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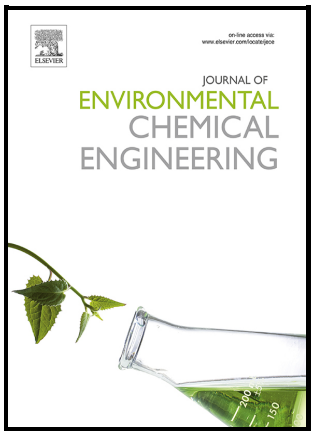

PII: S2213-3437(21)01076-9

DOI: https://doi.org/10.1016/j.jece.2021.106099

Reference: JECE106099

To appear in: *Journal of Environmental Chemical Engineering*

Received date: 19 April 2021
Revised date: 5 July 2021
Accepted date: 19 July 2021

Please cite this article as: Collin G. Joseph, Yan Yan Farm, Yun Hin Taufiq-Yap, Chuan Kian Pang, Janice L.H. Nga and Gianluca Li Puma, Ozonation treatment processes for the remediation of detergent wastewater: A comprehensive review, *Journal of Environmental Chemical Engineering,* (2021) doi:https://doi.org/10.1016/j.jece.2021.106099

Ozonation treatment processes for the remediation of detergent wastewater: A comprehensive review

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ABSTRACT

Laundry detergent wastewater is a potential renewable resource that can be recycled and reused in order to mitigate water scarcity. The treatment of laundry detergent wastewater is very challenging because of its multicomponent composition, large discharge volumes to the environment resulting from increasing usage of detergent as the global population grows and ineffectiveness of conventional treatment technologies. Ozonation as one of the most effective advanced oxidation processes (AOPs) has shown tremendous potential in the treatment and reclamation of laundry detergent wastewater. Complete mineralization of the water contaminants by molecular ozone is not economical due to the high ozone generation

cost and other limitations such as pH dependence, short lifetime of ozone, low ozone solubility in aqueous solution and mass transfer limitation. Strategies such as modification of ozonation processes including combination with hydrogen peroxide (peroxone process) and catalytic ozonation have received much attention. The effectiveness of the ozonation process can be further enhanced by photocatalytic ozonation resulting in higher rate of pollutants mineralization. This paper reviews the recent studies on ozonation treatment of surfactant containing laundry wastewater generated from various sources including domestic, industrial and commercial or public premises. Lastly, important remarks on the ozonation treatment of laundry detergent wastewater and suggestions for future works are presented.

Keywords:

Ozonation Treatment, Advanced Oxidation Process (AOP), Detergent, Laundry Wastewater, Surfactants

1. Introduction

Environmental degradation due to ever growing contamination of water resources, increasing human activities, industrial wastes and climate change has had a significant impact on fresh water supplies [1,2]. Thus, water reuse increasingly being introduced as a method to mitigate water shortages while preserving the environmental status of the remaining fresh water resources [3]. One of the major sources for water contamination originate from every growing world population and increasing amount of laundry wastewater [4]. The large discharge amount of laundry detergent wastewater in the range of 200 to 2,100 litres per day produced by domestic, industrial and institutional sectors [5–7] in the average daily discharge amount ranging from 200 to 2100 litres per day represents a potential renewable resource for water reuse on a variety of applications in the agriculture, industrial and urban sectors [8] supplementing the existing water shortages.

The characteristics of laundry wastewater depend on the type, concentration, and amounts of chemicals added, the type of cleaned items and the degree of soil removed during the laundry process [9]. The organic loads measured by chemical oxygen demand (COD) are in the range of 1,200 to 20,000 mg/L are reported for laundry wastewater [3]. High concentration of surfactants, phosphates, suspended solids, organic matters, pathogens, nutrients and salts that emerging from the residues of laundry detergents and fabric softener [10–12] are often encountered. Laundry wastewater generally has high pH due to the presence of alkaline materials and surfactants [13,14]. Phosphates as the sequestering and chelating agents are added to achieve stronger laundry detergent cleaning power particularly in industry and public institutions [15,16]. Phosphates act as the nutrients to the environment and promotes excessive growth of algae, which cause eutrophication of waters. Besides that, it will cause the formation of white foam which will block the entry of oxygen and light to the water eventually affects its fauna and flora [12].

The different characteristics of laundry wastewater originated from domestic, industrial and hospital sectors are shown in Table 1.

Table 1 Characteristic of laundry wastewater generated by domestic, industrial and hospital sectors [14]

Parameters	Domestic Laundry	Industrial Laundry	Hospital Laundry
pH	9.3-10	9-11	11.4-11.6
EC (µS/cm)	190-1400	640-3000	808-2000
TDS (mg/L)	400-6000	420	456-800
TSS (mg/L)	200-987	4-7000	66-71
TH(mg/L $CaCO_3$)	-	44	53-68
TA(mg/L $CaCO_3$)	83-200	128	302-375
TOC (mg/L)	8.0-35	71.5-11790	25-26
Phosphate (mg/L)	4-27.6	3.43	10.8-167
BOD_5 (mg/L)	48-1200	218-9810	44-50
COD (mg/L)	375-4155	80-212,000	477-876
Turbidity (NTU)	14-400	40-150	87.9

The commonly used methods to treat laundry wastewater are adsorption, chemical precipitation, coagulation, flotation and biological treatments [3]. Concentration of detergent

and surfactants in untreated domestic wastewater can be found in the range from 1 to 10mg/L [17]. Incomplete biodegradation of detergents and surfactants often results in the formation of toxic metabolites, which are more harmful than the parent compound, or of derivatives with relatively stable structure which may accumulate in the environment [18–20].

Advanced oxidation processes (AOPs) based on the generation of highly reactive radical oxidative species (ROS) such as hydroxyl radical ($\cdot OH$), but also other relative species such as hydrogen radical ($\cdot H$), hydrated electron (e_{aq}^-), singlet oxygen ($^1O_2$), hydroperoxyl radicals ($HO_2^{\cdot}$), superoxide radicals ($\cdot O_2^-$) and so forth [21,22] can be highly effective in the removal of recalcitrant pollutants and can serve as advanced methods for the treatment of laundry wastewater. The radical oxidative species (ROS) for different AOPs are shown in Table 2.

Table 2
Radical oxidative species (ROS) for different AOPs [21–23]

AOPs	Reactive species
Fenton oxidation	$\cdot OH$, $\cdot HO_2$
Ozonation	$\cdot OH$, $\cdot HO_2$, O_3, $\cdot O_2^-$, $\cdot O_3^-$, $\cdot HO_3^-$, H_2O_2, O^-
Photocatalysis oxidation	$\cdot OH$, h^+, $\cdot O_2^-$, e_{aq}^-, $\cdot HO_2$, $\cdot HOO$
Electrocatalysis oxidation	$\cdot OH$, e^-, H_2O_2
Wet air oxidation	$\cdot OH$, $\cdot HO_2$, H_2O_2
Ultrasonic oxidation	$\cdot OH$, $\cdot HO_2$ $\cdot H$, H_2O_2
Persulfate-based oxidation	$\cdot OH$, $\cdot SO_4^-$, $\cdot SO_5^-$, $^1O_2$
Radiation	$\cdot OH$, e_{aq}^-, $\cdot H$, H_2O_2

Different AOPs have been effectively used for the degradation of detergent surfactants or treatment of laundry wastewater such as ozone-based AOP [24–29], Fenton process [30–35], electrochemical oxidation [36–38], photolysis [39–44], photocatalysis [45–47], ultrasound [48–50] or in some cases a combination of these methods with conventional methods [51–55]. However, different sources and characteristics of the laundry wastewater may affect the performance or application steps of the various AOPs. In addition, challenges

related to excessive energy consumption, chemical input, corrosion of the catalyst supports, inefficient light utilization, limited mass transfer of ozone and the operating costs has often restricted their application in wastewater treatment [21,56]. For example, the main challenges of electrochemical AOPs (e.g., electro-Fenton, anodic oxidation and indirect oxidation) resides on the costs and life of electrodes and the high electrical energy consumption [38,57]. The same problems such as requirement of acidic pH [31,35,58] and sludge formation associated with the Fenton and photo-Fenton process can be found in electro-Fenton [37]. These processes are not suitable to treat laundry wastewater which has an alkaline nature due to the large pH adjustment needed [13,14,57].

