AN INTEGRATED UV/OZONE REACTOR WITH OZONE GENERATED FROM THE SAME UV LAMP

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ABSTRACT

The concept of using ultraviolet (UV) light at 254 nm and ozone co-generated from the same UV lamp at 185 nm has been revisited and put into integrated UV/ozone reactors. There was an optimum reactor diameter and an optimum gas flowrate to yield the highest ozone concentrations in the product gas mixture. The ozone generation also monotonically increased with increasing gas pressure and decreasing relative humidity. Compared to the common bubble diffuser, the venturi injector and the nano-bubble generator were better for transfer of the produced ozone from the gas phase to the liquid phase by producing much smaller bubbles. The integrated UV/ozone reactors using the venturi injector, with or without the nano-bubble generator, to deliver ozone into the water phase demonstrated more efficient micropollutant degradation and disinfection than the conventional UV system. The attributing reasons are unclear but are likely associated with the production of hydroxyl radicals and the enhancement in mixing and UV light reflection. This single, integrated UV/ozone reactor, without an external ozone source and external energy for ozone generation, provides an energy- and space-saving option to achieve simultaneous UV/ozone advanced oxidation and UV photolysis. Ozone generation and transfer remain the major challenges that deserve further investigation and development.

Key words: Advanced oxidation process (AOP), disinfection, nano-bubble generator, ozone generation, ultraviolet (UV).

INTRODUCTION

Ultraviolet (UV) or ozone treatment alone has been widely used for water disinfection and/or oxidative destruction of micropollutants (Clancy et al., 2000; von Gunten, 2003). Individually, they are selective and may not be effective in controlling some pollutants (Friedberg et al., 1995; Crittenden et al, 2005). Combination of UV and ozone gives benefits for pollution control by 1) their complementary mechanisms in pollutant destruction and disinfection (Meunier et al., 2006; Jung et al., 2008), and 2) production of highly reactive hydroxyl radicals (Reisz et al., 2003). However, the high cost of these two energy-intensive processes limits their combination in practice.

Current low-pressure, mercury UV lamp technology converts about 40% of electric power to germicidal UV-C radiation at 254 nm and around 6~9% of the power to VUV light at 185 nm. When the UV lamps are designed for disinfection only, the lamp envelope in fact is doped to prevent the emission of 185-nm VUV light, and thus this part of energy is inherently wasted (Crittenden et al., 2005). It is well known that the 185-nm VUV light can split oxygen to generate ozone (Horowitz et al., 1988). Using low-pressure mercury UV lamps to generate ozone and to achieve UV and ozone (UV/ozone) coexposure had been considered in earlier work (Dohan and Masschelein, 1987; Bolton and Denkewicz, 2008). This consideration, however, suffered from inability of the UV lamps in generating sufficient ozone. Recent advancement in UV lamp design and manufacturing, nevertheless, enables the possibility to realize this concept by increasing the output intensity and the penetration of light at 185 nm. This concept is revisited by using the 185-nm UV light to produce ozone from air flowing between a low-pressure UV lamp and its quartz sleeve. The produced ozone is introduced into the water phase in flow-through UV systems to create UV/ozone coexposure for enhancing chemical oxidation and disinfection.

The development and evaluation of the reactor consisted of the following three objectives: 1) to enhance ozone production in the gas phase by changing potential affecting factors, including lamp types, reactor diameters, installation of oscillatory baffles, gas pressure, flowrates, temperature, and humidity; 2) to enhance transfer of the produced ozone into the water phase by using various diffusers; and 3) to optimize reactor configurations with aims to maximize pollutant destruction and disinfection in the water phase.

GAS-PHASE OZONE GENERATION

Low-pressure, mercury UV lamps (GPH212T5VH/4 and GPHO212T5VH/4, Heraeus) were used for ozone generation, with feeding of ambient air or compressed air into the sleeves to supply oxygen. The former lamp is a low-output lamp (10 W) made from natural quartz glass and the latter is a high-output lamp (20 W) made with synthetic quartz. The effects of various parameters, including lamp types, reactor diameters, installation of oscillatory baffles, gas pressure, flow rates, temperature, and humidity, on the ozone generation by the UV lamps were evaluated.

