Iraqi Journal of Oil & Gas Research

Journal homepage: https://ijogr.uotechnology.edu.iq

A trends in Ozone Treatment of Wastewater: a Review

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Article information

Article history:

Received: February, 1, 2022

Accepted: February, 25, 2022

Available online: April, 8, 2022

Keywords:

Ozone,

Wastewater,

Tri-oxygen

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Abstract

In recent years, great efforts have been done by the scientific community to develop many green chemical processes to treat water and wastewater effluent. Taking into consideration these new approaches, a new environmentally friendly technique is giving special attention such as ozonization. Molecular ozone, or tri-oxygen, can be considered as an inorganic molecule with the chemical formula of O_3. Ozone is a pale blue gas having pungent smell and far less stable molecule than the molecular O_2, decaying in the lower atmosphere to free O_2 gas. Ozone is an efficient gas for killing bacteria and eliminating endotoxins and biofilm, with effectiveness being dependent on concentration and operating time. In addition, because the ozone is a strong oxidant, suitably compatible materials ought to be adopted. Moreover, the manufacture of ozone should always be supervised and its level checked occasionally in the surrounding environment to keep a pollution air standard of less than 0.1 ppm. In this paper, essentials and practical features dealing with ozonization system and its utilization are presented and discussed.

DOI: http://doi.org/10.55699/ijogr.2022.0201.1016 , Department of Petroleum Technology, University of Technology-Iraq

1. Introduction

The improper release of polluted wastewater from various resources is a major factor behind environmental issues nowadays. The growth of human activities has countless harmful effects on freshwater quality and its quantity. Thus there is a pressing requirement for apposite procedure of wastewater treatment. Certain physicochemical and biological restrictions should be exhibited to make water usable for everyday use and within adequate range. Therefore, to get this request, practical treatment is necessary and many approaches have already been emerged recently. One of the developing technologies is to use ozone gas. There are in the literature a number of published papers on the ozone treatment for water and wastewater [1-3], these are described in this reviewing paper. The extraordinary reactivity of ozone was illustrated by Schönbein in his primary research, dictating an attentive selection of constituents in which to contain the ozone. The reactivity of ozone originates from its characteristic instability that arises from its molecule´s inclination to take an electron, reducing ozone to O_2, then the electron donor converts oxidized. What provides the ozone its extraordinary redox potential, which makes it capable to experience redox reactions. The ozonation trend is suggestively inclined by wastewater structure. The constituents found in the water could pledge, indorse, or hinder the radical chain reactions. Owing to the ozone high selectivity, there are many prospects to use it as an oxidation procedure for the disinfection of wastewater [4-8].

2. Type of Ozone Treatment

Ever since the finding of O_3, this reactive gas has been considerably adopted for cleansing water/wastewater, because of its remarkable disinfectant action [9]. The making of municipal water necessitates the elimination of many constituents (i.e. toxic impurities and humic elements) and O_3 performs by oxidizing numerous water pollutants, including various compounds (organic and inorganic), besides the removal of many pathogenic organisms (e.g. viruses, parasites) [10-20]. There are some presents papers suggested guidelines for the existence of by-product created by disinfection, for example trihalomethanes and haloacetic acids, that are obtained from cleansing of organic contaminants by using halogenic materials [21-43]. These papers reported the characterization of treated water for the ozone application, and examined some parameters such as the ozone capacity, and sequential ozone dose. They exposed that ozonation is an effective technique for water/wastewater treatment and found that the quantity of O_3 needed for water remediation relying on the volumetric flow rate and the characteristics of the treated water. The kinetics plays an important part in the viability of removing organic contents found in water. Differences associated with the direct/indirect nature of the ozone reaction route were also reported, showing the direct ozonation kinetics for many well-known pollutants.

2.1. Ozone in drinking and groundwater water treatment

Water contaminations, particularly in the surface water that is in close interaction with various pollutants, force authorities to add chloride to water, which is commonly used all over the world. This may create some sort of opposing influences on the residential users, i.e. producing by-products such as chlorinated chemicals and Trihalomethanes. O_3 is more oxidant than chlorine as disinfecting substance besides that other oxidants are relatively unstable in aqueous environment, having a half-lifetime of around 25 min at 20 ^oC. Remediation with O_3 should easily eradicate most bad tastes and odors in water supplies upcoming from pollution besides the influence of other contaminates (i.e. algae and actinomycetes) that may generate unpleasant tastes [36, 44-47].