The main challenges in photolytic and photocatalytic methods are the low UV-transmittance due to turbidity levels [56,59]. Sonolysis is highly energy intensive process unsuitable to treat large wastewater volumes and it is usually used as assisting method for other AOPs [60]. Hence, the efforts on the study of AOPs in terms of wastewater treatment and water reclamation research are still on-going with various improvement and strategies such as the development of heterogeneous catalytic ozonation [61–63], heterogeneous Fenton processes [64,65], plasmonic metal particle-incorporated TiO_2-SiO_2 composite as the solar photocatalyst for photocatalysis process [66] and novel application of industrial waste product such as red mud as catalyst support for the catalytic processes of AOPs [67]. All these efforts are constituting AOPs as the promising, efficient and environmental-friendly wastewater treatment methods.

Ozonation and ozone-based AOPs on the other hand, can be effectively and easily implemented for the treatment of recalcitrant pollutants in wastewater without generating a secondary waste [68]. Ozone is 13 times more soluble than oxygen in water [69] and it is widely applied to degrade many organic compounds causing odour, taste, colour and for disinfection purposes. Ozone is the powerful oxidant that can oxidize a lot of organic and

inorganic compounds [68]. Under alkaline conditions such as in laundry wastewater, the greater oxidation power of hydroxyl radicals generated from its decomposition results in a highly effective treatment. Thus, ozonation can be an excellent choice for the treatment of laundry wastewater. Some of the main challenges of ozonation are the limited mass transfer of ozone, short lifetime of the generated ozone and ROS and the relatively high energy consumption for ozone generation [61]. The efficiency of wastewater treatment by ozonation and ozone based AOPs can also be increased by combination with other oxidising agent such as hydrogen peroxide, UV irradiation and the catalyst [24,29,70].

This review focuses on the removal of surfactants as the main contaminants in laundry wastewater by ozonation processes and ozone-based processes. The ozonation processes treatment backgrounds and important parameters affecting the efficiency of ozonation processes in laundry detergent wastewater treatment are discussed. The perspectives for future studies and recommendations towards development of laundry detergent wastewater treatment by ozonation processes are presented.

2. Development of ozonation process technology and theoretical background

2.1 History of ozone discovery, production and properties of ozone

Ozone was first discovered by Christian Friedrich Schonbein (1799-1868) in 1840. He noticed that the electrolysis of water created an odorous gas. The odorous gas has a pungent smell described as “fresh air after a thunderstorm”. Schonbein gave a Greek-based name to the gas as ozone. The term was derived from a Greek word “ozein” which means the act of smelling [71]. Ozone is an unstable gas and having a short life span [72]. The chemical and physical properties of ozone are summarized in Table 3. Due to the rapid decomposition of ozone to oxygen and ozone cannot be accumulated, ozone has to be generated *in situ* and produced continuously when needed by ozone generator [73].

Ozone can be produced from pure oxygen or oxygen-containing air by various methods such as electrical (corona) discharge, photochemical (UV), radiochemical, chemical, and electrolytic methods. Ozone generation via corona discharge is the most common method that applied commercially. There are many different names taken for corona discharge such as silent discharge, dielectric barrier discharge, cold plasma, plasma block, cold plate discharge. They are all corona discharge using different techniques to produce ozone with an electrical charge across an air gap [74]. In this method, a dried, dust-free, oil free air or an oxygen-containing gas or pure oxygen is passing through the space of a high-energy electrical field (corona) between two electrodes separated by a dielectric material. One electrode is a grounded medium and the other is a dielectric one [75].

Table 3
Chemical and physical properties of ozone [76]

Chemical and Physical Properties	**Value**
Boiling Point,ºC	-111.9±0.3
Melting Point, ºC	-192.5±0.4
Critical Temperature at 54.6 atm, ºC	-12.1
Evaporation Heat, Kcal/mol	3.63
Dipole Moment, Debye	0.53
Vapor Pressure at -192.5 ºC , mmHg	0.00859
Gaseous Density, g/L	2.1415
Solid Density at 77.4 K, g/cm^3	1.728
Thermal Conductivity at -183 ºC, ºC/cm	0.000531
Dielectric Constant -183 ºC	4.74

2.2 Direct reaction mechanism of ozonation

Ozonation processes are pH dependent and can react with target substances through direct and indirect mechanisms. The direct pathway involves molecular ozone whereas indirect pathways involve hydroxyl radicals (OH·) and also other radical species [21,22]. Oxidation of target substances by molecular ozone take place at the acidic pH whereas

indirect pathways happens at alkaline pH [69,77]. The electrophilic character of ozone enables it to react efficiently with unsaturated functional group, amines, sulphides, and other reducing compounds.

On the other hand, the nucleophilic property of one oxygen atom of ozone causing it to react with carbonyl, carbon-nitrogen bond containing substances and strong Bronsted acids [76]. This is a slow and selective reaction with the rate constant in the range of 1.0 -10^6 M/s [69]. The molecular ozone reaction can be classified into 4 categories, namely oxidation-reduction reactions, dipolar cycloaddition reactions, electrophilic substitution reactions and nucleophilic addition [63,78].

2.2.1 Oxidation –reduction reaction

Ozone as one of the strongest oxidizing agents with the oxidation potential of 2.07V can oxidize various compound by oxidation-reduction reactions. The redox reactions are mostly occurred through electron transfer. They can be observed in the reaction between ozone and hydroperoxide ion or superoxide ($\cdot O_2^-$) as shown in (1) and (2) respectively.

$$O_3 + HO_2^- \rightarrow \cdot O_3^- + \cdot HO_2 \quad (1)$$

$$O_3 + \cdot O_2^- \rightarrow \cdot O_3^- + O_2 \quad (2)$$

2.2.2 Cycloaddition reaction

This is an addition reaction with the combination of 2 molecules to yield a third compound. Ozone is known to react with an unsaturated compound (with a carbon double bond or π electrons) and an electrophilic compound to form a new compound as shown in reaction (3) [63].

$$-C = C + XY \rightarrow -XC - CY - \quad (3)$$

Reaction of ozone with any olefinic compound and aromatics will form a cyclic intermediate called ozonide which will subsequently undergo series reactions such as abnormal ozonolysis and produces smaller molecules such as ketones, aldehydes, or carboxylic acids. The series of reaction (Criegee mechanism) are shown in the Fig. 1 with the three steps: i) formation of primary ozonide (or five-member ring); ii) zwitterion ion generation; iii) different reaction pathways of zwitterion and formation of final products.

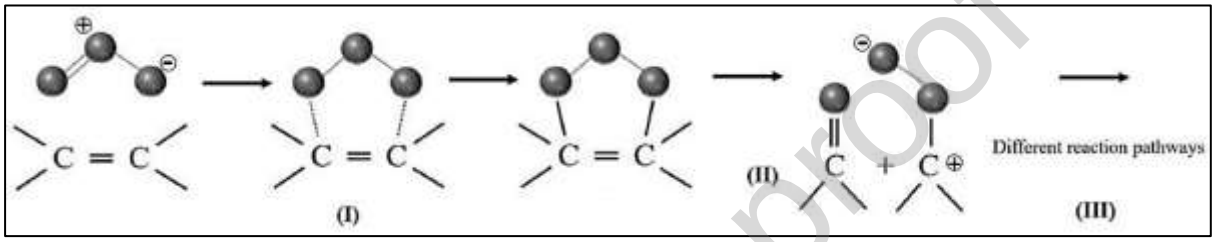

Fig. 1. Criegee mechanism of ozone reaction [63]

2.2.3 Electrophilic substitution reaction

Ozone as an electrophilic agent can attack the nucleophilic position of the organic substances and substitute one part of the molecule. The groups such as OH^-, NO_2^- and Cl^- in the aromatic molecule have strong influence on the reactivity of the aromatic ring with ozone [63].

2.2.4 Nucleophilic reaction

Ozone shows nucleophilic property because of its resonance structure. Nucleophilic addition takes place when there is shortage of electrons and particularly with the compound containing carbonyl or double and triple nitrogen bonds. This reaction only proposed in few cases in non-aqueous solution systems [63,78].

2.3 Indirect reaction mechanism of ozonation

The indirect reaction mechanism of ozonation is relevant to the treatment of laundry wastewater, therefore is presented here in detail. Ozone at pH > 7 begins to decomposes in water to form mainly hydroxyl radical (OH·) and other reactive radicals such as hydroperoxyl ($\cdot HO_2$), hydroxyl (OH·) and superoxide ($\cdot O_2^-$) at alkaline pH [77]. The hydroxyl radical which has an unpaired electron are highly unstable and immediately reacts with the other compounds to gain the missing electron [69]. Hydroxyl radicals (OH·) have higher oxidation potential (2.80V) than the ozone molecule (2.07V) alone as shown in Table 4.

