1–3% ozone can be produced, however, using pure oxygen allows yields to reach up to 6% ozone (Rice et al., 1981). Consequently, ozone concentration cannot be increased beyond the point that the rates of formation and destruction are equal (Manley & Niegowski, 1967). Ozone gas cannot be stored since ozone spontaneously degrades back to oxygen atoms (Kogelschatz, 1988; Wickramanayaka, 1991; Coke, 1993).

4. Antimicrobial effects of ozone

The bacteriocidal effects of ozone have been studied and documented on a wide variety of organisms, including Gram positive and Gram negative bacteria as well as spores and vegetative cells (Fetner & Ingols, 1956; Foegeding, 1985; Ishizaki, Shinriki, & Matsuyama, 1986; Restaino, Frampton, Hemphill, & Palnikar, 1995).

Restaino et al. (1995) also investigated the antimicrobial effects of ozonated water against food related microorganisms and determined that ozone effectively killed such gram positive bacteria as Listeria monocytogenes, Staphylococcus aureus, Bacillus cereus, Enterococcus faecalis, and such Gram negative bacteria as Pseudomonas aeruginosa, and Yersinia enterocolitica. Restaino et al. (1995) also determined that ozone destroyed the yeasts Candida albicans and Zygosaccharomyces bactilli and spores of Aspergillus niger. Ozone destruction of bacteria is accomplished by attack on the bacterial membrane glycoproteins and/or glycolipids. Khadre and Yousef (2001) compared the effects of ozone and hydrogen peroxide against foodborne Bacillus spp. spores. It has been found that ozone was more effective than hydrogen peroxide.

Guzel-Seydim, Bever and Greene (2004) worked on the efficacy of ozone to reduce bacterial populations in food components using sterile Class C buffer, whipping cream, and 1% solutions of locust bean gum, soluble starch, and sodium caseinate in sterile distilled, deionized water. These substrates were inoculated with spores of Bacillus stearothermophilus or vegetative cells of Escherichia coli or Staphylococcus aureus and, then ozonation was applied. Spore populations at 10 min were reduced by 4.93 log cycles in buffer, 4.56 in starch, 0.95 for locust bean gum, and 0.24 for caseinate (p < 0.05). There was not a significant reduction in the bacterial populations in the cream (p > 0.05). Statistically significant (p < 0.05) log cycle reductions in the E. coli populations at 10 min were observed in buffer (6.10), starch (6.11), locust bean gum (3.86), caseinate (3.76), and whipping cream (1.98). For the S. aureus, statistically significant (p < 0.05) log reduction were detected at 10 min in buffer (6.48), starch (6.47), locust bean gum (4.94), caseinate (1.47) and whipping cream (1.02). The locust bean gum provided an intermediate level of protection, while the caseinate and whipping cream provided the greatest levels of protection to the bacterial populations.

It is the potent oxidation capacity that makes ozone very effective in destroying microorganisms. Ozone has been demonstrated to destroy a wide range of viruses.

5. Suggested mechanisms of antimicrobial power

Ozone destroys microorganisms by the progressive oxidation of vital cellular components. The bacterial cell surface has been suggested as the primary target of ozonation. Two major mechanisms have been identified in ozone destruction of the target organisms (Victorin, 1992): first mechanism is that ozone oxidizes sulfhydryl groups and amino acids of enzymes, peptides and proteins to shorter peptides. The second mechanism is that ozone oxidizes polyunsaturated fatty acids to acid peroxides (Victorin, 1992). Ozone degradation of the cell envelope unsaturated lipids results in cell disruption and subsequent leakage of cellular contents. Double bonds of unsaturated lipids are particularly vulnerable to ozone attack. In Gram negative bacteria, the lipoprotein and lipopolysaccharide layers are the first sites of destruction resulting in increases in cell permeability and eventually cell lysis (Kim, Yousef, & Dave, 1999). Chlorine selectively destroys certain intracellular enzyme systems; ozone will cause widespread oxidation of internal cellular proteins causing rapid cell death (Mudd, Leavitt, Ongun, & Mcmanus, 1969; Hinze, Prakash, & Holzer, 1987; Takamoto, Maeba, & Kamimura, 1992; Kim et al., 1999). Cellular death can also occur due to the potent destruction and damage of nucleic acids. Thymine is more sensitive to ozone than cytosine or uracil. Ozone also destroys viral RNA and alters polypeptide chains in viral protein coats (Kim et al., 1999).