2.2. Ozone for reducing suspended solids

The ozone procedure lessens the degrees of the suspended solids without any TDS added to the effluent, besides its operative virus removal and cost effective over other treatment systems. In this process the dosage of ozone needed is 10 mg/l [48]. G. Moussavi et al. [49] reported the behavior of ozone reaction on sludge with the purpose of evaluating the efficiency of changing ozone concentrations on sludge dissolution and mineralization using TS, TSS, and VS. The outcomes specified that the ozone at 0.125-2 gO3 g-1 TS would reduce the values of TS, TSS and VS, concluding that the ozone could mineralize a great portion of volatile solids so that the minimum reduction of VS in waste activated sludge was attainable via this process.

2.3. Ozone and removal of turbidity

Ozone has been found to be an effective procedure in reducing turbidity, and changing the suspended solids/ dissolved solids into the particular stabilized formula. Loss Angeles city in California has an ozone treatment plant with a capacity of 580 mgd of tap water used to reduce turbidity to about 0.3 NTU. This large water treatment plant was established in 1986 depending on ozone system. In this plant, ozone is produced (with a dose of 7,300 Ib/day) to treat water and reduce turbidity to less than 0.3 NTU. The high filtration rate is made applicable by adopting O_3 as a flocculant [48]. More recently, P. Setareh et al. [35,50] showed that pre-treatment with US/O_3 would improve the action of coagulation process in eliminating the turbidity of the surface water. The influence of many input parameters (i.e. ozone concentration, power intensity, US frequency, exposure time, coagulant type, and coagulant concentration were studied by the authors. Specific coagulation procedure with ferric chloride reduced turbidity to about 15%, yet pre-coagulation with ferric chloride integrated with US/O_3 enhanced the reduction of turbidity up to 32%.

2.4. Ozone in paper industry effluent treatment

Pollution triggered by discharging of wastewater produced from pulp and paper manufacturing has been treated using ozonation technique. This waste comprises of many types of Hgno-cellulose’s structure, deeply polluted and highly colored, particularly when the system utilizing synthetic coloring agents. Here traditional wastewater treatment frequently shows to be of inadequate efficiency that makes ozone necessity to remove color from the discharged water having humic material. It is hard to conclude the precise chemical outcome of ozone on the humic structure. O_3 interacts with the agents accountable for coloration, decaying species with high electron densities. These contain pollutants; the tannins, humic acids, and lignin generally originated in vegetal substances. O_3 dose in the range of 10-15 mg/l is needed to lower the contamination of this industrial wastewater water to substantial degree [39, 51]. In addition, Z. Wang and T. Yang [52] reported the treatment of waste paper pulping effluent using ozone-water combined with other processes. The authors claimed that this process can lessen wastewater treatment cost significantly, and surge the processing speed of wastewater.

2.5. Ozone in dyeing industry effluent treatment

The waste formed by dyeing production holds various synthetic coloring agents. The discharged water from this industry can be classified according to different factors. Physical properties such as color, turbidity (SS and DS) may be included, while the properties may contain the constituents of basic and acidic dyes, sulphides dyes, azo dyes, and metallic complex dyes. In general, biological remediation may be used to treat the wastewater color but within limited range due to limited biodegradable of the molecules responsible for coloration. Nevertheless, this can be enhanced by applying aeration/oxygenation procedure. This may include the annihilation of the double nitrogen-nitrogen bonds, which is established by the presence of NO and NO_2 radicals following the O_3 system [53]. Guendy et al. [10] Studied decolonization of dyes (Direct Pink 3B and Reactive Violet SH- 2R) using ozone in aqueous environment in with some operative variables. The process was found to be effective for the decolonization of textile wastewater (98% decolonization in few minutes). Changing some regulatory parameters would provide very efficient decolonization [20].

2.6. Ozone in treatment of toxic waste

O_3 has been applied in the water and wastewater treatments especially in the tertiary stage for the eradicating all traces of dangerous toxic substances. The ozone here gives great advantage in the treatment process where adding additives is required, which may increase toxicity than the pollutants removed. In toxic assortments, it is difficult to make ozone react specifically with the constituent that we demand to eradicate. Thus, it is desirable to apply conventional purification systems before using ozone procedure as a later stage. As we mentioned the concentration levels are a critical issue when dealing with toxicity. Accordingly, ozone should be applied as a final stage in the treatment process to ensure removing all traces of harmful constituents [17, 36].