First, increasing the reactor diameter from 25 to 100 mm increased and then decreased the ozone yields for both the high-output and low-output lamps (see **Figure 1**) with the maximum occurring at a reactor diameter of 56 mm. The changes in net ozone production with changes of the reactor diameter depend on the generation and decomposition of ozone, which are influenced by the residence time of the air and the distance from the UV lamp. Increasing the residence time increases ozone generation; however, a longer distance results in further light dissipation and more ozone decomposition than ozone generation. The high-output lamp produced only 10-30% more ozone than did the low-output lamp, although the former delivered UV power at 185 nm three times higher than that with the latter, suggesting photolytic ozone decomposition could be more important.

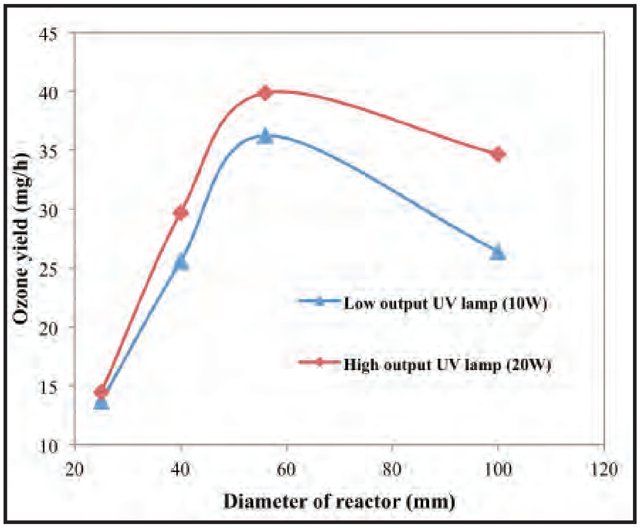

Figure 1. *Effects of lamp types and reactor diameters on ozone yields. Gas flow rate 1 L/min, temperature 22 °C and gauge pressure 0 bar.*

Increasing the gas pressure increased ozone yields monotonically (not shown), in agreement with Dohan and Masschelein (1987). Increasing the gas flow rate increased the ozone yields (Figure 2), because of the increase in the quantity of oxygen passing through the reactor per unit time. The rate of the increase, however, gradually declined at higher gas flow rates, likely because of the decrease in the residence time. The balance of the two factors resulted in an optimum gas flow rate to produce the highest ozone concentration. Therefore, the effects of gas flow rates and pressure on ozone generation followed the ideal gas law. Increasing the relative humidity from 45% to 90% decreased monotonically the ozone production from 35 to 21 mg/h. The decreasing ozone production with increasing relative humidity can be explained partially by the fact that water molecules compete with oxygen molecules in absorbing 185-nm UV photons (Falkcnstein, 1999; Creasey et al., 2000; Salvermoser et al., 2008). It is also partially arises from the enhancement of ozone photolysis in the presence of water vapor (Laforge et al., 1982). The effects of oscillatory baffles and gas temperature on ozone generation were minor (not shown).

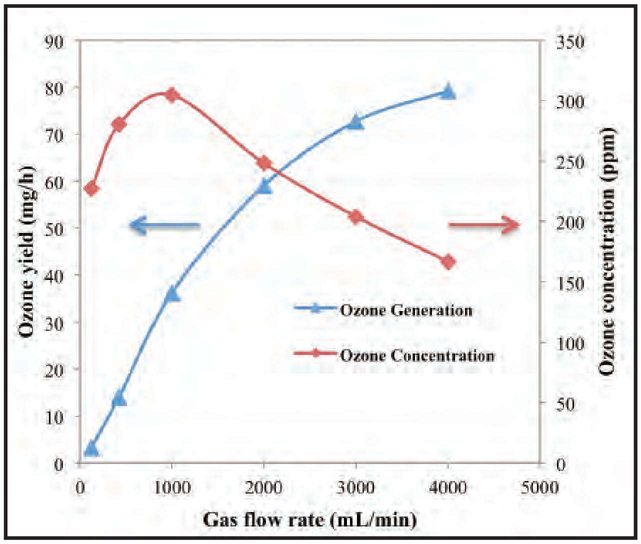

Figure 2. *Effects of gas flowrates on ozone yields and concentrations. Gauge pressure 0 bar, temperature 22 °C and reactor diameter 56 mm.*

