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Ozonation and UV irradiation—an introduction and examples of current applications

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Abstract

This paper was written to introduce the 2001 AES Issues Forum's '*Ozone and UV Treatment*' session by providing an overview of ozone and ultraviolet (UV) irradiation technologies as well as several examples of current ozone and UV irradiation applications in aquaculture.
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1. Introduction

Ozone is a powerful oxidizing agent that has seen wide use in aquaculture applications for achieving both disinfection and water quality improvements (Rosenthal, 1981; Owsley, 1991; Cryer, 1992; Wedemeyer, 1996; Summerfelt and Hochheimer, 1997). Ozone is added to aquaculture system waters to inactive fish pathogens, oxidize organic wastes (including color) and nitrite, or supplement the effectiveness of other water treatment units. Ozone has some advantages because it has a rapid reaction rate, produces few harmful reaction by-products in freshwater, and oxygen is produced as a reaction end-product.

Ultraviolet (UV) irradiation is also being widely applied within aquaculture systems. However, the primary objective of UV irradiation is disinfection. In

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contrast to ozonation, UV irradiation is not generally considered to be a process that is applied to supplement the effectiveness of other water treatment units.

2. Ozonation

Ozone application within aquaculture systems requires ozone generation, ozone transfer into solution, contact time for ozone to react, and possibly ozone destruction to ensure that no ozone residual makes it into the culture tanks (Summerfelt and Hochheimer, 1997). These requirements are discussed, along with certain key issue regarding the application of ozone within recirculating aquaculture systems.

2.1. Ozone generation

Ozone is typically generated within an enriched oxygen feed gas using an electrical corona discharge. Enriched oxygen feed gases are often used because ozone production is 2–3 times more energy efficient when an oxygen feed gas is used instead of air (Masschelein, 1998), and because purified oxygen feed gas supplies are already used to maximize carrying capacity within many intensive aquaculture systems. Corona discharge generation using purified oxygen feed gas requires about 10 kW h of electricity to produce 1.0 kg of ozone (Masschelein, 1998). Also, generating ozone in oxygen feed gas can produce a 10–15% (by weight) concentration of ozone, which nearly doubles the concentration of ozone that can be generated using air as the feed gas. The relatively high concentrations of ozone can be generated to reduce the overall mass of oxygen required to supply ozone. Yet, it is less energetically efficient to produce ozone concentrations of 10–15% (by weight) than to produce ozone concentrations of 4–6% (Carlins and Clark, 1982). Taking all of this into account, ozone production can be optimized according to the demands of the aquaculture system and economic considerations of feed gas cost and energy usage.

2.2. Ozone transfer

Ozone generated within either an air or oxygen feed gas must be transferred into water for microbiological inactivation or other oxidative purpose. The ozone gas can be transferred into the water using any of the typical oxygen transfer devices (Summerfelt and Hochheimer, 1997). Effective transfer of ozone into water is important because the cost of producing ozone is not insignificant, especially if the ozone is carried within a purified oxygen feed gas that is either purchased or produced on site.

The rate of ozone transfer and the subsequent rate of ozone decomposition depends upon the contact system efficiency and the reaction rates of ozone with constituents in the water. The ozone reaction rate depends on the water temperature and on the concentration and type of constituents contained in the water. Rapid

reaction with oxidizable inorganic and organic material will maintain a low apparent equilibrium concentration of ozone within the liquid film and increase the rate of ozone transfer compared to water's without oxidizable inorganic and organic material. The driving force for ozone transfer is maximized when the ozone absorbed is rapidly consumed by reaction with constituents within water. In fact, when ozone reacts very fast, ozone decomposes at the gas surface and no molecular ozone is transferred into the water (Bablon et al., 1991).