Table 4
Oxidation agent and their oxidation potential [79]

Oxidizing Agent	Symbol	Oxidation Potential (V)	Relative Potential of Ozone
Fluorine	F_2	3.06	1.48
Hydroxyl Radical	·OH	2.80	1.35
Atomic Oxygen	O	2.42	1.17
Ozone	O_3	2.07	1.00
Hydrogen Peroxide	H_2O_2	1.77	0.85
Hydroperoxide radical	$\cdot HO_2$	1.70	0.82
Permanganate	MnO_4^-	1.67	0.81
Chlorine Dioxide	CO_2	1.50	0.72
Hypochlorus Acid	HClO	1.49	0.72
Chlorine Gas	Cl_2	1.09	0.66
Bromine Gas	Br_2	0.87	0.53
Iodine Gas	I_2	0.54	0.26
Oxygen Gas	O_2	0.40	0.19

The process of ozone decomposition in water to form hydroxyl radicals consists of a series of multiple reactions which include initiation, propagation and termination reactions [80]. There are two main initiation reactions which are pH dependent.

2.3.1 Initiation Steps

i) At acidic to neutral pH:

$$O_3 + OH^- \rightarrow \cdot O_2^- + \cdot HO_2 \qquad K_{i1}= 70\ M/s \qquad (4)$$

ii) At alkaline pH:

$O_3 + OH^- \rightarrow HO_2^- + O_2$ K_{i2}= 40 M/s (5)

$O_3 + HO_2^- \rightarrow \cdot HO_2 + \cdot O_3^-$ K_{i3}= 2.2 × 10^6 M/s (6)

Hydroxyl ions can react with ozone to form the superoxide anion ($\cdot O_2^-$) and hydroperoxyl radical ($\cdot HO_2$). The initiation reactions of (4) and (5) are usually slow and these will become the limited steps for the ozone decomposition. At alkaline pH, the hydroperoxide ion (HO_2^-) can reacts with ozone molecule much faster and generates a hydroperoxyl radical ($\cdot HO_2$) [80]. This hydroperoxyl radical has an acid-base equilibrium when value of Pk_a equals to 4.8. Above this value, it decomposes to form a superoxide anion ($\cdot O_2^-$) [77].

The chain reactions are propagated in the following steps.

2.3.2 Propagation Reaction:

$\cdot HO_2 \rightarrow \cdot O_2^- + H^+$ K_{1a}= 7.9 × 10^5 M/s (7)

$\cdot O_2^- + H^+ \rightarrow \cdot HO_2$ K_{1b}= 5 × 10^{10} M/s (8)

$O_3 + \cdot O_2^- \rightarrow \cdot O_3^- + O_2$ K_2= 1.6 × 10^9 M/s (9)

$\cdot O_3^- + H^+ \rightarrow \cdot HO_3$ K_3= 5.2 × 10^{10} M/s (10)

$\cdot HO_3 \rightarrow \cdot O_3^- + H^+$ K_4= 3.3 × 10^2 M/s (11)

$\cdot HO_3 \rightarrow \cdot OH + O_2$ K_5= 1.1 × 10^5 M/s (12)

$\cdot OH + O_3 \rightarrow \cdot HO_4$ K_6= 2 × 10^9 M/s (13)

$\cdot HO_4 \rightarrow \cdot HO_2 + O_2$ K_7= 2.8 × 10^4 M/s (14)

The superoxide anion ($\cdot O_2^-$) then reacts with ozone to form an ozonide anion($\cdot O_3^-$). It will immediately decompose via hydrogen trioxide ($\cdot HO_3$) to a hydroxyl radical ($\cdot OH$) in the step of (17). The hydroxyl radical ($\cdot OH$) can react with ozone in the steps of (13) and (14) to form ($\cdot HO_4$). The chain reaction can restart with the decomposition of ($\cdot HO_4$) into oxygen and a hydroperoxyl radical ($\cdot HO_2$). Some organic molecules, R that contain functional groups

that react with hydroxyl radical can also act as promoters and form organic radicals (·R) or organic peroxyl radicals (·ROO) with the presence of oxygen according to the reactions (15) and (16) respectively. The organic radicals can further react by re-enter into the propagation reaction (17-18). However, there are also possibilities that reaction of ozone with some organics or inorganics substances forming secondary radicals that do not produce superoxide radicals which are known as radical scavengers [77].

$$H_2R + \cdot OH \rightarrow \cdot HR + H_2O \quad (15)$$

$$\cdot HR + O_2 \rightarrow \cdot HRO_2 \quad (16)$$

$$\cdot HRO_2 \rightarrow R + \cdot HO_2 \quad (17)$$

$$\cdot HRO_2 \rightarrow RO + \cdot OH \quad (18)$$

2.3.3 Termination Step:

$$\cdot OH + \cdot HO_2 \rightarrow O_2 + H_2O \qquad K_8 = 3.7 \times 10^{10}\ M/s \quad (19)$$

$$\cdot OH + HCO_3^- \rightarrow OH^- + \cdot HCO_3 \qquad K_9 = 1.5 \times 10^{7}\ M/s \quad (20)$$

$$\cdot OH + CO_3^{2-} \rightarrow OH^- + CO_3^- \qquad K_{10} = 4.2 \times 10^{8}\ M/s \quad (21)$$

$$\cdot OH + H_2PO_4^- \rightarrow OH^- + H_2PO_4 \qquad K_{11} < 10^{5}\ M/s \quad (22)$$

$$\cdot OH + HPO_4^{2-} \rightarrow OH^- + H_2PO_4 \qquad K_{12} < 10^{7}\ M/s \quad (23)$$

The chain reaction can be terminated by reaction of two radicals as shown in the step (19) [77]. The chain reaction also can be terminated by radical catchers or scavengers such as bicarbonate (HCO_3^-), carbonate (CO_3^{2-}), dihydrogen phosphate ion ($H_2PO_4^-$) and hydrogen phosphate ion (HPO_4^{2-}) through the reaction of (20) to (23) [63]. Presence of tert-butanol, p-chlorobenzoate, and humic substances also act as hydroxyl radical scavengers. Hydrogen peroxide generation is also suggested under alkaline condition by the reaction (24). However, hydrogen peroxide is an initiator and promoter of ozone decomposition which is known as

peroxone process ($O_3 + H_2O_2$). The combination of the reactions (20) and (7-14) give the overall of reaction (25) which shows that three ozone molecules will produce two hydroxyl radicals [77].

$$HO_2^- + H^+ \rightarrow H_2O_2 \qquad K_{13}= 5 \times 10^{10}\ M/s \qquad (24)$$

$$O_3 + OH^- + H^+ \rightarrow 2 \cdot OH + 4O_2 \qquad (25)$$

The hydroxyl radical generated can initiate the oxidative destruction of organic contaminants present in wastewater by the processes such as radical addition, hydrogen abstraction, electron transfer and radical combination [21].

2.3.4 Hydrogen atom abstraction (Dehydrogenation)

A hydrogen atom can be removed by the hydroxyl radical from the organic compound (C-H bonds) to form a radical organic compound (·R) and water. The radical compound can initiate a chain reaction by reacting with oxygen to form a peroxyl radical (·ROO) which can react with another organic compound and so on. The reactions are shown in (26-27).

$$R + \cdot OH \rightarrow \cdot R + H_2O \qquad (26)$$

$$\cdot R + O_2 \rightarrow \cdot ROO \qquad (27)$$

2.3.5 Radical addition

The addition of hydroxyl radical to an unsaturated aliphatic or aromatic organic compound resulted in the production of a radical organic compound that can be oxidised further by oxygen or ferrous ion to form stable oxidized end products as shown in (28).

$$R + \cdot OH \rightarrow \cdot R - OH \qquad (28)$$

2.3.6 Electron transfer

The electron transfer is through the oxidation–reduction process which causes the formation of ions of a higher valence. Oxidation of a monovalent negative ion will result in the formation of an atom or a free radical (29-30).

$$R^n + \cdot OH \rightarrow R^{n-1} + OH^- \quad (29)$$

$$RX + \cdot OH \rightarrow \cdot RX^+ + OH^- \quad (30)$$

2.3.7 Radical combination

Two radicals can combine to form a stable product as shown in (31-32)

$$\cdot OH + \cdot OH \rightarrow H_2O_2 \quad (31)$$

$$\cdot R + \cdot R \rightarrow R - R \quad (32)$$

Ozonation processes can react with target substances through direct or indirect pathways. Indirect reactions of ozone are more powerful than direct pathway of ozonation due to the higher oxidation potential of the hydroxyl radical compared to the molecular ozone. Both pathways should always be considered when using ozonation processes as the treatment method for wastewater [77]. In the direct reaction of molecular ozone, molecular ozone selectively attacks molecules containing unsaturated bonds such as nitro-aromatic compounds, surfactants, pesticides and dyes leading to the formation of saturated compounds including aldehydes, ketones, and carboxylic acids [81–86]. However, these intermediates are resistant to ozone attack and accumulated in the solution. The extension of the degradation can be further improved by hydroxyl radical attacks [87].

