



Towards Enhancing Ozone Diffusion for Water Disinfection—Short Notes

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How to cite this paper: Ghernaout, D. and Elboughdiri, N. (2020) Towards Enhancing Ozone Diffusion for Water Disinfection—Short Notes. *Open Access Library Journal*, 7: e6253.

<https://doi.org/10.4236/oalib.1106253>

Received: March 23, 2020

Accepted: April 4, 2020

Published: April 7, 2020

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Abstract

Thanks to its performance in eliminating color, UV absorbance, trace organic compounds, and microorganisms, ozone application in treating water has acquired vogue. The intensification of mass transfer and mixing in ozone-founded disinfection methods stays so crucial to attaining the desired disinfection level at an acceptable price. This work discusses the growing tendency of improving ozone diffusion for better water disinfection. Researchers suggested a new procedure of ozonation supported by a liquid whistle reactor (LWR), which produces hydrodynamic cavitation, for disinfecting water. This integration has been observed to be a cost-efficient method for attaining extreme disinfection paralleled to the solo running of hydrodynamic cavitation (lower magnitude of disinfection) and ozonation (higher prices of treatment frequently attributed to higher cost of ozone production). Further, juxtaposed to the traditional bubble reactor, the achievement of high disinfection performance at a small Ct estimate in a rotating packed bed (RPB) depicted that RPB is an efficacious ozone disinfection contactor for killing microorganisms in water. Finally, electrogenerated ozonation could be a surpassed technique confronted with chlorination in terms of disinfection and poisoning reduction.

Subject Areas

Atmospheric Chemistry, Biological Chemistry

Keywords

Ozone, Disinfection By-Products (DBPs), Microorganisms' Killing, Electrochemical Advanced Oxidation Processes (EAOPs), Rotating Packed Bed (RPB), Liquid Whistle Reactor (LWR)

1. Introduction

Handling wastewater remains a subject of great importance [1]. At the time that wastewater is contaminated with risky anthropogenic contaminants, traditional treatments stay not constantly helpful. In this direction, electrochemical advanced oxidation processes (EAOPs), founded on forming and employing hydroxyl radicals, have revealed their fitness and surprisingly their distinguished effectiveness for dealing with such grave ecological issues [2] [3]. Between the EAOPs, it is imposing to focus on ozonation and electrolysis. The former is founded on the generation and activation of ozone [4] [5] [6] [7]; however, in the latter, a mixture of oxidants could be produced and activated thanks to the oxidation and/or reduction of constituents of the wastewater [8] [9]. Therefore, in electrochemically-founded techniques, more than ozone, several additional species could be generated like the cathodically produced hydrogen peroxide or the anodically formed chlorine, peroxocarbonates, peroxosulfates, etc. This shows the elevated performances gained by such technique in dealing with diverse contaminants spreading from dyes [10] [11] to pesticides [12] passing via numerous additional anthropogenic contaminants [13] [14] [15].

Since the 1990s, doped diamond coating materials have been suggested as anodes in electrochemical cells [1] [16] [17]. They have proved excellent characteristics like a very large electrochemical window, which is in charge of their superb potential for generating oxidants, as hard to be acquired like peroxophosphates, peroxocarbonates of perbromates [18] [19] [20]. Due to that, in employing such materials for remedying with wastewater, it is frequent that the pathways utilized to interpret the decomposition of organic compounds do not only take into account the direct oxidation via electron transfer on the anode surface but, most possibly, the mediated oxidation through a huge collection of oxidants produced from the various salts carried in the electrolyte, as well as ozone and hydrogen peroxide [21]. This begins to be an issue in disinfection due to the potential formation of toxic perchlorate [22] [23]; however, it is very favorable participation in the electrolysis of wastewater [24].

Ozone remains a powerful oxidant utilized in disinfecting water, that a stage has been performed in the implementation of wastewater treatment in traditional treatment plants. Nevertheless, one of the hardest obstacles of ozonation stays the transfer of mass among the gaseous and liquid phases. For such trouble, numerous dispositions and kinds of reactors have been engineered with a view to augment the quantity of ozone in solution. One more hurdle remains the small percentages of mineralization of organic matter (OM) due to its minimum selectivity [25]. Producing electrochemically ozone *in situ* can possess a better mass transfer and mineralization performance for remedying wastewater [1].