Ozone transfer units that have a continuous liquid-phase (i.e., units that disperse gas bubbles within a liquid)—such as Speece cones (Fig. 1), U-tubes (Fig. 2), aspirators, bubble diffusers, and enclosed mechanical surface or subsurface mixers—provide both ozone transfer and some reaction time. Ozone transfer units that have a continuous gas-phase (i.e., units that disperse liquid drops and films within a gas)—
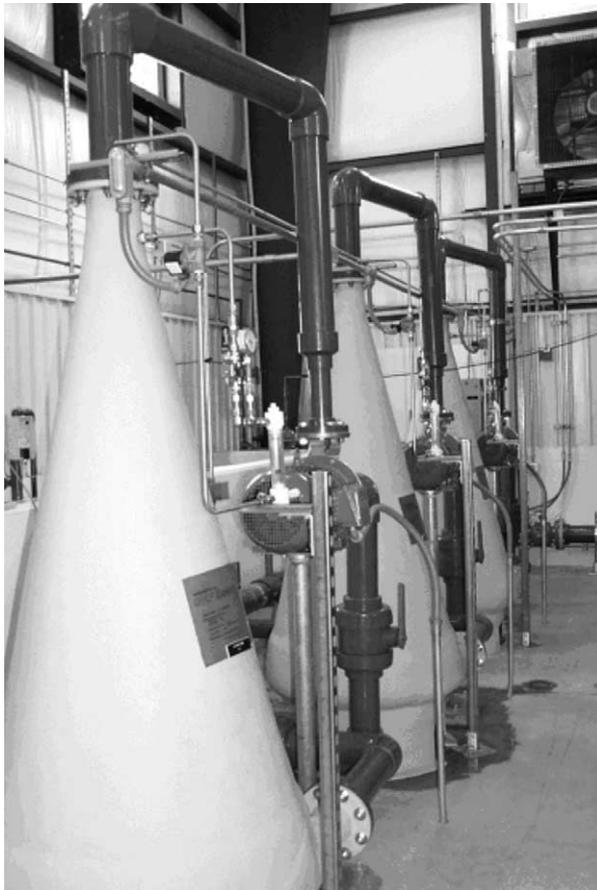

Fig. 1. An ozone/oxygen feed gas is injected into water within three Speece cones (plumbed in parallel for redundancy and variable flow requirements). The system shown is used to disinfect 400–2400 l/min of surface water at the US Fish and Wildlife Service's Northeast Fishery Center in Lamar, PA (Summerfelt et al., in press).
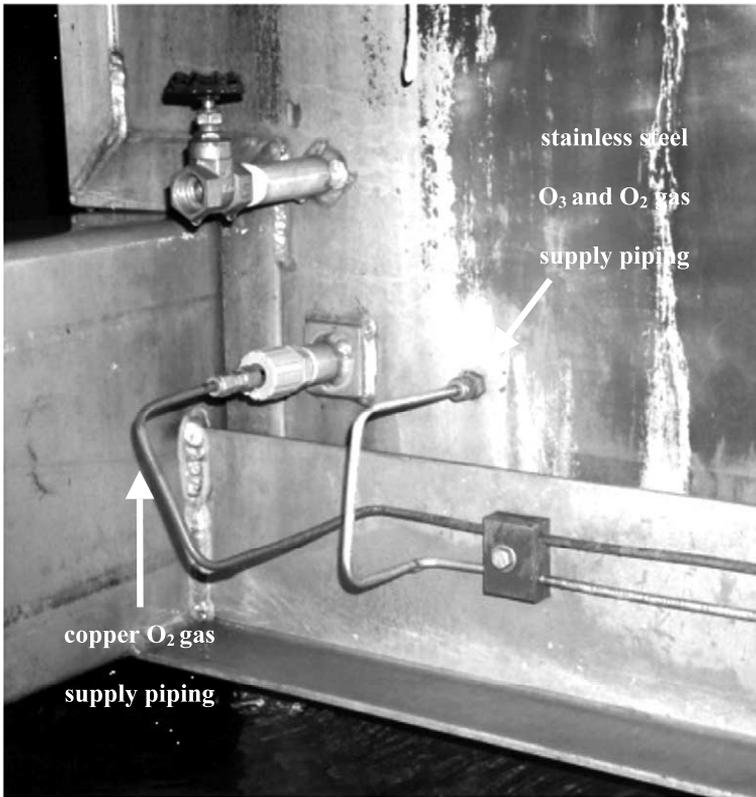
Fig. 2. A copper pipe is used to supply the pure oxygen feed gas to a LHO unit, while a stainless steel pipe is used to carry the ozonated feed gas. The LHO is used to oxygenate/ozonate 4800 l/min of water in a recirculating system at the Conservation Fund Freshwater Institute (Shepherdstown, WV).

such as spray columns, packed columns, and multi-stage low head oxygenators (Fig. 3)—provide efficient transfer but very little time for reaction (Summerfelt and Hochheimer, 1997). Continuous gas-phase transfer units are best suited for use in situations where the maximum amount of ozone needs to be transferred in the shortest time. Continuous liquid-phase transfer units are usually selected for situations where reaction is rate limiting and an ozone residual must be maintained for a specific length of time (Bellamy et al., 1991).