2.7. Ozone in removing heavy metals

Ozone can easily oxidize transition metals to their higher oxidation state to produce less soluble oxides, thus easily can be separated by filtration procedure. Furthermore, the ozone system can also be adopted to eliminate metals like Cd, Cr, Co, Cu, Pb, Mg, Ni, and Zn. In general, in systems such as mineral extraction plant, O_3 oxidizes heavy metals to generate metallic oxides/hydroxides, later can be precipitated and separated easily from water [54]. Recently, the removal of zinc ion from water is tried by S. Khanom and N. Hayashi [55] using Dielectric Barrier Discharge Plasma system. A maximum removal rate of 29% of zinc ion from the treated water was achieved at 10 min, and pH $\leq$7.4. The pH of the treated water depends on both zinc ion and ozone dose in the water.

2.8. Ozone in elimination of phenols

There are many purification approaches that exist to remove phenols harmful compounds. One important procedure is the oxidation of O_3 gas. Thorough investigations have been carried out into the approach O_3 reacts with phenols. Principally, ozone splits phenols to create oxalic acids and oxygen. The mechanism reviled that several parameters may influence the effectiveness of this system. Mainly, among these parameters are mostly the pH and the system temperature. Therefore, when the pH holds in the range 8.0-11.0, it will double the rate of reaction at which phenols are splitting. O_3 can particularly attack phenols at higher pH values in comparison to the other oxidizable matter, thus reducing reaction times [9]. O_3 can effortlessly react with organic contents with low reaction rate, especially with saturated organic compounds, which limits its efficacy. Furthermore, a high conc. of O_3 must be used in order to mineralize organic species, resulting in high treatment costs. Presently, it was suggested that the concurrent adopting of O_3 with absorbent constituents (i.e. silica-gel, activated carbon (AC), and zeolites) would improve the mineralization of phenol in wastewater and significantly decrease the costs. The performance of phenol removal in the O_3/AC system were thoroughly investigated by W. Xiong [56]. It was established that the phenol deprivation and TOC removal efficacy were affected by the ozone dosage, AC dosage, and pH of the water. This work offers more elucidation of the mineralization mechanism of phenol in the O_3/AC process.

2.9. Ozone in Wastewater Treatment

Ozone can break down the cell membrane to yield disinfection, causing a drop in SS value, and a rise in soluble COD contents; accordingly, the process can be considered as an effective step for industrial wastewater treatment [57]. O_3 may be adopted in a one-step remediation method, as a pretreatment system tailed by other traditional procedures, or can be applied as a post-treatment stage to attain discharge limits and particularly to eradicate micro-pollutants that have not yet been approached in the treatment policies and regulations [21-38]. Among literature reviews concentrated on the applied utilization of micro-pollutants elimination techniques, studies designed at covering current wastewater treatment plants using other tertiary treatment processes [12, 26, 36, 58]. This is because those plants are the major causes of pollutants in relations to their supply to separate sections of the marine ecology. The advantages of using O_3 with other processes is that they can offer a comprehensive mineralization of organic constituents to CO_2 and H_2O or enhancing bio-treatability by converting hydrophobic organic mixtures into more polar mixtures [59].

Ozone maximum concentration generated from air/O_2 is around 4–8%, together with low active efficacy of the manufacture and the obligation of a dry air feeding [15, 18]. Operating efficiency is quite highly which relies on the effective gas–liquid mass transfer, hard to reached because of the small solubility of O_3 in water environment. On the other hand, the main cons of O_3 process is the deficient mineralization of contaminants and the establishment of changeable by-products. Yet, O_3 indorses the incomplete oxidation of toxins. Accordingly, recently advanced procedures adopting O_3 just for the purging of unsafe constituents and for incomplete oxidation of resilient wastewater organic contaminants to enhance their features [60].