2.4 Ozone-based AOPs

In addition to the individual ozonation processes, ozone-based AOPs which focused on the enhancement of the efficiency of ozonation by combination with other oxidising agent such as O_3/hydrogen peroxide, catalytic ozonation, ozone/UV and others are also receiving

much attention. In this combined processes, the oxidising capability is increased by higher production of hydroxyl radicals and reduction of ozone dosage [81]. The ozone based AOPs are discussed in the following sections.

2.4.1 Ozone/hydrogen peroxide

Combination of ozone and hydrogen peroxide is also known as peroxone process with the goal to expedite the transformation ozone to hydroxyl radicals. Peroxone process is probably the best studied and implemented AOP. Ozone decomposition to hydroxyl radicals is accelerated with the low concentration of hydrogen peroxide (10^{-5} to 10^{-4} M) [80]. It is through the reaction between ionic form of hydrogen peroxide (hydroperoxide ion, HO_2^-) and ozone to form hydroxyl radicals according to the reaction (33-35). The optimum stoichiometry for this process is in the ratio of 1 mole of hydrogen peroxide react with 2 mole of ozone [59]. The efficiency and reaction rate of peroxone is higher than the ozonation alone besides reduce the formation of toxic-by-product such as bromate [88]. However, excessive amount of hydrogen peroxide will act as radical scavenger and residual of hydrogen peroxide is harmful to the environment [69].

$$H_2O_2 \leftrightarrow H^+ + HO_2^- \quad (33)$$

$$O_3 + HO_2^- \rightarrow \cdot OH + O_2 + \cdot O_2^- \quad (34)$$

$$H_2O_2 + 2O_3 \rightarrow 3O_2 + 2 \cdot OH \quad (35)$$

2.4.2 Ozone/UV

The principle of ozone/UV is based on the coupling between ultraviolet radiation and ozone. Ozone undergoes photolysis reaction to form hydrogen peroxide or atomic oxygen radical ($\cdot O$) as shown in reactions (36) and (37) respectively [78]. The hydroxyl radicals are formed by reaction of $\cdot O$ and water. Alternatively, hydrogen peroxide molecules may

undergo ultraviolet irradiation to form hydroxyl radicals too according to the reaction (38) [79]. The practical application of this system is restrained due to low hydroxyl radical yield [89] and high energy demands for the UV lamps and ozone generator [56].

$$O_3 + H_2O + hv \rightarrow H_2O_2 + O_2 \quad (36)$$

$$O_3 + hv \rightarrow O_2 + \cdot O \quad (37)$$

$$\cdot O + H_2O \rightarrow 2 \cdot OH \quad (38)$$

2.4.3 Ozone/UV/hydrogen peroxide

This system consists of the three components that combined to increase the generation rate of hydroxyl radicals. However, the parameters such as operating pH, ozone dosage, hydrogen peroxide dosage and the light intensity need to be optimized in order to achieved high mineralisation of target pollutants and energy efficiency [90]. Comparative evaluations of the processes, UV, UV/O_3, UV/H_2O_2 and UV/H_2O_2/O_3, have been studied for the degradation of organic pollutants such as Reactive Red 45 (organic dye) after one hour treatment, with the UV/H_2O_2/O_3 process showing the highest mineralization extent and the efficiency of the processes follows the order: UV< UV/H_2O_2< UV/O_3< UV/H_2O_2/O_3 [91]. However, the high cost of this system in terms of ozone generation, UV light and hydrogen peroxide preparation limits the broader use of the system.

2.4.4 Catalytic ozonation

The ozonation technology can be further enhanced by the addition of catalyst and the system is known as catalytic ozonation [92]. The application of various catalysts on the course of ozonation will produce more reactive hydroxyl radicals with lower ozone demands compared to ozonation alone [56]. The catalytic ozonation are classified into homogenous and heterogenous types according to the water solubility of the catalyst. Homogenous

catalytic ozonation employs metal ions as the catalyst whereas heterogenous catalytic ozonation uses metal oxide or metal oxide (or metal) on supports as the main catalysts [93].

Catalytic ozonation has shown better performance than single ozonation in achieving higher degree of mineralisation for some intermediate products such as aldehydes, ketones and carboxylic acids [81]. Besides having better performance in mineralisation of organic pollutants, the catalytic ozonation also reduce the operation cost since it does not need other energy cost for UV generation or cost for pH adjustment as its effectiveness in wide range of pH values [63]. The varieties of homogenous and heterogeneous catalysts are shown in the Fig. 2.

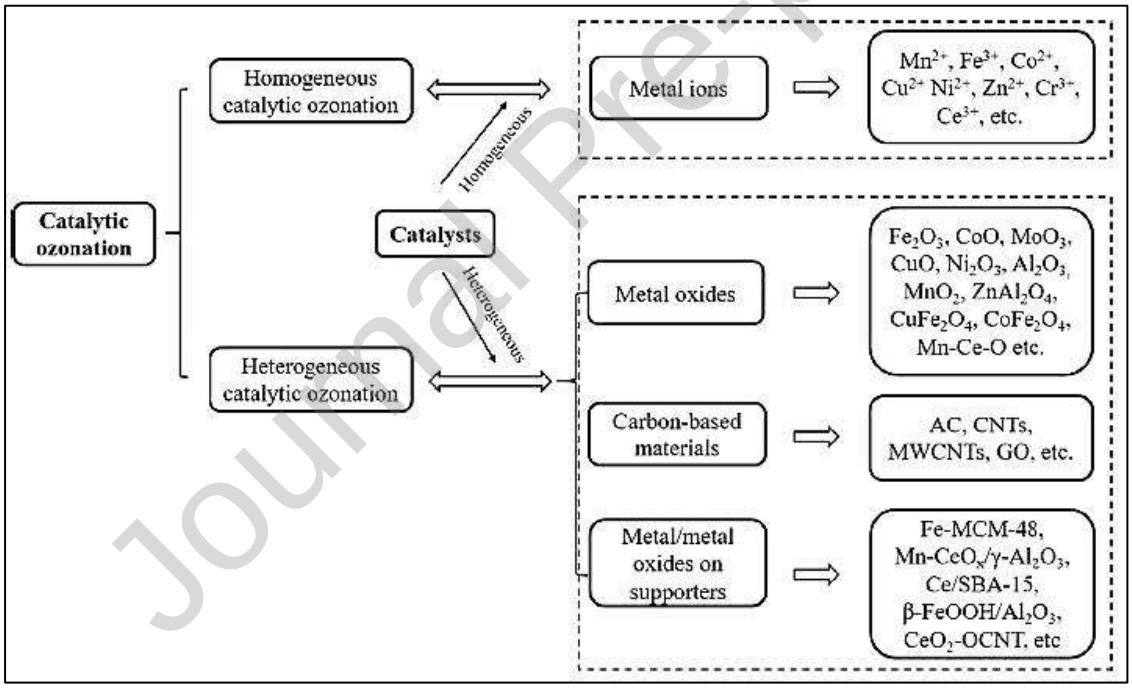

Fig. 2. Examples of homogeneous and heterogeneous catalysts [63]

The liquid catalysts, especially the transition metal ions such as cobalt (II), nickel (II), iron (II), copper (II), manganese (II), zinc (II) and so forth were used for the process of homogeneous catalytic ozonation for the removal of organic compounds that are hard to be removed by ozonation alone [94–99]. The nature or characteristics of the transition metals

determine the ozone consumption, reaction rate and selectivity [100]. There are two main pathways of homogenous catalyst mechanisms. The first one involved enhancement of the decomposition of ozone to generate more hydroxyl radicals. The other pathway is combination of metal ions and organic pollutants to form intermediates or complexes, which are then oxidized by ozone and other oxidizing species [63,69].

However, the use of metal ions in solutions can cause secondary pollution because the catalysts will need to be removed after the oxidation of the organic compounds. The particles are unstable and accumulated to form large particles [81]. Hence, the applications of homogenous catalytic ozonation need to accompany by a technique of removing the metal ion from the treated effluents, which will increase the costs of water treatment. In addition, the toxicity of some metals may limit the application homogenous catalytic ozonation [100]. The problems are overcame by the usage of solid catalysts or heterogeneous catalysts such as metal oxides, carbon based materials and metal supported catalysts which are more stable and efficient as shown in Fig. 2 [63].