Most ozone contactors rely on continuous liquid-phase units that bubble ozone into the liquid (Bellamy et al., 1991). High column bubble diffusers are frequently used for aquacultural applications and can achieve more than 85% ozone transfer to the liquid phase (Liltved, 2001). These units are particularly well suited to situations where reaction is rate limiting and an ozone residual must be maintained for a specific length of time, such as during disinfection. Speece cones (Fig. 1), U-tubes, and low head oxygenators (Fig. 2) are also being used to efficiently and rapidly
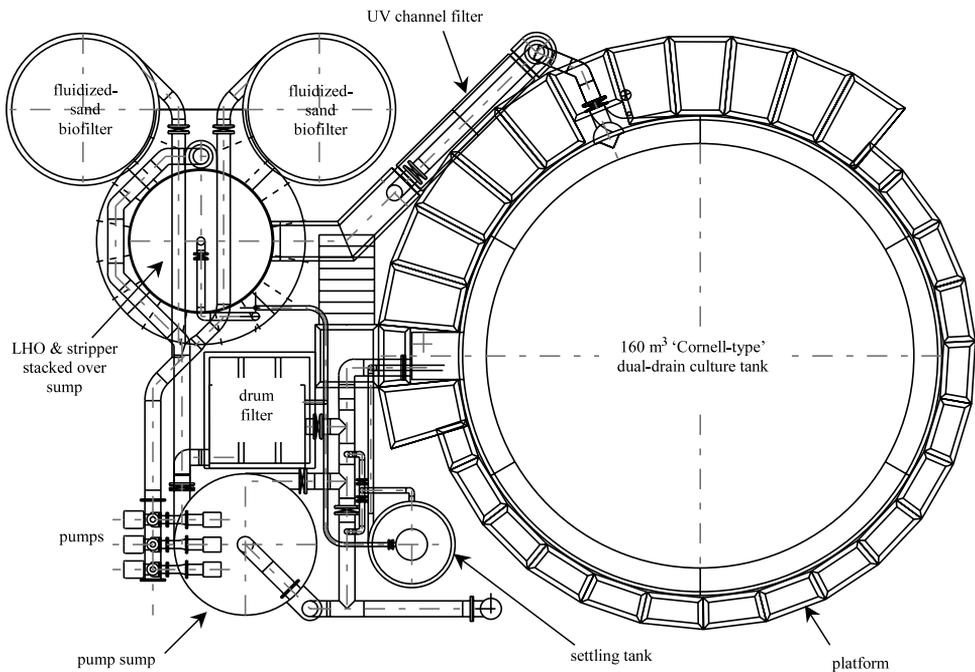
Fig. 3. The 4800 l/min recirculating system at the Freshwater Institute was designed for ozone addition within the purified oxygen feed gas supplied to the LHO unit. Drawing courtesy of Marine Biotech, Inc. (Beverly, MA).

transfer ozone/oxygen feed gas within recirculating aquaculture systems (Summerfelt et al., 2000), where the primary goals of the gas transfer units are to:

- supply supersaturated levels of dissolved oxygen that will increase the culture tank carrying capacity, and
- transfer ozone (carried within the purified oxygen feed gas) to oxidize nitrite and organic matter and supplement the effectiveness of other treatment processes.

Ozone transfer within continuous gas-phase units is not as common as within continuous liquid-phase units (Bellamy et al., 1991). When ozone transfer has been reported within continuous gas-phase units, the applications are mostly within packed columns and more recently within low head oxygenators that are used in recirculating system applications (Fig. 3). However, the relatively high transfer efficiency and relatively small vessel requirement of continuous gas-phase transfer units do make these units attractive when compared to the transfer efficiency and foot print of high column bubble diffuser systems.

If ozone transfer is not 100% efficient, then the off-gas discharged from the transfer unit will contain some ozone. Because ozone is toxic, the ozone in these off-gas flows must be treated to destroy the remaining ozone before the gas is discharged.

2.3. Ozone disinfection and maintaining an ozone residual

Ozone oxidation can kill microorganisms, but disinfecting the water requires maintaining a certain dissolved ozone concentration for a given contact time. Thus, disinfecting efficiency depends on the product of the ozone residual concentration multiplied by its contact time. An ozone contact vessel should provide the time necessary for the ozone residual to react with and inactivate the target microorganism(s). Disinfecting water can require maintaining a residual ozone concentration of 0.1–2.0 mg/l in a plug-flow type contact vessel for periods of 1–30 min, depending upon the target microorganism. Wedemeyer (1996) and Liltved (2001), and Summerfelt et al. (in press) provide reviews on ozone dosing requirements for various fish pathogens. These reviews indicate that many pathogenic organism can be inactivated by ozone $c \times t$ dosages of 0.5–5.0 min mg/l. Unfortunately, certain spore forming organism are especially hard to inactivate with ozone.