Ozone system can be integrated with many systems, i.e. photochemical processes [61,62], photo-Fenton processes [63], and photocatalysts methods [64]. Thus, combined both chemical and biological oxidation procedures have a recognized possibility of eliminating stubborn and anthropogenic constituents from treated wastewater. It may be illustrated that advance oxidizing processes could occasionally yield by-products, that may constrain the bi-systems when O3 integrated with the biological treatment processes [2, 12, 21-38, 58]. A new process of combining ozonation with coagulation process was studied for the elimination of o-dichlorobenzene and humic acid from tap water [65]. In this process, applying clays has improved the coagulation process, producing an improvement in the elimination efficiency of humic acid. Previous reported experimental work has reviled that O_3 can be positively applied to eradicate pollutants from sewage [66]. A wide range of organic micro-pollutants has been removed effectively using ozonation system in Regensdorfe plant - Switzerlan. A removal efficiency of more than 90% was obtained at relatively low ozone doses. Contaminates such as hormones, antibiotics, analgesics, etc. were practically totally detached. Thus, after ozonation, additional step is essential, i.e. filtration to eliminate reactive oxidation products, besides removing bacteria, scent, color and foam.

An integration diverse filtration devices (i.e. trickling filtration slow and sand filtration) plus ozonation was studied as an improved tertiary stage of urban wastewater subsequently after the bio-process [67]. The filtration approaches demonstrated to be unproductive for the elimination of micro-pollutants in the wastewater. However, a third filtration method that used activated bio-carbon stated a removal efficiency of 99%. Yet, the running cost was found to be high for fruitful complete application. The ozone procedure detached most of the pollutants from the wastewater but yielded in producing more discharged nonselective toxicity. It appears that the best procedure for removing micro-pollutants and preventing toxicity of secondary effluent was by using slow sand filtration followed by the ozone system.

Another points needed to be considered during the application of ozonization that is the substantial upsurge in geno-toxicity which was elucidated as a result of the founding of many hazardous changeable products. This disadvantage was solved by using a sequential sand filter. Outcomes specified a larger reduction in the landfill leachate toxicity by O_3 in comparison to the Fenton process. The practical outputs maintain the anticipation that the integrated processes of O_3/bio-filtration may improve the efficiency of the remediation [68 ,69].

The creation of additional slurry in the biological treatment systems may not be evaded. The utilization of stabilized sludge to the soil can origin the discharge of risky, damaging, and toxic constituents into the soil and groundwater. Furthermore, compounds adsorbed on the activated sludge may be discharged into the surroundings as volatile species and other products. An example is the process of using O_3 to eradicate carcinogenic and toxic materials from waste of biological sludge. The system adopted over O_3 of the anaerobically, it stabilized sludge and re-exchanged into the stabilization reactor. In combination with H_2O_2, the RE of aromatic hydrocarbons increased by 31% [70, 71]. Other results have also established the O_3 of anaerobically stabilized sludge and its re-exchanging into the reactor, besides increasing the reduction reactions in organic content, upsurge bio-gas making and decreased the contents of micro-pollutants. Also, the O_3

treatment of extra sludge may lead to increase the removal efficiency of endocrine-disrupting materials absorbed onto sludge [72].

3. Evaluation and Future Work

As have been previous agreed with, the ozonation and associated oxidation processes have been widely recognized as advance procedure in water treatment technology, due to their numerous advantages over other techniques such as chlorine. We can conclude that the key ozone advantages of wastewater disinfection are listed as follows:

- Ozone can be considered relatively safe if dissolved in water and comes in contact with the human skin, there will be no irritation to the skin, nose, or ears. Also the ozone will not leave any chemical particles on skin or make the skin dry out.
- There will be no hostile taste or smell produced from ozone solubility in water.
- Because ozone is unstable gas, we cannot increase the added dose to the treated water since the excess, insoluble ozone will simultaneously convert to free oxygen.
- Ozone have no side effect on water and produce no damaging by-products in comparison with the unsafe chlorinated by-products in the treated water.
- Adding chemicals to water may cause long-term negative effects on the environment whereas ozone causes no such problem.