OZONE TRANSFER

The direct delivery of the produced ozone into tap and DI water using two different injectors, a common bubble diffuser and a venturi injector (Ozone Solutions) were compared using the optimum ozone generation conditions and at same water and gas flowrates (**Table 1**). The transfer of produced ozone from the gas phase to the water phase was achieved better by the venturi injector, resulting in doubling the dissolved ozone concentrations in the water, than that achieved by the bubble diffuser. Much smaller air/ozone bubbles were generated by the venturi injector, which enhanced the air-water interfacial mass transfer rate and duration. The dissolved ozone concentrations were not sensitive to the water and gas flow rates.

Table 1. *Comparison of dissolved ozone concentrations in tap and DI water by the venture injector and a bubble diffuser*

Water flow rate (mL/min)	Gas flow rate (mL/min)	Ozone yield (mg/h)	Ozone concentration (mg/L) in			
			tap water		DI water	
			Venturi	Diffuser	Venturi	Diffuser
1100	500	18.1	0.055	0.024	0.080	0.030
1500	800	26.8	0.064	0.031	0.072	0.033
2000	1200	36.2	0.067	0.028	0.074	0.038

The venturi injector was also used together with a nano-bubble generator. The combination produced air/ozone bubbles in the micro/nano size, which further increased the

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ozone transfer rate. However, because of the significant pressure drop in the venturi injector resulting from flow restriction in the nano-bubble generator, only a relatively lower gas flow was drawn, which made the dissolved ozone concentration undetectable.

DESIGN, FABRICATION AND EVALUATION OF UV/SELF-GENERATED OZONE REACTOR

Design and Fabrication

Based on the knowledge learned from the above, three integrated UV/self-generated ozone reactors were designed and fabricated. One used a common bubble diffuser with gases delivered by an air pump. The other two used a venturi injector, with or without a nano-bubble generator (see **Figure 3**). The reactors can be operated in either flow-through or semi-batch mode. In all cases, the ozone produced in the space between the UV lamp and the quartz sleeve was transferred into the water phase by the different diffusers at a water flow rate of 2.0 L/min. The off-gas was circulated back to the gas phase (the space between the UV lamp and the quartz sleeve) to produce more ozone. **Figure 4** displays photos of the reactor using the venturi injector and the nano-bubble generator with UV light on/off and a photo of the nano-bubble generator.

Figure 4. *Photos of the reactor and the nano-bubble generator.*

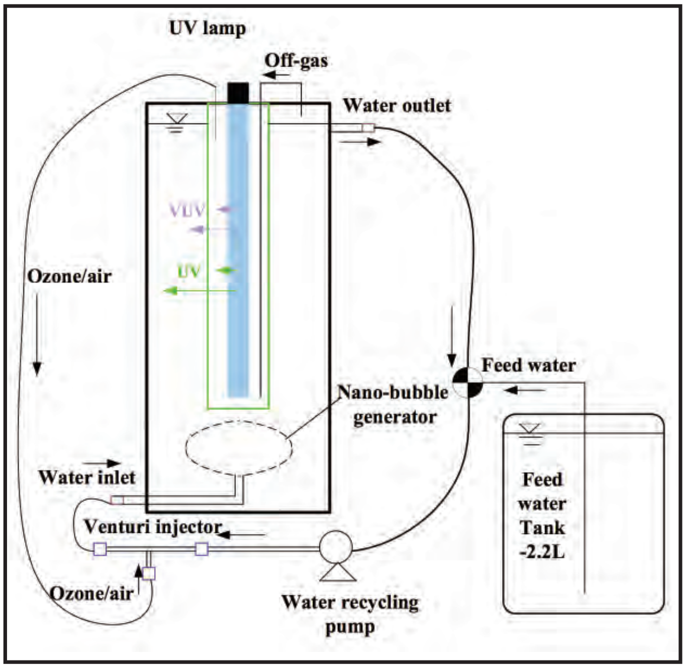

Figure 3. *The schematic presentation of the UV/self-generated ozone reactor using the venturi injector with or without the nano-bubble generator (dash line)*