Ozone has seen frequent use for pre-treating surface waters supplied to fish farms (Liltved, 2001) and state or federal fish hatcheries (Roselund, 1975; Owsley, 1991; Cryer, 1992; Summerfelt et al., in press) in situations where water born pathogens are a significant concern or problem. On occasion, ozone has also been used to disinfect fish hatcheries discharges in an attempt to prevent the potential for the release of fish pathogens to the receiving watershed (Liltved, 2001).

In order to achieve the desired disinfecting $c \times t$ (i.e., the product of the ozone residual concentration at the end of the contact vessel multiplied by the hydraulic retention time of the contact tank), an ozone dose sufficiently high to account for the initial ozone demand of the water must be provided. In natural waters and in waters found within recirculating systems, additional ozone will be lost in reactions with organics and other compounds at rates that depend upon the water temperature. According to ozone demand tests on a high quality trout stream water being ozone disinfected at the US Fish and Wildlife Service Northeast Fishery Center (Lamar, PA), an ozone concentration of 2–4 mg/l must be transferred to maintain a 0.2 mg/l ozone residual concentration after 10 min (Summerfelt et al., in press). Cryer (1992) reported similar ozone demand results in tests on surface water supplies that were being disinfected at US Fish and Wildlife Service salmonid hatcheries in North America. All of the surface water supplies examined in these studies exhibit relatively high water quality with low concentrations of oxidizable organic material, iron, and manganese (Cryer, 1992; Summerfelt et al., in press), yet the ambient ozone demand reduces the half-life of ozone to less than a few minutes. In comparison, the half-life of ozone dissolved in pure water at 20 °C is 165 min (Rice et al., 1981). The ozone demand of water within recirculating aquaculture systems, which contains much higher levels of organic material and nitrite, creates an even shorter ozone half-life (e.g., < 15 s), which makes maintaining an ozone residual difficult (Bullock et al., 1997). For this reason, achieving large microorganism reductions in recirculating systems requires much greater ozone dosages than are required for simply controlling water quality within these systems (Bullock et al., 1997) and also much higher ozone dosages than are typically required for disinfecting single-pass inflows.

When sufficient ozone has been transferred to create a disinfecting ozone residual concentration at the end of the contact chamber, then that residual will need to be removed before the water reaches aquatic organisms in the culture tanks. Residual ozone can be lethal to fish at concentrations as low as 0.01 mg/l, but the actual concentration depends upon species and life stage (Section 2.4). To abate this potential problem, dissolved ozone can be removed by providing extended contact times, aeration and degassing, intense UV light doses, or reaction with hydrogen peroxide (Section 2.5).

The surface water filtration and ozonation system at the US Fish and Wildlife Service's Northeast Fishery Center (Lamar) is an example of the ozone contacting and removal sequence that can be employed to provide contact time for disinfection and also protect the fish culture system from ozone residual (Summerfelt et al., in press). This system (Fig. 4) first uses a pair of 60-μm microscreen drum filters to remove fine particulates that might shield pathogens from dissolved ozone. The system then uses Speece cones (Fig. 1) to transfer ozone into the water and subsequently provides contact time for the ozone within a two-reactor sequence that is followed by a ventilated cascade column (Fig. 4). The first vessel in the sequence provides the ozone contact time (e.g., 10 min HRT) required to achieve disinfection and the second larger vessel requires the contact time (e.g., 20 min HRT) for dissipation of much of the ozone residual. A dissolved ozone probe is used to monitor the ozone concentration exiting the first contact vessel and this information is used in a PID control loop to adjust the ozone generator output for maintaining a constant 0.2 mg/l of ozone residual concentration following the first ozone contact
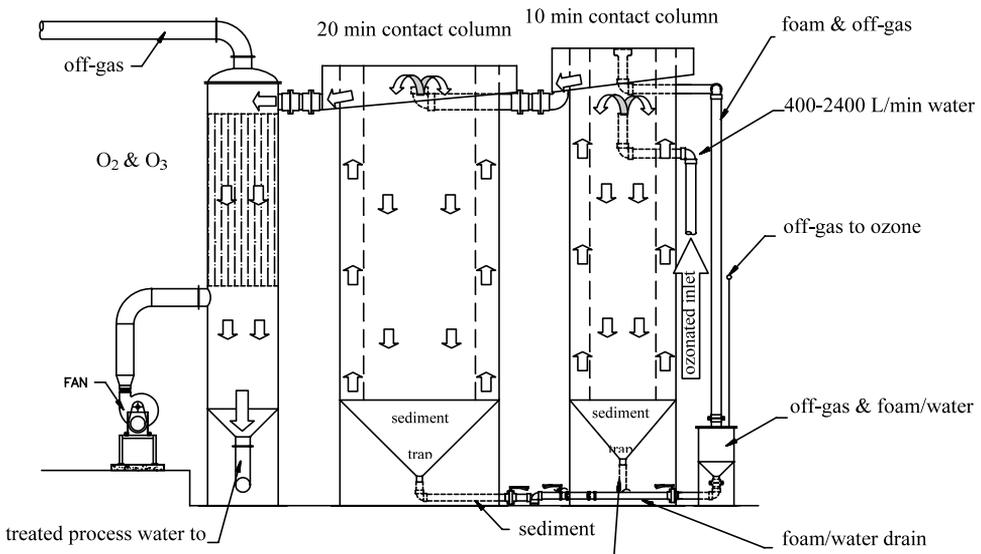