6. Use of ozone for food hygiene

Ozone has been used for disinfecting recycled poultry chill water and disinfection of poultry carcasses (Sheldon & Brown, 1986). According to the Code of Federal Regulations (USDA, 1997), there must be at least 60% reduction in total microorganisms and similar reduction in coliforms, E. coli, and Salmonella spp. Waldroup, Hierholzer, Forsythe, and Miller (1993) investigated the use of ozone for recycling of poultry chill water and determined that there were no viable E. coli or presumptive coliforms after ozonation. In addition, the total aerobic plate count was low and use of ozone for disinfecting poultry chill water met USDA recycling requirements. Sheldon and Brown (1986) applied ozone directly to poultry carcasses. Ozone destroyed more than 2 log-units of all carcass microorganisms with no significant lipid oxidation, off-flavor development or loss in carcass skin color. They concluded that using ozone on recycling chilled water partially fulfilled USDA requirements.

Treating fruits and vegetables with ozone has been used to increase shelf-life (Norton, Charig, & Demorandville, 1968; Rice et al., 1982). Treatment of apples with ozone resulted in lower weight loss and spoilage. An increase in the shelf-life of apples and oranges by ozone has been attributed to the oxidation of ethylene. Fungal deterioration of blackberries and grapes was decreased by ozonation of the fruits (Beuchat, 1992). Onions have been treated with ozone during storage. Mold and bacterial counts were greatly decreased without any change in chemical composition and sensory quality (Song, Fan, Hildebrand, & Forney, 2000). Shredded lettuce in water bubbled with ozone gas had decreased bacterial content (Kim et al., 1999). Ozone has been used experimentally as a substitute for ethylene oxide for the decontamination of whole black peppercorns and ground black pepper (Zhao & Cranston, 1995). Ozone treatment of ground black pepper resulted in slight oxidation of volatile oil constituents but ozone had no significant effect on the volatile oils of whole peppercorns. Caryophyllene was the most abundant component in the pepper followed by δ-2-carene, limonene, β-pinene, α-pinene, and α-phellandrene. No new components were detected as a result of ozonation. Because ozonation successfully reduced microbial loads and did not cause significant oxidation of the volatile oils in whole black peppercorns, this method was recommended for industrial treatment of the spice (Zhao & Cranston, 1995). A number of patents have been issued for using ozone to treat fruits and vegetables. Ozone has been used experimentally to control mold on Cheddar cheese surfaces and in cheese rooms. At high ozone concentrations ozone appeared to destroy the molds present. However, upon termination of ozonation, mold populations flourished (Gibson, Elliot, & Beckett, 1960).