Concerning the electrochemical generation of ozone, even if fundamentally reported in several publications, it appears that it is not as motivating as else manners of producing such oxidants and there are numerous sides to be examined [15] [26] [27]. This could assist to clarify the weak submitting of the

technique in the market, in the face of the great capability of the implementation that it can have related. Partially, this weak performance is explicated in the matter of the minimum solubility of oxygen (ozone precursor), which successively may be in charge of stimulating else secondary responses in the electrochemical reactor. However, such subject stays of huge commercial attention thanks to the considerable utilization it may acquire in disinfecting water. Taking into account that, there are numerous commercial prototypes, most of them founded on the implementation of polymer exchange membrane (PEM) electrolyzers like the two cells estimated by Lara-Ramos *et al.* [1], fabricated by CONDIAS GmbH, and which have shown excellent effectiveness throughout their application in killing pathogens electrochemically [28] [29] [30]. At all events, the presence of such fascinating devices, a more extensive comprehension of the technology stays a large dare [1] [31].

2. New Manner of Ozonation and a Liquid Whistle Reactor for Disinfecting Water

Chand *et al.* [32] investigated a new procedure of ozone treatment supported by a liquid whistle reactor (LWR), which produces hydrodynamic cavitation, for disinfecting water employing a simulated effluent carrying *Escherichia coli*. They introduced a suspension possessing an *E. coli* [33] level of around 10^8 to 10^9 CFU/mL into the LWR to evaluate the impact of hydrodynamic cavitation single and in integration with ozone. Almost 75% disinfection could be obtained in around 3 h of treatment time employing a regulated integration of hydrodynamic cavitation and ozonation. This integration has been observed to be a cost-efficient method for attaining extreme disinfection paralleled to the solo running of hydrodynamic cavitation (lower magnitude of disinfection) and ozonation (higher prices of treatment frequently attributed to higher cost of ozone production).

3. Catalytic Ozonation and Biofiltration for Reducing the Generation of Disinfection By-Products

Chen *et al.* [34] focused on the influence of catalytic ozonation in a fluidized bed reactor (FBR) on the generation of single disinfection by-products (DBPs) [35]-[44]. They employed a biofiltration column to assess the reduction performance of biotreatment on DBP precursors. They controlled dissolved organic carbon (DOC) [45] [46] [47], simulated distribution system trihalomethanes (SDS THMs), and six simulated distribution system haloacetic acids (SDS HAA6). The source water was contaminated via urban and agricultural effluents. Catalytic ozonation reduced the level of DOC by 8.2% - 51.4% following the injection of the catalyst. The reductions of SDS THMs and SDS HAA6 were 41.3% - 51.2% and 31.7% - 48.3%, respectively, below the identical running circumstances. Biotreatment considerably enhanced the elimination effectiveness of DOC and diminished the production of DBPs. Up to 81.7%, 76.1%, and 81.3% of

DOC, SDS THMs precursors, and SDS HAA6 precursors were reduced following the catalytic ozonation pursued via biofiltration, respectively. The treatment methods also affected the fractions of solo DBP species. The fraction of bromine-containing species from the SDS THMs and SDS HAA6 augmented in water samples following being treated by biofiltration alone, ozonation alone, catalytic ozonation, and catalytic ozonation pursued by biofiltration.