Fig. 4. Ozone Treatment system for disinfecting 400–2400 l/min of surface water at the US Fish and Wildlife Service's Northeast Fishery Center in Lamar, PA (Summerfelt et al., in press). Drawing courtesy of Oak Point Associates, Biddeford, ME.

tank. The ventilated cascade column (Fig. 4) is used to strip the remaining ozone residual while reducing excessive levels of dissolved oxygen before the water flow is supplied to the fish culture tanks.

2.4. Ozone toxicity

Although ozone has a rapid reaction rate and few harmful reaction products, it is toxic to aquatic life at low levels (Wedemeyer et al., 1979; Langlais et al., 1991). Ozone gas is also toxic to humans. Standards set by the federal Occupational Safety and Health Administration only allows for a maximum single exposure level of 0.3 ppm for less than a 10 min duration and of 0.1 ppm on a time-weighted average for an 8-h period (Occupational Health and Safety Administration, 1993). Therefore, care must be used when transferring an ozone containing air or purified oxygen gas mixture into water, when providing time for reaction of dissolved ozone with the targeted constituents in the water, and when considering the removal or monitoring of dissolved ozone before the water enters the culture tanks (Summerfelt and Hochheimer, 1997).

2.4.1. Freshwater applications

The maximum safe level of chronic ozone exposure for salmonids is 0.002 mg/l (Wedemeyer et al., 1979). A compilation of results from several studies indicates that most fish exposed to ozone concentrations greater than 0.008–0.06 mg/l will develop severe gill damage that can result in serum osmolality imbalances and can kill fish immediately or leave them more susceptible to microbial infections (Bullock et al., 1997).

Only limited and expensive technology exists to continuously monitor dissolved ozone at concentrations low enough to be safe for fish. Oxidation–reduction potential probes have also been used, with varying degrees of success, as an indirect means to monitor and control dissolved ozone levels (Bullock et al., 1997).

2.4.2. Seawater applications

Ozone reacts with bromide ions in brackish and seawater systems to form the oxidants hypobromous acid (HOBr) and hypobromite ion (OBr$^-$), which are relatively stable and toxic to fish and shellfish (Crecelius, 1978; Huguenin and Colt, 1989; Blogoslawski and Perez, 1992; Keaffaber et al., 1992). Prolonged ozonation can further oxidize hypobromite ion to bromate (BrO$_3^-$), which is another persistent and toxic compound. Unfortunately, the production conditions and toxicity towards aquatic animals of these ozonation by-products are not well understood.

2.5. Ozone destruction

Supplying an adequate level of ozone residual at the end of the contact chamber to ensure disinfection will also require that this same ozone be removed prior to the water reaching the aquatic organisms. In many cases, residuals are eliminated by water retention within tanks immediately after ozonation (Fig. 4) or by applying

small doses of a reducing agent, e.g., 1 mg/l of sodium thiosulphate (Hemdal, 1992). Dissolved ozone can also be stripped into air when passed through a forced-ventilation packed aeration column (Fig. 4) (Cryer, 1992; Summerfelt et al., in press). However, air stripping will also remove dissolved oxygen concentrations that are in excess of saturation, which may or may not be desirable. Dissolved ozone can also be destroyed by passing the water through a biofilter or bed of activated carbon, reaction with low levels of hydrogen peroxide, or contact with high intensity UV light (catalyzing the conversion of O_3 to O_2). Achieving ozone destruction with UV electromagnetic radiation depends on the wavelength of the UV light source and the quantity of energy transmitted (Rodriguez and Gagnon, 1991; Hunter et al., 1998). Ozone residuals are destroyed at UV light wavelengths ranging from 250 to 260 nm. Ironically, UV wavelengths of 185 nm can be used to generate ozone.