The efficiency of heterogenous catalytic ozonation depends on the solution pH, chemical structure of the reactants and also the properties of the catalyst such as surface area, pore volume, porosity and mechanical strength [92]. The common metal oxides that studied and applied in heterogenous catalytic ozonation are such as manganese oxides (MnO_2), aluminium oxides (Al_2O_3), titanium oxides (TiO_2), zinc oxides (ZnO) and so forth [96,101–103]. The carbon based materials were also being studied for catalytic ozonation such as activated carbon, multi-walled carbon nanotube [104,105]. The solid catalysts are easy to be segregated and have longer time of catalytic activity. It is a promising technology for water and wastewater treatment especially for the degradation of various persistent and toxic organic pollutants such as phenols, pesticides, dyes, pharmaceuticals, nitrobenzenes,

surfactants and also the treatment of synthetic and industrial effluents [63,92]. However, inconsistent mechanisms were reported by different research groups and there are still undefined for the detailed mechanism of catalytic ozonation [63,106,107].

The mechanism of the heterogenous catalytic ozonation can be complex since many reaction steps are three phases (gaseous (ozone gas), liquid (water) and solid (catalyst). There are three possible mechanisms of heterogeneous catalytic ozonation (Fig.3.). In the first case, ozone is adsorbed onto the catalyst surface and decomposes into free radicals. The radicals react with the organic molecules and oxidized products are formed. Secondly, organic molecules are adsorbed on the surface of the catalyst and then attacked by ozone molecules or other species to form oxidized products. In the third case, both ozone and organic molecules adsorb on the catalyst surface and then react [63].

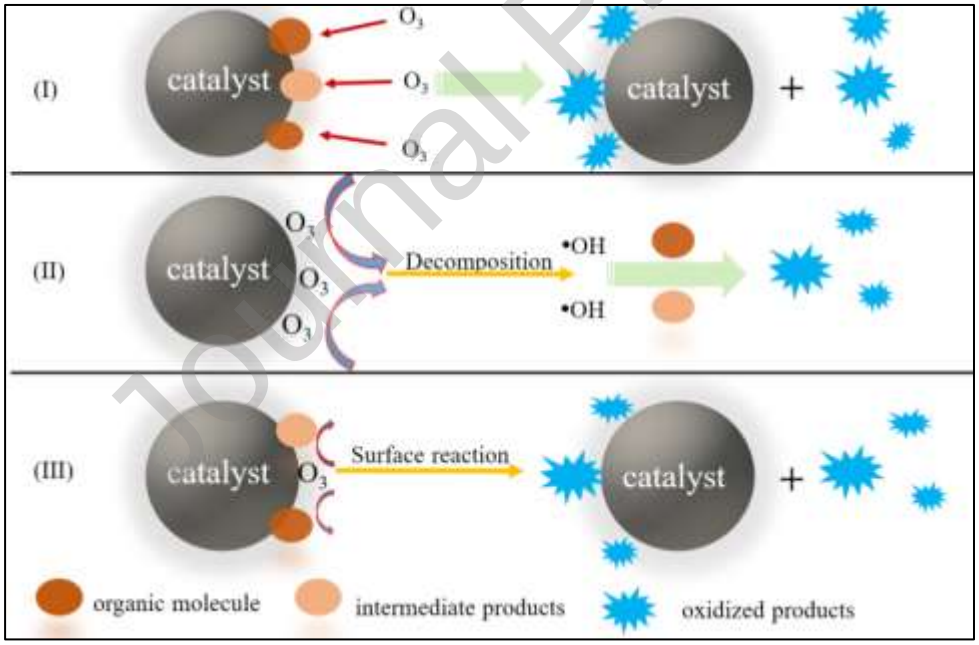

Fig.3. The Mechanisms of heterogeneous catalytic ozonation [63]

2.4.5 Photocatalytic ozonation

Photocatalysis and ozonation are combined to achieve higher extent of mineralisation of pollutant. The common catalyst used for photocatalytic ozonation reactions is titanium dioxide (TiO_2) and TiO_2-based materials. Titanium dioxide catalyst is widely used because of its chemical stability, low cost, non-toxicity and optical properties. However, there are some limitations of its application such as: i) its photocatalytic activity can only initiated by ultraviolet light due to its wide band gap (3.2eV), ii) low quantum yield and photocatalytic activity which caused by the relatively high rate of electron/hole recombination and iii) the photocatalyst recovery and its reusing requires several steps that increase the operational costs [108]. The other non-TiO_2-based catalysts are such as nickel-zinc ferrite magnetic nanoparticle (NZFMN), copper ferrite nanoparticle (CFN), multiwalled carbon nanotube (MWCNT), bismuth oxide (Bi_2O_3) nano rods and so forth [109].

In the photocatalytic ozonation reaction, photocatalyst is irradiated with a photon of energy greater than their band gap energy so that electron/hole pairs are formed. Ozone molecules act as the electron trappers, lead to the formation of ozonide ion radicals $(\cdot O_3^-)$ and then leads to the generation of hydroxyl radicals according to the reactions (39) and (40) [110].

$$O_3 + e^- \rightarrow \cdot O_3^- \quad (39)$$

$$\cdot O_3^- + H_2O \rightarrow O_2 + OH^- + \cdot OH \quad (40)$$

There are various operation parameters that will affect photocatalytic ozonation such as pH, reaction temperature, ozone dosage, light intensity, natural and concentration of the substrate and properties of the photocatalyst [111]. The degradation efficiency of photocatalytic ozonation are presented and reviewed in many studies. However, most of them are employed in lab-scale and few of them have been performed on real wastewater treatment [110]. Photocatalytic ozonation is considered as more costly treatment method and its use for

the removal of biodegradable pollutants from water is not economically feasible. However, it can be applied to mineralise poorly-biodegradable organic compounds or enhance the biodegradability of organic contaminants such as pharmaceutical compounds, surfactants, detergents, colouring matters, pesticides, saturated aliphatic carboxylic acids and so forth [112].

2.5 Summary of ozonation and ozone-based processes

Overall, ozonation processes can be categorised into non-catalytic ozonation and catalytic ozonation as shown in Fig. 4.

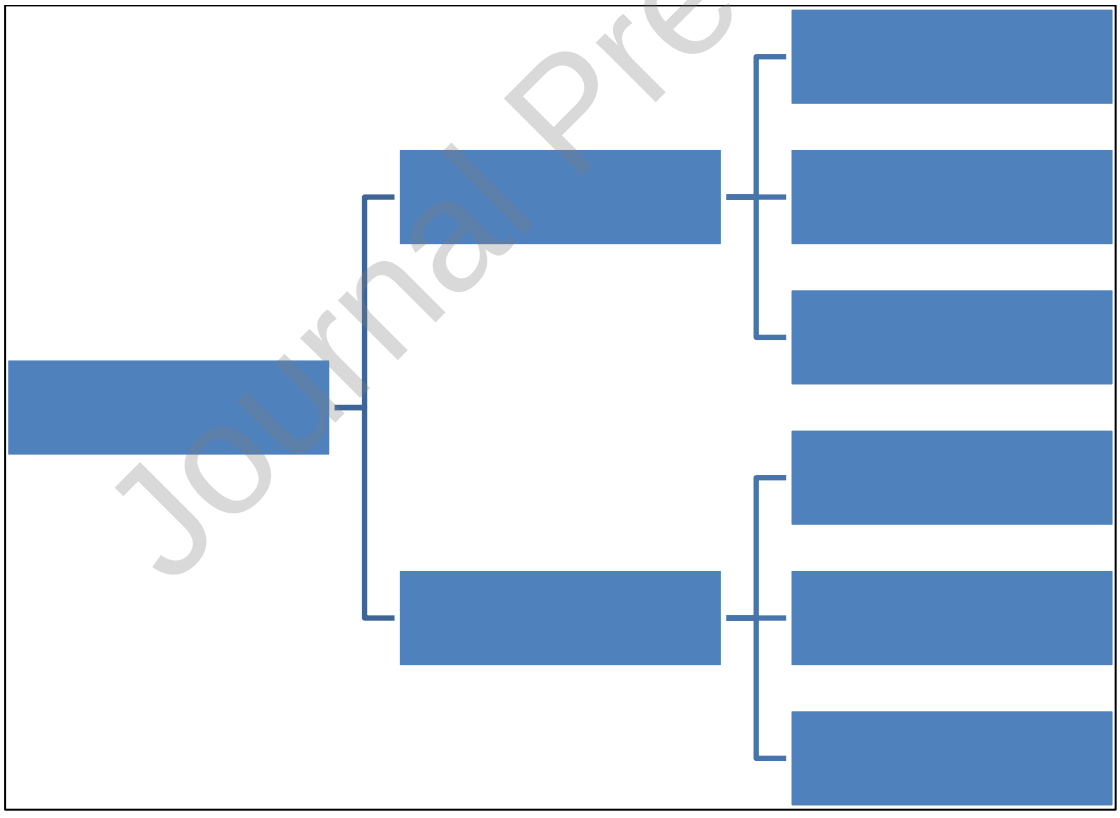

Fig. 4 Schematic diagram of various ozonation processes.