A similar investigation was performed by Arnold *et al.* [48] who focused on the influences of ozone injection and empty bed contact time (EBCT) in ozone-biofiltration setups on disinfection by-product formation potential (DBPFP). They assessed the probability of employing DBPFP as a substitutional guideline for total organic carbon (TOC) removal in drinking reuse usages. They employed a pilot-scale ozone-biofiltration setup with O₃/TOC ratios spreading from 0.1 to 2.25 and EBCTs extending from 2 to 20 min. The biofiltration columns carried anthracite or biological activated carbon (BAC). Bench-scale chlorination was realized employing the uniform formation conditions (UFC) procedure, and quenched samples were analyzed for total trihalomethanes (TTHMs) and regulated haloacetic acids (HAA5s). They proved that ozone-biofiltration attained TOC reductions extending from ~10% to 30%, following running circumstances; however, biofiltration without ozone usually obtained <10% TOC removal. UFC testing illustrated that the ozone solo was performant in converting bulk OM and decreasing DBPFP by 10% - 30%. The interactive integration of ozone and biofiltration obtained average global removals in TTHM and HAA5 formation potential of 26% and 51%, respectively. A maximum TOC level of 2.0 mg/L was fixed as a recommended treatment target for reliable compliance with TTHM and HAA5 regulations for drinking reuse setups in the United States.

4. Ozonation vs. Chlorination in Terms of Disinfection *Ct* Values

Poisoning emerging from deadly DBPs remains an unwanted consequence of disinfection throughout the water recovery [49]-[54]. To make sure secure water recovery treatment, it is crucial to promote a disinfection procedure with a lower generation of total poisoning in the recuperated water [55] [56] [57] [58]. The cumulative disinfectant concentration over time (*Ct*) is a helpful idea for micro-organisms monitoring throughout reuse water disinfection [59]-[66]. Dong *et al.* [67] estimated the poisoning influence of *Ct* estimates and numerous techniques to attain similar *Ct* estimates via ozonation or chlorination of wastewaters from four agricultural sources on mammalian cells. They followed N-acetylcysteine (NAC) reactivity of the wastewater organic extracts to expose their effect on the thiol-specific biological detoxification mechanism. They illustrated that for two sources and for both ozonation and chlorination, greater *Ct* estimates improved cytotoxicity. The ozonated waters were at least 10% less toxic and as much as 22.4 times less toxic than either the non-disinfected controls or the chlorinated waters. Chlorination systematically provoked bigger cytotoxicity than ozonation

by between 2.2 and 22.4 fold, respectively, and produced identical or greater cytotoxicity than the non-disinfected controls, by at most 4.4 fold. Assuming the identical Ct estimates, integrating elevated disinfectant concentration and short contact time generated finished wastewaters with bigger toxicity than the integration of low disinfectant concentration and long contact time. NAC thiol reactivity was positively and importantly matched with mammalian cell cytotoxicity and agreed with 80% of the cytotoxicity rank order. This proposes that the induction of cytotoxicity implied reactions with agents that acted as thiol pool quenchers. The total findings depict that the cytotoxicity of wastewaters could augment if bigger Ct estimates are implemented to demobilize recalcitrant microorganisms. To offset the possible elevation in cytotoxicity at elevated Ct estimates, for both ozonation and chlorination, smaller disinfectant injection and longer contact time could be followed.

As shown above, the intensification of mass transfer and mixing in ozone-founded disinfection methods stays so crucial to attaining the desired disinfection level at an acceptable price. Liu *et al.* [68] utilized rotating packed bed (RPB), an efficacious process intensification apparatus, as an ozone-disinfection contactor, and assessed its effectiveness on disinfection and mass transfer through the demobilization of *E. coli* and mass transfer coefficient ($k_L a$). The ozone exposure (Ct estimate) and the log demobilization of *E. coli* in the RPB attained 0.0008 - 0.0014 min \times mg/L and 6.8 - 7.3 in phosphate-buffered saline buffer, respectively. And the $k_L a$ in RPB augmented from 0.030 to 0.186 s⁻¹ with the elevation of liquid flow rate from 10 to 60 L/h. The elevation of the rotation speed of the RPB ameliorated the log demobilization and $k_L a$ together. Further, it was shown that a bigger gaseous ozone level at a similar used ozone dosage is suitable for demobilizing *E. coli*. The computation of Hatta number revealed that the demobilization of *E. coli* in RPB is a diffusion-controlled process for which RPB is well convenient. Paralleled to the traditional bubble reactor, the achievement of high disinfection performance at a small Ct estimate in RPB depicted that RPB is an efficacious ozone disinfection contactor for killing microorganisms in water.