2.6. Ozone applications in recirculating aquaculture systems

Ozone is often used to improve water quality within intensive recirculating systems that are designed to maintain high quality water (Summerfelt et al., 2001), especially within recently constructed salmonid production systems. Ozone is most often applied to recirculating systems at doses that promote water quality improvement (Colberg and Lingg, 1978; Otte and Rosenthal, 1979; Rosenthal and Otte, 1980; Williams et al., 1982; Paller and Lewis, 1988; Rosenthal and Black, 1993; Brazil, 1996; Bullock et al., 1997; Summerfelt and Hochheimer, 1997; Summerfelt et al., 1997; Krumins et al., 2001a,b). This author has reached the following conclusions after considering the above listed research on ozonation within recirculating systems:

- Ozone is thought to impart water quality improvements by oxidizing larger and relatively complex organic molecules and thereby creating smaller and more biodegradable molecules.
- Ozone will break apart refractory organic molecules, which can reduce the color of water.
- Ozone will oxidize nitrite to nitrate.
- Ozonation may enhance fine solids removal by changing particle size (i.e., microflocculating fine particulate matter) and surface properties, which can make particles easier to settle, filter, or float (Reckhow et al., 1993). However, these effects are still not clearly defined.

In addition, ozonation of recirculating systems can reduce fish disease simply by improving water quality, which reduces or eliminates environmental sources of stress (Brazil, 1996; Bullock et al., 1997). These studies, as well as experience with ozone application at numerous commercial recirculating systems, indicates that both water quality and fish health can be improved by adding approximately 13–24 g ozone for every 1.0 kg of feed fed to a recirculating system (Brazil, 1996; Bullock et al., 1997).

2.6.1. More discussion on the use of ozone to oxidize nitrite

The fact that ozone decreases nitrite levels in a recirculating system is a substantial benefit on those occasions when bacterial conversion of nitrite to nitrate in the biofilter is lost. However, because ozone reduces the nitrite concentration going to the biofilter, it also reduces the quantity of bacteria converting nitrite to nitrate and thus reduces the total acclimated nitrite removal capacity of the biofilter. Nitrite concentrations can rapidly accumulate within fully recirculating systems when ozone addition is interrupted, because ozone can be responsible for removing a fairly large fraction of the total nitrite produced.

In addition, ozone has been occasionally applied to recirculating systems as an afterthought in order to overcome design or operational errors. For example, ozone has been added when biofilters used in recirculating aquaculture systems were found to be incapable of converting all of the nitrite produced into nitrate. These biofilter problems may be due to insufficient surface area (or volume) for completing the two-step nitrification process or may be due to an insufficient supply of dissolved oxygen. Insufficient dissolved oxygen is sometimes caused by poor solids removal that increases heterotrophic respiration within the biofilter. Ozone is then added to these systems as an afterthought to prevent nitrite from accumulating to unsafe levels. However, adding ozone as a fix is not the ideal solution. Ideally, the biofilter will be designed and operated with sufficient surface area and dissolved oxygen to complete the nitrification process, especially when it must treat higher organic and ammonia loading rates. Improved solids control before and within the biofilter will often improve biofilter nitrification and reduce the ozone requirement in many applications.