The combined methods provide the prospect of improving extra treatments, such as coagulation/flocculation or filtration with granular activated carbon (GAC). Furthermore, they permit the deprivation of impurities that were intractable to chlorine, due to a higher oxidative potential. Obviously, information of both the reaction rate constants and the kind of the by-products are of key significance to guarantee an effective procedure (i.e. removal of the toxic chemicals originally found in water, with succeeding reduction of the produced by-products). Yet, decomposition is rarely comprehensive and many materials commonly endure after oxidation. This may create numerous issues such as the regrowth of bacteria in the distribution network and increasing the activity of the mutagenic. Thus, the ozonoization process should be followed by a biological stage (either sand or GAC filtration) before the final cleansing and the distribution procedure.

Previous practical results suggest that the remediation with O_3 are of great promise for advanced COD reduction of industrial wastewaters. However, the biological treatment prior to the ozonation procedure is necessary to remove biodegradable organic contents that may compete with intractable organics for oxidants. Thus, biological removal of organics preceding ozonation would assist an effective and economic chemical oxidation of the bio-refractory constituents. This process may also form humic acids which have been revealed to trigger ozone itself to decay organic contents which are not attacked by ozone in the absence of humic. Clearly, H_2O_2 is shaped by the ozonation of humic material and reacts with ozone causing in the making of OH radicals. Partial chemical oxidation of organic wastewater constituents with O_3 or O_3/H_2O_2 will not constantly improve the biodegradability. But refractory organics of biologically treated oil reclaiming wastewater can be changed by ozonation to constituents which are vulnerable to biological decomposition.

Economic issues of ozonization are also a vital concern in treating water/wastewater. The costs related to ozone making have fallen by 50% in the last decade and, thus, a great number of novel industrial utilizations has performed in recent years. A vital feature of ozone usage in water remediation is its application as sterilizer in filtered water loops for the pharmaceutical and electronic industries. In addition, it is worthy to remark that O_3 claim, different from that of chlorine, which does not produce damaging deposits such as haloforms after interaction. Consequently, the eco- advantages of O_3 over halogens validate its relatively higher cost of production for a growing number of utilizations.

Finally, studies have shown that ozone was effective against several types of viruses and suggested that ozone disinfection could be used as a disinfection method. Low ozone concentrations can be used to treat the air in naturally ventilated hospital rooms, providing an additional tool for hospitals that do not have heating, ventilation and air conditioning systems. Ozone gas has a considerable advantage in disinfecting virus contamination in the aerosol, whereas ozone water is more effective in surface disinfection simply by immersing contaminated material in the ozone water. However, both forms of ozone must be administrated with caution to prevent lung injury due to the inhalation of ozone gas.

Future prospective of ozonization technology for water and wastewater treatment should emphasis on the following topics:

(1) Research and development should explore new combined processes to maximize the efficiency of different procedures to evade the process limitations. Our lab here at University of Technology-Iraq currently works on AOP combined with ozonization to treat mostly oily wastewater.
(2) Comprehensive investigations of water/wastewater decomposition mechanism, are required to improve remediation effectiveness and reduce operating costs.
(3) Support the environmentally friendly approach and sustainability to research.

4. Conclusions

Ozonization is a chemical procedure used for water/wastewater treatment relying on the infusion of ozone into an aqueous. Ozone has a wide range of applications in water/wastewater treatment, since it is effective disinfector besides its degradation ability against organic and inorganic contaminants. Ozonization possesses a number of advantages over other chemical disinfectors, i.e. its fast reaction with bacteria, viruses and protozoa at a wide range of pH, its tougher germicidal characterization in comparison to chlorination, no chemicals need to be added to the treated water, its high removal efficacy for organics, inorganics, color, taste and odor. On the other hand, Ozonization drawbacks includes: its high operating and mountainous costs, its energy consumption is high, it requires trained technicians and engineers for the design and equipment maintenance, in some cases and with the presence of certain chemicals such as brome, potentially damaging by-products may be formed during treatment of water, and finally the possibility of causing fire and toxicity with its generation is likely. In general, O_3 disinfection is accomplished at medium to large sized plants following up a secondary treatment. Ozone treatment of water and wastewater has the potential of achieving high levels of disinfection in comparison to other halogenated compounds or UV. However, the capital costs besides maintenance expenses are still high to make it not competitive with the accessible options. Therefore, we can determine that ozone is applied only sparingly, predominantly in exceptional circumstances where alternatives are not operative. Finally, future work may include further comprehensive studies to reduce operation cost and improve the ozone generation technology. Meanwhile, extra research studies are required to use combined processes in order to improve water/wastewater treatment efficiency.

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