7. Use of ozonated water for plant equipment sanitation

In the food industry, after proper cleaning many different types of sanitizing agents are used such as derivatives of chlorine, acid, iodine, and quaternary ammonium compounds (Marriott, 1994). Some food
industries use thermal sanitation methods and/or irradiation. Thermal sanitation is very effective in destroying contaminating microorganisms; however, steam and hot water are quite expensive to generate and excessive heat can be damaging to food processing equipment (Troller, 1993). Radiation methods are not practical for use in food processing plants because of the inherent hazards associated with radioactive materials. Chemical sanitation methods are the most commonly utilized sanitizers in the food industry. Chlorinated agents are used worldwide for disinfecting water, wastewater and for sanitizing food processing plant equipment. Even though chlorine sanitizers have several disadvantages including being harmful and irritating in high concentrations, being prone to forming carcinogenic compounds and being toxic to the environment, these compounds are economical bactericides that inactivate all types of vegetative cells. Chlorine has been a preferred disinfectant in the food and water industries for many years. Although chlorination efficiently decreases the spread of food borne infectious disease, chlorine combines with many organic compounds to form toxic by-products. These by-products are released in drinking water and adversely affect public health as well as the environment (Bellar, Lichtenberg, & Kroner, 1974; Trussell and Umphres, 1978). Among these by-products, trihalomethanes (THM) and haloacetic acids (HAA) are mutagenic and carcinogenic. Trussell and Umphres (1978) have stated that levels of halofroms (such as chloroform), THM and HAA increased markedly after chlorination of organic-laden water. The effect of chlorinated compounds on the environment is of major concern. Aquatic humus compounds are common natural substances found in rivers, streams and lakes. Metahydroxy aromatic ring, the functional group in humus structures, reacts with chlorine and forms hazardous trihalomethane compounds (Trussell and Umphres, 1978). Releases of chlorinated compounds into the environment can result in formation of carcinogenic THM in rivers, streams and lakes. Potential drinking water can be affected along with native aquatic and terrestrial species. Consequently, efforts are being made to minimize releases of chlorine into the environment. Although the hazards of carcinogen/mutagen formation from chlorine are extensive, lack of suitable sanitizer/disinfectant would result in much greater illness and loss of life due to microbial contamination of foods and water. Therefore, chlorine use continues, even though it is known to be hazardous. It is imperative that effective, yet safe sanitizers are needed for use in food and water processing (Bellar et al., 1974; Trussell & Umphres, 1978; Krasner et al., 1989; Minear & Amy, 1995).

Food researchers are searching for alternative cleaning and sanitizing agents effective against food spoilage and pathogenic bacteria, yet harmless to humans and the environment. Additionally, these agents must also be non-corrosive to expensive food processing equipment. Ozone is a potential alternative to chlorine for use in the food industry.

Greene, Few, and Serafini (1993) proposed the use of ozonated water as a sanitizer for dairy and food plants. They compared the effectiveness of ozonated water and chlorinated sanitizer for the disinfection of stainless steel surfaces which had been incubated with UHT milk inoculated with either Pseudomonas fluorescens (ATCC 949) and Alcaligenes faecalis (ATCC 337) at 32°C for 4–24h. Stainless steel plates were used to simulate dairy plant equipment. It has been found that ozone was as effective as chlorination against dairy surface attached bacteria, both treatments reduced bacterial populations by 99%. Ozone, chlorine and heat were compared for killing effectiveness against food spoilage bacteria in synthetic broth (Dosti, 1998). Fresh 24h bacterial cultures of P. fluorescens (ATCC 948), Pseudomonas fragi (ATCC 4973), Pseudomonas putida (ATCC 795), Enterobacter aerogenes (ATCC 35028), Enterobacter cloacae (ATCC 35030), and Bacillus licheniformis (ATCC 14580) were exposed to ozone (0.6 ppm for 1 and 10min), chlorine (100 ppm for 2 min) or heat (77 ± 1°C for 5 min). One min ozonation had little effect against the bacteria. There were significant differences (p<0.05) among 10 min ozonation, chlorine or heat inactivation of all bacteria except Bacillus licheniformis. Ten min ozonation caused the highest bacterial population reduction with a mean reduction over all species of 7.3 log units followed by heat (5.4 log reduction) and chlorine (3.07 log reduction).

If the cleaning process is ineffective, microorganisms may form biofilms on equipment surfaces. Microorganisms accumulate and proliferate in the porous biofilm material. Once embedded in this matrix, the microorganisms often become resistant to the action of sanitizers. Dosti, (1998) tested ozone against bacteria which are able to form biofilm, P. fluorescens (ATCC 948), P. fragi (ATCC 4973) and P. putida (ATCC 795) for 24–72 h. After biofilm formation, the SS metal coupons were rinsed with phosphate buffered saline (1 min) and exposed to ozone (0.6 ppm for 10 min) and chlorine (100 ppm for 2 min). Results indicated that both ozone and chlorine significantly reduced the biofilm bacteria adhered to the SS metal coupons as compared to the control (p<0.05). However, there was no significant difference (P>0.05) between ozone and chlorine inactivation of the bacteria with the exception of P. putida. Ozone killed P. putida more effectively than chlorine (Dosti, 1998).