3. UV irradiation

UV irradiation can be used to destroy ozone residuals (catalyzing the conversion of O_3 to O_2) and to denature the DNA of microorganisms, causing the microorganisms to die or lose their function. Achieving ozone destruction and microorganism inactivation with UV irradiation depends on the wavelength of the UV light source and the quantity of energy transmitted (Rodriguez and Gagnon, 1991; Hunter et al., 1998). Ozone residuals are destroyed at UV light wavelengths ranging from 250 to 260 nm, while microorganism inactivation can be achieved at UV wavelengths ranging from 100 to 400 nm, although a wavelength of 254 nm is most effective. Low pressure UV bulb systems are almost an industry standard and supply monochromatic irradiation specific to the 254 nm wavelength (Fig. 5). Medium pressure bulb systems are also available, but not as commonly used, to supply a broader UV spectrum (Fig. 6). To achieve a given UV dose, medium pressure UV systems generally require far fewer bulbs (5–20% of the bulbs) but possibly 2–3 times more power than traditional low pressure and low intensity bulb systems. A low pressure but high intensity bulb system has recently been introduced to supply efficient mono-chromatic irradiation that requires only 1/3rd to 1/6th of the bulb required by traditional low pressure and low intensity bulb systems.
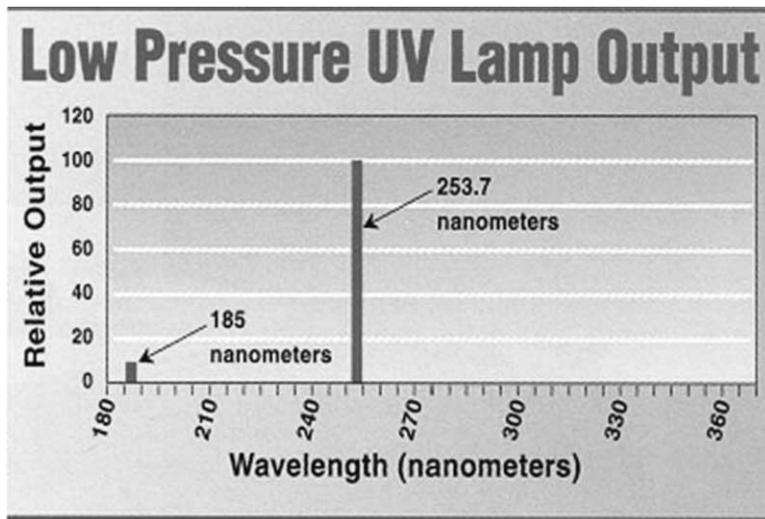
Fig. 5. Nearly monochromatic irradiation specific to the 254 nm wavelength can be supplied by low pressure UV bulb systems (Courtesy of Trojan Technologies, London, Ont., Canada).
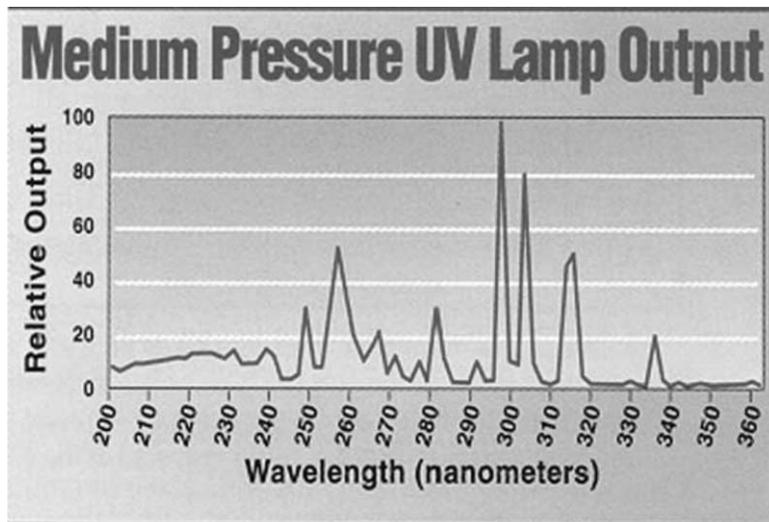

Fig. 6. Medium pressure bulb systems supply a broader UV spectrum (Courtesy of Trojan Technologies, London, Ont., Canada).

UV light intensity is described in terms of mW/cm^2 and UV dose in terms of mW s/cm^2. According to White (1992), contact times of 10–30 s are typical of many commercial UV units.

UV doses of 60–75 mW s/cm^2 have been reported to completely destroy ozone residuals as high as 0.5 mg/l (Hunter et al., 1998). UV doses required to inactivate

microorganisms can vary tremendously, from only 2 mW s/cm^2 to more than 230 mW s/cm^2 (at 254 nm), depending upon the target organism and the required kill rate (Wedemeyer, 1996). Research summarized by Wedemeyer (1996) and Liltved (2001) indicates that many fish pathogens are inactivated by UV doses of 30 mW s/cm^2, excepting of Saprolegnia, white spot syndrome baculovirus, and IPN virus (which require extremely high UV to inactivate).

However, before the UV dose can even reach the target organism, it must be able to transmit through the water. Therefore, the lowest expected UV transmittance of the process water should be established and used to predict how much UV intensity must be generated to transmit the desired UV dose through the water between the target organism and the light source. The UV filter unit should also be sized to account for the 40% decline in bulb intensity that occurs over the typical 12 month expected lamp life.