On the other hand, because of the quick response of ozone with OM, dissolved ozone is usually not measurable and thus, the frequent disinfection monitoring parameter, concentration integrated over contact time (Ct) cannot be acquired. In such circumstances, substitutional parameters have been proved to be helpful as surrogate measures for microbial elimination comprising a variation in UV₂₅₄ absorbance (ΔUVA), change in total fluorescence (ΔTF), or O₃:TOC (or O₃:DOC). Even if such measures have proved potential, some cautions stay. These involve uncertainties in the relationships among such measurements and microorganisms' demobilization. Then, former utilization of seeded microbes with greater disinfection sensitivity juxtaposed to autochthonous microbes can conduct to over-estimation of proper log credits. Carvajal *et al.* [69] ozonated secondary treated wastewater from a full-scale plant in a

bench-scale reactor employing five increasing ozone injections. Throughout the trials, the reduction of four indigenous microbial indicators representing viruses, bacteria, and protozoa was followed at the same time with Δ UVA, Δ TF, O_3 :DOC and PARAFAC derived components. They utilized Bayesian methods to fit linear regression models, and the uncertainty in the posterior predictive distributions and slopes provided a comparison between previously reported results and those reported here. Integrated findings illustrated that all surrogate parameters were helpful in anticipating the elimination of pathogens, with a better fit to the models using Δ UVA, Δ TF in most situations. Average adjusted determination coefficients for fitted models were elevated ($R_{\text{adjusted}}^2 > 0.47$). With Δ UVA, one unit decrease in log10 reduction value (LRV) corresponded with a UVA mean removal of 15% - 20% for coliforms, 59% for *C. perfringens* spores, and 11% for somatic coliphages. With Δ TF, a one-unit decrease in LRV corresponded with a TF mean removal of 18% - 23% for coliforms, 71% for *C. perfringens* spores, and 14% for somatic coliphages. Compared to former researches also analyzed, these findings propose that microbial removals were more conservative for autochthonous than for seeded microbes. Their results proposed that site-specific analyses have to be performed to produce models with lower uncertainty and that indigenous microorganisms are helpful for the measurement of system efficiency even when censored observations are obtained.

5. Integrating Coagulation and Ozonation before Ultrafiltration

Ultrafiltration (UF) process is seen as a performant water treatment technology. Lately, there is an increasing tendency to improving the treatment performance of UF and mitigating membrane fouling [70] [71]. An efficacious strategy is to attain this target via employing coagulation as a pre-treatment to UF; nevertheless, throughout a long term running, membrane fouling could happen because of the remaining OMs and the formation of microbes. Bu *et al.* [72] merged coagulation and ozonation as a pre-treatment of UF and paralleled four kinds of pre-treatment: coagulation, ozonation, coagulation followed by ozonation (C-O) and ozonation followed by coagulation (O-C). They established that the ozonation sole did not do well. Both integrations of coagulation and ozonation ameliorated the OM reduction performance, even if membrane fouling was lowest following the C-O pre-treatment since the microbial load was decreased (Figure 1). During the time that a small augmentation in the yield of DBPs was detected with the introduction of ozone, the level stayed under the Chinese standard for tap water quality (100 mg/L).

6. Conclusions

This work presents a growing interest in improving the diffusion of ozone in order to ameliorate the disinfection of water efficiency. From this work, the following conclusions can be drawn:

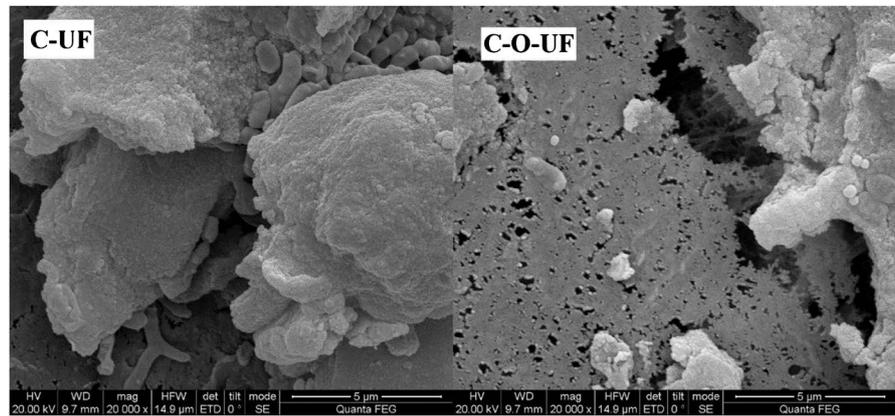


Figure 1. Scanning electron microscope (SEM) images of the membrane surface from the C-UF and C-O-UF systems [72].

1) Employing an LWR single, hydrodynamic cavitation produced findings in only 22% disinfection; however, ozone-assisted running with a lower time of ozone treatment outputs 75% disinfection. Thanks to the circulatory flow in the LWR, the ameliorated mass transfer rates attained increase the efficient usage of ozone. Chand *et al.* [32] deduced that the best working treatment protocol is the introduction of ozone in phases that helped via hydrodynamic cavitation at lower running temperatures. If keeping lower running temperatures remains not practical, then the identical magnitude of disinfection could be obtained through employing higher working pressures in the LWR (elevated support from the hydrodynamic cavitation). Bigger pressures and identical circumstances of ozone injections stay mostly suggested for disinfecting wastewaters with some organic loads and different pollutants. Regulating ozone injection remains important in coupling with hydrodynamic cavitation with a view to suggesting a technique, which will expend less energy and simultaneously be more cost-effective.

2) Dong *et al.* [67] deduced that bigger disinfectant subjection, in general, conducted to greater degrees of mammalian cell cytotoxicity in agricultural wastewaters. Considering the same disinfectant subjection, smaller disinfectant concentration integrated with longer contact time was proved to form smaller poisoning than bigger disinfectant concentration integrated with shorter contact time. Disinfectant boosting method, where disinfectant is utilized at smaller Ct estimates, and at the end of a disinfectant subjection period another low Ct estimate boost disinfectant is used to meet the total Ct design, may diminish poisoning. Ozonation could be a surpassed technique confronted with chlorination in terms of disinfection and poisoning reduction.

3) Throughout their examination of the demobilization of *E. coli* via using RPB as an ozone disinfection contactor, Liu *et al.* [68] discovered that a favorable demobilization impact was obtained at a smaller Ct estimate in the RPB juxtaposed to the bubble reactor, and a bigger rotation speed of RPB and gaseous ozone level were appropriate for the mass transfer of ozone and the demobilization of *E. coli*. In addition, it was noted that the $k_t a$ in RPB augmented with the

elevation of liquid flow rate and was greatly bigger than that in the traditional bubble column reactor. Thanks to the singular fluid flow features, there are a great gas-liquid interface and violent interface renewal rate in RPB. Consequently, it is easy to reach the desired demobilization impact of *E. coli* at a small *Ct* estimate, which implies that RPB could be used in disinfecting water with the merit of small price and small volume of equipment.

4) Juxtaposed to distinct pre-treatments, integrating coagulation and ozonation improved the UV₂₅₄ reduction performance of UF [72]. As for the DOC elimination, there was no considerable gap among the C-UF, C-O-UF, and O-C-UF methods. The aggregation of OM was reduced in the C-O method via fragmenting hydrophobic macromolecules into smaller hydrophilic molecules. Injecting ozone following coagulation somewhat augmented the DBP yield; this must be a worry while employing C-O-UF procedure in treating. C-O-UF technique attained the smallest rate of elevation of transmembrane pressure. Introducing ozone reduced microbial products such as polysaccharides and proteins and extenuated membrane fouling [72].

Acknowledgements

This research has been funded by the Research Deanship of University of Ha'il, Saudi Arabia, through the Project RG-191190.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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