Ozonation processes have been extensively applied in wastewater treatment due to the strong oxidation ability based on the direct reaction of molecular ozone and the production of

hydroxyl free radicals. However, complete mineralisation of the contaminants by molecular ozone is not economical due to the high ozone generation cost and other factors such as short half-life ozone, poor solubility of ozone in water, low utilization of ozone during ozonation and limited mass transfer of ozone [84,92,113].

The indirect free radical mechanism of ozonation which required an elevated pH to produce hydroxyl radicals which is short lived and extremely potent oxidizing agent can degrade and mineralize the pollutants unselectively. This indirect ozonation process is capable to remove 90% of the contaminant from water and it is widely used for drinking water treatment because of its oxidation potential and disinfectant properties [114]. However, the indirect ozonation process is suppressed by presence of some radical scavengers that may exist in natural water bodies or wastewater such as, bicarbonate ion (HCO_3^-), carbonate ion (CO_3^{2-}), dihydrogen phosphate ion ($H_2PO_4^-$) and hydrogen phosphate ion (HPO_4^{2-}) [115].

Therefore, ozone based AOPs are widely investigated in order to overcome some of the drawbacks of conventional ozonation process. Among them, catalytic ozonation and peroxone process have received much attention for its ability to remove organic matter from the effluent completely [92]. It is foreseen that in the near future there will be increasing studies in ozonation and ozone-based processes due to its technical evaluation as shown in the following strengths, weaknesses, opportunities and threats (SWOT) diagram (**Fig. 5**.).

Fig. 5 The SWOT analysis for ozone technologies [116]

3. Conventional ozonation treatment of laundry wastewater and the operation parameters

The treatment of laundry wastewater is very challenging because of the negative impact to the environment, increasing usage of detergent with intense population growth and lacking of treatment infrastructure improvements [10,19]. Conventional ozonation or single ozonation treatment refers to the direct and indirect reaction (AOP) of molecular ozone in wastewater treatment. The efficiency of ozonation treatment depends on various process parameters, such as temperature, solution pH, initial concentration of pollutant, ozone dosage, mixing conditions and time of treatment. Laboratory scale studies are done on the synthetic laundry detergent wastewater treatment (using model surfactants) or on real effluents such as domestic laundry, hospital laundry, textile industry laundry wastewater. In this review, we distinguish 3 major types of surfactants namely anionic, non-ionic and cationic surfactants

and also their metabolites that formed from biodegradation processes. The impacts of the process parameters and further details are discussed in the following sections.

3.1 Effect of Temperature

Ozone solubility decreases with temperature, thus a decrease in dissolved ozone concentration usually results in lower degradation of target pollutants. The ozonation/UV treatment of synthetic laundry wastewater containing potassium laurate and alkyl benzene sulfonate (LAS) measured by total organic carbon (TOC) reduction has been reported almost the same at 10 and 20 ºC, but significantly decreased at 40 ºC [117]. The results suggested that, the treatment efficiency is not improved by the higher temperature due to the limited ozone solubility in the solution. However, another study on ozonation of LAS in the range of 25 to 55 ºC reported that higher temperature causes higher degradation of LAS, although the ozone concentration in the aqueous solution decreased at higher temperature [118].

In another study, the removal of the non-ionic surfactant, nonylphenol (NP) was improved when the temperature was increased from 22 to 30ºC with a reaction time of 20 minutes [119]. The degradation of sodium dodecyl sulphate (SDS) with ozone in aqueous solution, on the other hand, in the temperature range from 20 to 50ºC was influenced by the presence of sodium nitrate ($NaNO_3$) [120]. The rate of the SDS decomposition increased at higher temperature in the absence of $NaNO_3$, but decreased in the presence of high concentration of sodium nitrate above 100g/L. This was explained by the competition of $NaNO_3$ with SDS for accessible ozone in the direct ozonation. The ozone solubility and ozone mass transfer were also affected by the presence of salt. However, under alkaline conditions, in the presence of sodium hydroxide (NaOH), the competition from $NaNO_3$ for ozone molecules did not take place due to the predominant indirect ozonation mechanism that

predominates at elevated pH. Thus, the decomposition of SDS increased again as the temperature was raised up to 50ºC.

Although the ozonation process is limited by the solubility of ozone in water, a temperature increase implies faster reaction rate of degradation [121]. In general, most of the ozonation experiments are carried out at ambient temperature and manage to achieve high degradation of pollutants. Higher temperature may increase the reaction kinetics and may improve the mass transfer between ozone gas and the solution [119], although overall ozone solubility decreases. More studies are needed to investigate the effect of temperature on the mass transfer of ozone in solution. In order to effectively transfer the ozone from gas phase to liquid phase or for the mass transfer improvement, the use of metals, solid catalyst as in catalytic ozonation [61,63], bubble column or gas diffusers [88], production of ozone nano bubbles [69], ultrasonic catalytic microbubbles ozonation process [60] and microbubbles by oscillatory flow in a baffled column [122] are being studied.

Overall, the performance under indirect ozonation are better than under direct ozonation as the latter is a highly selective process. In addition, the presence of other organic compounds in the wastewater other than the target compound will also affect the efficiency of the ozonation process, particularly in the presence of radical scavengers such as dissolved organic carbon, nitrite, bicarbonate, carbonate, dihydrogen phosphate ion and hydrogen phosphate ion (HPO_4^{2-}) that are naturally present in wastewater [63,88]. Therefore, characterisation of the wastewater is needed before applying the ozonation process. A pre-treatment steps such as filtration, coagulation and biological treatment can also be applied prior to ozonation process in order to remove the deleterious oxidants [88]. A techno-economic optimization process is needed to select the most efficient treatment methods. There are no details on the cationic surfactant degradation by ozone in terms of temperature

in the literature. Table 5 summarizes the effect of temperature on the ozonation treatment of surfactant wastewater.

Table 5
Effect of temperature on the efficiency of the ozonation treatment

Target Compound(s)	Temperature studied	Results/ Remarks	References
Anionic Surfactant			
Linear alkyl benzene sulfonate (LAS)	T = 25, 35, 45 and 55ºC with 15ppm of LAS and duration of 10 min.	The concentration of LAS decrease with an increase in temperature from 25 to 55ºC.	[118]
Sodium Dodecyl Sulfate (SDS)	T = 20 and 50ºC with 100ppm of SDS	The SDS degradation rate increase with temperature increase from 20 to 50 ºC. With the presence of sodium nitrate in the solution, the SDS decomposition decreased.	[120]
LAS and Potassium Laurate	T = 10, 20 and 40ºC in the Ozone/UV system	The TOC deduction at 10ºC is almost the same for 20ºC. Higher temperature of 40ºC did not improve the efficiency due to limited solubility of ozone	[123]
Non-ionic surfactants			
Nonylphenol (NP)	T = 22 and 30ºC with 200ppm initial concentration and reaction time of 20 min.	NP degradation increase with the temperature increase	[119]

3.2 Effect of pH Solution

The pH of the wastewater is a key factor impacting the effectiveness of the ozonation treatment process because it determines the type of ozonation mechanism such as direct or indirect reaction [73]. At higher pH values, the formation of non-selective hydroxyl radicals is favoured. On the other hand, at lower pH values, the direct electrophilic attack by molecular ozone is predominant [124]. The initial pH for domestic, industrial and hospital laundry wastewater can reach values in the range 9 to 12 respectively [14], thus the prevalent mechanism of ozonation of these laundry wastewater is by indirect ozonation process and hydroxyl radicals.

In general, a pH decrease is observed during the ozonation of laundry wastewater containing surfactants thus the mechanism of reaction may switch from indirect to direct during the treatment. For example, the ozonation treatment of anionic and non-ionic surfactants in aqueous solution at pH of 7 resulted in a final acidic pH of 4.5 [125]. Same observation has been reported for the ozonation of anionic surfactants such as sodium dodecylbenzenesulfonate (SDBS) and sodium lauryl sulphate (SLS) [126]. Both surfactants degraded at similar rates and achieved 90% conversion and the pH decreased from 7 to 2.6 for the case of SLS. The acidic conditions of the final solution due to the presence of organic acids resulting from the ozonation process. The analysis and identification of the final products are needed in order to clarify the degradation mechanism of the target pollutant by ozonation process. Besides that, anionic surfactants with aromatic rings such as linear alkylbenzene sulfonates, or non-ionic surfactants which is alkylpolyglucoside (APG) exhibits lower toxicity after ozonation [125]. This is an added advantage of applying ozonation in laundry wastewater treatment where the toxicity of wastewater is reduced.