Comparison of Reactors' Performance

To evaluate and compare the performance of the three reactors, *E. coli* and MS2 coliphage were chosen as the indicator microorganisms in disinfection tests and N-nitrosodimethylamine (NDMA) and nitrobenzene (NB) were the target micropollutants in pollutant degradation tests in ultrapure water. The results are shown in Figures 5 and 6. For *E. coli*, MS2 coliphage and NB, the reactors using the venturi injector alone (Reactor II) and the venturi injector and the nano-bubble generator (Reactor III) performed much better than that with a common bubble diffuser (Reactor I). The higher specific surface area of the smaller bubbles produced in Reactors II and III improved the mass transfer efficiency and, thus, should enhance the formation of hydroxyl radicals to extend the chemical destruction and microorganism inactivation. Also, the smaller bubbles existing in the water, which was not found to affect UV transmittance, might enhance the disinfection and oxidation processes by enhancing mixing and UV-C light reflection. As for NDMA removal, which relies primarily on UV photolysis, the performance difference among the three reactors was less significant.

Comparison of UV, UV/Air and UV/Ozone Reactors

A comparison was made on the performance difference for the UV, UV/air and UV/ozone reactors using Reactor III. *E. coli* was chosen as the indicator in both ultrapure water and tap water. For UV treatment alone, the venturi injector and the nano-bubble generator were removed so that only water was flowing through the reactor. For the UV/air and UV/ozone cases, Reactor III was operated in the same way as above but ambient air and ozone-containing air produced from the same UV lamp, respectively, were drawn by the venturi injector. **Figure 7** displays the results. In both ultrapure water and tap water, the conventional UV reactor, relying solely on UV exposure achieved only close to 3-log *E. coli* inactivation; while both the UV/air and UV/ozone reactors achieved over 5-log *E. coli* inactivation. It is interesting that the incorporation of air bubble at nano size in the absence of ozone also significantly increased the *E. coli* kill, supporting the hypothesis that smaller bubbles might enhance mixing and UV-C reflection to enhance inactivation. In ultrapure water, the presence of ozone in the UV/ozone reactor further increased the *E. coli* kill from 5-log to 6-log reduction. However, the enhancement became insignificant in tap water, likely because of the ozone and hydroxyl radical demands in tap water. In addition, it was also found that different water matrices and water flow rates affected the NB degradation.

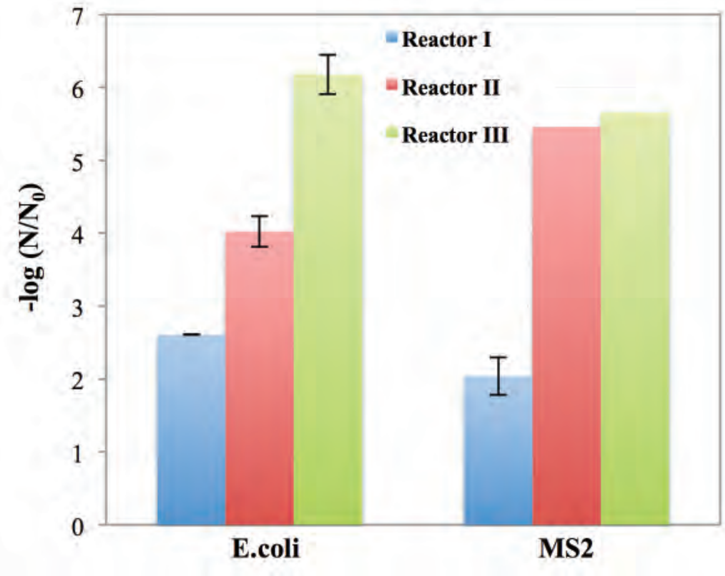

Figure 5. *Performance evaluation of the UV/ozone reactors on E. coli and MS2 disinfection in flow-through mode (40 s).*