Guzel-Seydim, Wyffels, Greene, and Bodine (2000) studied the use of ozonated water in dairy equipment. Soiled stainless steel coupons were treated with ozonated water as a pre-rinse. Results implied that ozone treatment removed 84% of dairy soil when compared to
warm water (40°C) treated samples removed only 51%. Scanning electron microscopy pictures were consistent with the results. From the results of this experiment, it is hypothesized that ozone use in the pre-rinse stage may allow for decreased detergent use in the cleaning solution recirculation step. Greene, Vergano, Few, and Serafini (1994) studied the effect of ozone and chlorine on seven gasket materials which are used in fluid food processing. Buna N, white Buna N, ethylene propylene diene monomer (EPDM), polyethylene, silicone rubber, polytetrafluoroethylene or Teflon (PTFE), and stream resistant Viton. Gaskets of each material were treated with either chlorine or ozonated water (0.5 ppm ozone) for 36 h at 25–33°C. Both treatments did not significantly affect the tensile strength. The elasticity of ozone treated PTFE gaskets was significantly affected by ozone treatment. Both treatments had a bleaching effect on black gaskets such as Buna N, EPDM, and Viton. The tensile strength of EPDM and Viton was decreased by ozone treatment but not significantly when compared to chlorine treatment (Greene et al., 1994).

“Criteria for the bacteriological quality of recirculating chilling water for dairy operations have been established in Appendix G of the United States Grade “A” Pasteurized Milk Ordinance.” (Anonymous, 1995) Ozone was suggested for using in chilled water to reduce bacterial content. Greene, Smith, and Knight (1999) studied the effect of ozone on metals in dairy chilling water systems. Aluminum, copper, carbon steel, 304 stainless steel and 316 stainless steel were examined to record durability of metals against ozone. Metals have been analyzed according to weight loss of metals and by using scanning electron microscopy. Carbon steel has the significantly different weight loss than the control. There was no significant difference among other metals within control and ozone treatments. SEM pictures showed that there was severe pitting on ozone treated copper samples (Greene et al., 1999).

Since food plant waste often contains large amounts of carbohydrates, fats, proteins and mineral salts, complete degradation of this effluent is complex. Dairy, meat, poultry, and seafood processing plant effluent may cause significant water pollution due to heavily loaded waste. These wastes are likely treated by biological methods. The primary concern with these food processing wastes is that organic matter provides a food source for microbial growth. Microorganisms can therefore proliferate rapidly and subsequently cause a decreasing amount of dissolved oxygen in the water. Biochemical oxygen demand (BOD) and chemical oxygen demand (COD) values of wastes are very important. Worldwide, hazardous cleaner and sanitizer compounds are also routinely dumped into sewage systems. Eventually, these chemicals enter the environment with deleterious effects on delicate ecosystems. These compounds complicate waste degradation and also maybe toxic to the environment. Artificial dairy waste samples were treated with ozone; ozonation treatment lowered the biochemical oxygen demand of dairy waste by 15% (Guzel-Seydim, 1996). Ozone pre-oxidizes organic material so that it is easier for biodegradation. With lowered treatment needs, required sewage treatment capacities and levied surcharges could possibly be reduced. Ozone is a promising cleaning and waste treatment agent for the dairy and food industry.

8. Ozone toxicity

Ozone toxicity is the most important criterion for approval of ozone in food and dairy processing plants. It is important to monitor people who might have contact with ozone in industry. In humans, ozone primarily affects the respiratory tract. Symptoms of ozone toxicity include headache, dizziness, a burning sensation in the eyes and throat, a sharp taste and smell, and cough. Chronic toxicity symptoms might cause headache, weakness, decreased memory, increased prevalence of bronchitis and increased muscular excitability (Hoof, 1982).

9. Conclusions

Food industry is seeking for more effective applications to ensure the safer food products. System design will be the most important issue for the food plant. There are sound advantages of ozone applications in food industry such as food surface hygiene, sanitation of food plant equipment, reuse of waste water, treatment and lowering BOD and COD of food plant waste. The bactericidal effects of ozone have been documented on a wide variety of organisms, including Gram positive and Gram negative bacteria as well as spores and vegetative cells. Even though it does not leave any residue due to quick decomposition of its structure, restrictions should be applied to human exposure to ozone.

References


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