The conversion of two commercial anionic surfactants, namely sodium dodecylsulfate (SDS) and tetraethylammonium dodecylbenzenesulfonate (DBS) after 30 minutes of reaction at basic pH of 9 (70%) was significantly faster than natural pH of 6.2 (20%) due to generation of stronger hydroxyl radicals than molecular ozone alone [127]. Similarly, the removal if SDBS by ozonation was especially fast at pH 10 rather than pH 7 or 2 due to faster reaction of SDBS with hydroxyl radicals [128].

The degradation of non-ionic surfactants such as octylphenol (OP) and nonylphenol (NP) through the direct and indirect reaction of ozonation was also examined at the pH range of 2 to 9 [129,130]. The reaction rate constant by molecular ozone for OP and NP were determined at pH 2 were 4.33×10^4 and 3.90×10^4 $M^{-1}S^{-1}$ respectively. In contrast, at pH 6 to 9,

the second order rate constants of the treatment with hydroxyl radicals were found to be $1.4x10^{10}$ and $1.10x10^{10}$ $M^{-1}S^{-1}$ for OP and NP respectively. The rate of ozone consumption increased with increasing pH as a result of ozone decomposition to hydroxyl radical. Similarly, the removal of nonylphenol (NP) increased as pH was varied from 4.86 to 10.34 [119].

Another study investigated the ozonation treatment of the two important cationic surfactants which are benzyl-dimethyl-hexadecylammonium-chloride (16-BAC) and benzyl-dimethyl-stearylammonium-chloride (18-BAC) at pH in the range between 5 and 6. The removal efficiency measured in terms of TOC reached 25 and 50% after 1.5 hours of ozonation respectively [131].

The performance of ozonation also depends on the molecular structure of pollutants. Electron-rich molecules easily react with molecular ozone. For example, the presence of a benzene ring in the surfactant structure and a long chain type of surfactant are harder to be degraded compared to molecules with a shorter chain and without benzene ring [132]. In this cases, indirect ozonation and other ozone-based AOPs which involves the generation of hydroxyl radicals are needed to achieve effective treatment.

The above studies indicate that higher removal efficiency of surfactants by ozonation can be attained at alkaline pH mediated by the strong hydroxyl radical, although, the optimal pH required for the maximum degradation of a pollutant need to be optimized in order to avoid excess utilisation of reagents. The data of the effect of pH solution on the efficiency of ozonation treatment are tabulated on Table 6.

Table 6 Effect of pH solution on the efficiency of the ozonation treatment

Target Compound (s)	pH studied	Results/ Remarks	References
Anionic Surfactant			
Sodium dodecyl benzene sulfate (SDBS)	pH 2,7 and 10 at T=20 ºC and initial concentration of 15ppm	SDBS degradation increase with the increasing in pH and Optimum pH is 10.	[128]
Sodium dodecylsulfate (SDS)	pH 6.2 and 9	Surfactant removal (MBAS method) of 20% and 70% for pH 6.2 and 9 respectively after 30 min.	[127]
Tetraethylammonium Dodecylbenzenesulfonate (DBS)	pH 6 and 9	Surfactant removal (MBAS method) of 50% and above 80% for pH 6 and 9 respectively after 30 min.	
Non-ionic surfactants			
Octylphenol (OP) and Nonylphenol (NP)	pH 2	The reaction rate constant by molecular ozone for OP and NP are $4.33x10^4$ and $3.90x\ 10^4\ M^{-1}S^{-1}$ respectively.	[129]
	pH in the range between 6 and 9	The second order rate constants of the treatment with hydroxyl radicals were found to be $1.4x10^{10}$ and $1.10x10^{10}\ M^{-1}S^{-1}$ for OP and NP respectively.	[130]
Nonylphenol (NP)	pH 4.86, 7.03 and 10.34 with 200ppm initial concentration	NP removal rate increased with the increase in pH.	[119]
Cationic surfactants			
Benzyl-dimethyl-hexadecylammonium-chloride (16-BAC) and Benzyl-dimethyl-stearylammonium-chloride (18-BAC)	pH in the range between 5 and 6	TOC removal between 25% and 50% after 1.5 hours of ozonation.	[131]

3.3 Effect of initial concentration

The pollutant removal efficiency by ozonation decreases with an increase in the initial concentration of the pollutants for a constant ozone dosage. This is due to higher rate of ozone consumption required at higher pollutant load. Jurado-alameda et al. [118] conducted the ozonation treatment for LAS with the initial concentration of 10, 15 and 25ppm. The efficiency of LAS ozonation was higher at lower LAS concentrations. Same observation has been reported for the degradation of a non-ionic surfactant (Sannonic SS-90) with the initial concentration of 5 to 30 mg/L. The decomposition of the surfactant decreased with the increase in the initial surfactant concentration. Besides that, TOC reduction was slower than

the reduction of surfactant concentration. This may due to the formation of intermediate oxidation by-products [133].

Another study investigated the ozonation treatment of non-ionic surfactant (Oxidet DM-4) with the concentrations of 50 and 100mg/L [134]. Mineralization degree of the surfactant was around 6% for the 50mg/L of Oxidet DM-4 (50% of ozone production) whereas 38% of mineralization achieved for 100mg/L concentration (100% of ozone production). The above results shown that higher rate of ozone consumption required for higher initial concentration of pollutants.

Besides that, different structure of the compounds will affect the effectiveness of the direct molecular ozone treatment process. Ozone as the electrophilic agent react selectively with the compounds containing electron-rich functional groups (e.g. unsaturated bonds, activated aromatic ring). On the other hand, the aromatic compounds containing electron-withdrawing groups (e.g. -Cl, -COOH, -NO_2, -CN) have low reactivity with ozone [79,135]. However, in the ozonation treatment of nonylphenoxy carboxylic acid (NP1EC) that containing carboxyl group, almost all fractions of NP1EC could be degraded within 4 to 6 minutes when the initial concentration was between 0.4 and 1.0mg/L [136]. This was explained by the electrophilic substitution reaction of molecular ozone which involved the substitution of hydrogen atom (-H) from the meta-position of the surfactants [63,79].

The average concentration of LAS in municipal wastewater treatment plants treating domestic wastewater range from 1 to 10mg/L. In the presence of higher LAS concentrations, biological treatment of wastewater becomes inefficient requiring long retention time to complete remove LAS, moreover foaming becomes a problem and the pH self-regulation capacity of the wastewater decreases. Hence, ozone has been proposed as an alternative agent with its high oxidizing power to address the problems mentioned above. The study done by

Beltran et al.[128] using real domestic wastewater and synthetic wastewater with 15mg/L of SDBS show that ozonation process alters the molecular structures of the organic matter initially presented in the wastewater and improve the biodegradability of the organic pollutants. This is also in accordance to the review done by Alvares et al. [124] which suggesting that ozonation can be used as pretreatment for biological treatment.

Amat et al. [127] evaluated the use of ozonation treatment for the degradation of dodecylbenzenesulfonate (DBS) and sodium dodecylsulfate (SDS) surfactants at the initial concentration of 20mg/L. For such a high concentration of surfactant of wastewater, it is preferable to employ the indirect ozonation treatment with generation of stronger hydroxyl radicals rather than direct reaction of molecular ozone alone. Their results shown that 70 to 80% of surfactant removal achieved in reaction time of 30 minutes [127].

Higher consumption of ozone is needed either for direct or indirect reaction of ozonation treatment in the degradation of high concentration of pollutants. Therefore, in order to overcome these limitations, optimisation of the ozone dosage and determination of the ratio of ozone molecules or hydroxyl radical's utilization with the target pollutants need to be performed. Table 7 shows the effect of initial concentration of surfactant in the detergent wastewater on the effectiveness ozonation treatment.