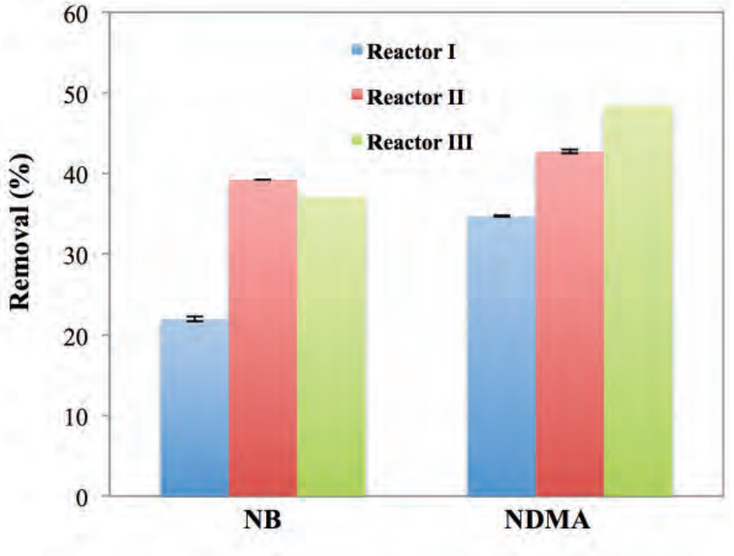

Figure 6. *Performance evaluation of the UV/ozone reactors on NDMA and NB degradation in semi-batch mode (2 min).*

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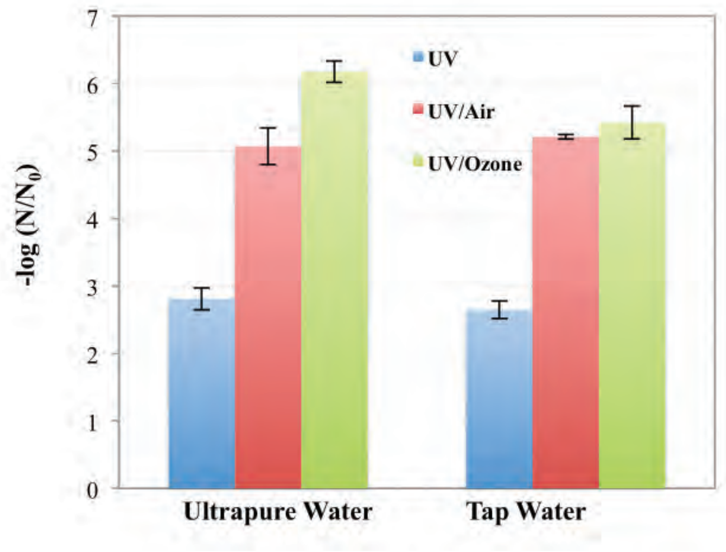

Figure 7. *Comparison of E. coli disinfection in ultrapure and tap waters by UV, UV/air and UV/ozone reactors in flow-through mode (40 s).*

SUMMARY

Ozone generation from the low-pressure UV lamp was largely affected by the reactor diameter and the gas flow rate. There was an optimum reactor diameter and an optimum gas flow rate to yield the highest ozone concentrations in the product gas mixture. The ozone generation also monotonically increased with increasing gas pressure and decreasing relative humidity. The ozone transfer efficiency by the venturi injector was higher than that by a common bubble diffuser. The integrated UV/ozone reactors using the venturi injector, with or without the nano-bubble generator, to deliver ozone into the water phase is energy- and space-saving. They provide more efficient degradation of micropollutants and disinfection than the reactor using a common bubble diffuser to transfer ozone into the water phase. The integrated UV/ozone reactor also performed much better than the conventional UV disinfection unit. Ozone generation and transfer remain the major challenges that deserve further investigation and development.

FUTURE WORK

More efforts are being made to further investigate and develop the UV/ozone system including:

1. Evaluating the effects of tap water and its composition on the system performance,
2. Optimizing the length of the UV system and the hole size of the nano-bubble generator, and
3. Studying the feasibility of using the nano-bubble generator alone to simplify the setup and to reduce the jet effect created by the venturi injector.

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