Achieving UV disinfection requires maintaining a minimum UV dose that is the product of the UV light intensity, the exposure time to this constant intensity, and a transmittance factor (see equation below). Therefore, the actual UV dose applied depends on the water flowrate (Q), the operating volume within the UV vessel (V_{vessel}), the lamp intensity (including losses through the quartz sleeve), and the UV transmittance of water (% Transm). An approximate relationship follows:

$$\text{UV dose} = (\text{UV intensity})(\text{exposur time})(\text{transmittance factor})$$

$$\cong (\text{UV intensity})\left(\frac{V_{vessel}}{Q}\right) a \exp^{(b\%\text{Transm})}$$

$$= \#\text{mW s/cm}^2$$

where a and b are coefficients that are specific to a given UV irradiation unit. The transmittance factor includes a correction for bulb spacing (as well as a correction for other factors), which is of note because UV intensity drops off as a square of the distance between the target organism and the light source (White, 1992).

The UV transmittance of spring water, partial-reuse system water, fully recirculating system water and of the facility's discharge water after microscreen filtration have been monitored at the Freshwater Institute (Table 1). The UV transmittance through the various water sources were found to be greatest in the water taken directly from the spring and degrades with intensity of use and especially with cleaning events (Table 1).

UV filters can be built as non-pressurized open channel units (Fig. 7) or as pressurized tube-and-shell units. The UV bulbs are usually contained within quartz sleeves to allow submergence in the process flow. The quartz sleeves must be kept clean to maintain transmittance (Fig. 7). Using UV light does not produce toxic residuals or form byproducts that pose a risk to aquatic organisms.

Table 1
Average UV transmittance data (across a 1-cm path length) measured on the Freshwater Institute's spring water, partial-reuse system water, fully recirculating system water and of the facility's discharge water after microscreen filtration

Water Source	UV Transmittance	
	Mean (%)	Range (%)
Spring water	97	95–98
Partial-reuse system water	96	88–98
Fully-recirculating system water	93	88–98
Facility discharge		
During normal operation	90	70–95
During cleaning period	40	30–50

Steven Summerfelt, unpublished data.

Fig. 7. Quartz sleeves are cleaned on an open channel UV system (with low pressure/low intensity bulbs) at the Freshwater Institute.

4. Concluding remarks

Care must be used when determining the effective ozone or UV dose that must be supplied to achieve disinfection. Certain pathogens may require much higher UV irradiation doses or higher ozone $c \times t$ values in order to achieve inactivation.

Applying UV irradiation for disinfection can be both less costly and less complex than using ozone. In addition, UV irradiation does not generate toxic residuals (as does ozone). However, UV irradiation may not work in situations where turbid water (and associated poor UV transmittance) may be encountered.

Applying ozone to disinfect aquaculture system influents or effluents can be quite complex and costly, yet disinfection is still necessary in many situations to control pathogen introduction. The process becomes even more complex if oxygen is produced on-site. However, there are several reasons why adding ozone within recirculating systems may not be as expensive (for a given flow treated) as adding ozone to disinfect aquaculture system influent and effluent flows. For one reason, ozone is not typically added to disinfect water flowing through a recirculating system, therefore, ozone doses added to recirculating flows are typically lower than ozone doses added to disinfect influent and effluent flows. Also, all ozone applications require ozone generation, ozone transfer into solution, contact time for ozone to react, and possibly ozone destruction to ensure that no ozone residual makes it into the culture tanks (as previously mentioned). However, adding ozone to a recirculating system can be less complicated than ozonating a water supply or hatchery effluent, because in recirculating aquaculture systems ozone transfer is sometimes accomplished using the same gas transfer unit that is used for oxygen supplementation—assuming that the transfer unit is fabricated from ozone resistant material (Bullock et al., 1997). In these situations, adding ozone to a recirculating system that is already using purified oxygen only requires installation of an ozone generator and the accompanying ozone distribution, monitoring, and control mechanisms (Summerfelt and Hochheimer, 1997). Most of the other necessary equipment (oxygen supply and distribution system, gas transfer units, and control mechanisms) are already in place. Also, the large ozone demand of the water typically found within a recirculating system causes the ozone dose to react and dissipate rapidly, which minimizes the requirement for a large ozone contactor and a dissolved ozone destruct unit (rapid ozone reaction is also a primary reason why ozone disinfection within recirculating systems is so difficult to achieve). Disinfecting system influent and effluent flows will require large ozone contactors and may also require dissolved ozone destruct units.

Acknowledgements

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