Table 7 Effect of initial concentration on the efficiency of the ozonation treatment

Target Compound (s)	Initial concentration	Results/ Remarks	References
Anionic Surfactant			
DBS and SDS	20ppm in reaction time of 30 min.	SDS removal (MBAS method) of 20% and 70% for pH 6.2 and 9 respectively. DBS removal (MBAS method) of 50% and above 80% for pH 6 and 9 respectively.	[127]
LAS	10, 15 and 15 ppm in reaction time of 10 minutes and temperature of 35ºC	LAS degradation rate decreased with the increase in the solution concentration	[118]
SDBS in synthetic and domestic wastewater	15ppm	Biodegradability rate increased.	[128]
Non-ionic surfactants			
Nonylphenol (NP), nonylphenol monoethoxylate (NP1EO) and nonylphenoxy carboxylic acid (NP1EC)	0.4 and 1.0ppm in reaction time of 4 – 6 min.	75-80% of NP removal. Ozonation was not so efficient for the breakdown of NP1EO. Almost all fractions of NP1EC degraded.	[136]
Polyoxyethylene Alkyl Ether (Sannonic SS-90)	5-30mg/L	20mg/L was completely decompose after 60 min. 95% of TOC mineralisation was accomplished after 160 min. The mineralisation rate of the surfactant decreased with the increase in the initial surfactants concentration	[133]
Oxidet DM-4	50 and 100 ppm in reaction time of 30 min.	Degradation of 5.67% for 50mg/L of Oxidet DM-4 with the 50% of ozone production whereas 37.64% of degradation achieved for 100mg/L with the 100% of ozone production	[134]

3.4 Effect of ozone dosage

Dantas et al. [131] accounted the effect of ozone dosage in their research on ozonation of cationic surfactants namely 18-BAC and 16-BAC. As expected, the TOC removal of the two compounds increased with the ozone dose. The TOC removal rate for 16-BAC achieved 25% when ozone dosage was 8.1g/L, while 45% when the ozone dosage was 11.3g/L after 90 minutes of ozonation. In the case of 18-BAC, the TOC removal rate achieved 35% when ozone dosage was 8.1g/L, while 50% when the ozone dosage was 11.3g/L for the same reaction time. Petrovic et al. [137] found that ozone alone is very effective in degrading the non-ionic surfactant which is the nonylphenol ethoxylates (NPEOs). 3.2ppm of ozone were

needed to complete breakdown the parent compounds whereas increasing of ozone concentration to 10.1ppm was able to eliminate the intermediate products completely.

The effect of ozone dosage was also examined by Carbajo et al. [84] in their study on the usage of ozonation as pre-treatment of activated sludge containing benzalkonium chloride (BAC) and NiO nanoparticles. The inlet ozone dosage was stepwise increased from 5 to 300 mg/L. They found that the mineralisation of BAC was not a single process but a series of reactions producing intermediate products. The increase of ozone dosage from 54 to 168 mg/L was necessary to attain a final concentration of BAC below 0.1mg/L. The increase of ozone dosage up to 54mg/L caused a gradual toxicity reduction. Another research done by Wang et al. [119] to study the removal of nonylphenol (NP) for different ozone dosages of 0.50mg/L, 0.75mg/L and 1.00mg/L yielded the removal rate of 63%, 82% and 95% respectively in 20 minutes.

The effect of ozone dosage was also examined in the treatment of laundry wastewater by using different ozone dosages of 32, 43, 53mg/L [138]. The other parameters such as pH and temperature are kept constant. At the ozone dosage of 32mg/L, COD and surfactant removal obtained are 60% and 80% respectively. At the ozone dosage of 43mg/L, COD and surfactant removal obtained are 57% and 77% respectively. Lastly, for the ozone dosage of 53mg/L, the COD and surfactant removal obtained are 53% and 71% respectively. These reverse results show that the surfactant degradation efficiency and COD removal decrease with increasing of ozone concentration.

In general, the increase of ozone concentration promotes the ozone transfer from the gas phase to liquid phase up to reaching the solubility limit at the temperature of the reaction. Once this is achieved further ozone supply will not be transferred to the solution and no

further improvement of removal rates will be observed. Thus an apparent decrease of the reation efficiency will be observed.

In terms of economical aspect, increasing ozone dosage for the wastewater treatment is not cost effective because this requires significant energy. Hence, the ozone dosage and mass transfer needs to be optimized in order to avoid ozone wastage. The ozonation process can be improved by some modification such as increasing of pH value or other ozone based AOPs such as peroxone process and ozone/UV without increasing the ozone dosage although the cost for providing UV and H_2O_2 needs to be considered. Modification of the ozonation process can save the ozone consumption and increased the treatment efficiency as shown in the degradation of anionic surfactant done by Panich et al. [120]. Table 8 shows the effect of ozone dosage on the efficiency of the ozonation treatment.

Table 8 Effect of ozone dosage on the efficiency of the ozonation treatment

Target Compound (s)	Ozone dosage	Results/ Remarks	References
Anionic Surfactant			
Laundry wastewater containing anionic surfactant	31.90, 42.75, 52.95mg/L	COD and surfactant removal of 60% and 80% respectively for ozone dosage of 31.9mg/L. COD and surfactant removal of 57% and 77% respectively for ozone dosage of 42.75mg/L. COD and surfactant removal of 53% and 71% respectively for ozone dosage of 52.95mg/L.	[138]
Non-ionic surfactants			
Nonylphenol (NP)	0.50, 0.75 and 1.00mg/L in reaction time of 90 min.	NP removal rate of 63.2%, 82.4% and 94.6% respectively.	[119]
Cationic surfactants			
Benzyl-dimethyl-hexadecylammonium-chloride (16-BAC)	8.1, 11.3g/L in reaction time of 90min.	TOC removal rate of 25% and 45% for ozone dosage of 8.1 and 11.3 g/L respectively	[131]
Benzyl-dimethyl-stearylammonium-chloride (18-BAC)		TOC removal rate of 35% and 50% for ozone dosage of 8.1 and 11.3 g/L respectively	
Benzalkonium chloride (BAC)	5 to 300 mg/L	Increase of ozone dosage from 54 to 168 mg/L needed to achieve final concentration of BAC below 0.1mg/L. The increase of ozone dosage up to 54mg/L will cause gradual toxicity reduction.	[84]

3.5 Effect of reaction time

Reaction time is another important parameter of ozonation treatment. Kinetics of the reaction between the contaminants and hydroxyl radical or molecular ozone can be studied and this information is important for the engineering design of ozonation and reactors [139]. The treatment time of laundry wastewater by ozonation was affected by the factors such as water matrices, chemical structure of surfactants, ozone dosage, initial surfactant concentration, presence of catalyst and the types of ozonation treatment (catalytic or non-catalytic types).

Gieldowska-Bulska et al. [132] reported that the ozonation treatment of surfactant was affected by the chemical structures of the surfactants such as presence of benzene ring in the molecule, degree of branching of aliphatic chain, polyoxythylene chain and the form of molecule in the solution. The molecular structure with the presence of benzene ring and shorter polyoxyethylene chain were harder to be decomposed by ozone. The linear molecules were faster decomposed than the corresponding molecules with a branched structure. The reaction rate was higher for the surfactant with lower concentration [132].

Another research done by Amat et al. [127] for the treatment of sodium dodecylsulfate (SDS) and tetraethylammonium dodecylbenzenesulfonate (DBS) showed that elevated pH of the solution will improve the ozonation process. Molecular ozone is effectively reacted with the unsaturated DBS but not so effective with aliphatic SDS. The better result is shown by basic pH where the indirect reaction of ozone is an unselective process. In addition, the authors reported that the surface tension increased to the value close to that of distilled water (72Mn/m) after 15 minutes of ozonation process. The active surface activities were changed by ozonation. Beltran et al [128] studied the chemical oxygen demand (COD) and total organic carbon (TOC) removal of sodium dodecylbenzene sulfonate

(SDBS) for 60 minutes ozonation time regardless of pH. They found that the COD and TOC were not completely removed after the treatment. However, the biodegradability of wastewater was improved because of changes in the molecular structure by ozone. Same result of high TOC and COD residual concentration were obtained by Brambilla et al. [140] for the treatment of non-ionic surfactants such as ethylene glycol and diethylene glycol mono-*n*-octylether.

Carbajo et al. [84] studied the ozonation of benzalkonium chloride (BAC) with the presence of nickel oxide nanoparticles. They reported that the extent of mineralization in terms of dissolved organic carbon (DOC) was limited (12%) and several intermediates of transformation products such as carboxylic acids were formed. This shows that the water matrix effect is another factor that will affect the performance of ozonation. The ozone also reacts slowly with aromatic compounds with electron-withdrawing substituents, aliphatic chains and quaternary amines. Other than that, ozonation helps to reduce the toxic effect of BAC [84]. Dantas et al. [131] also compared the efficiency of the photo-Fenton reaction with ozonation for the treatment of 16-BAC and 18-BAC. There is 50% of TOC removal achieved by ozonation after 90 minutes of reaction. Higher removal of 80% of TOC was achieved by addition of photo-Fenton for the same reaction time. One of the disadvantages with photo-Fenton process is the formation of iron (III) hydroxide precipitate. Other than that, the 18-BAC are more reactive with ozone compared to 16-BAC.

Fernandez et al. [141] studied the treatment of LAS mixtures by using ozonation alone and combination of hydrogen peroxide with ozonation. The results showed that the TOC removal is 84% achieved after 3 hours of ozonation process. The efficiency of ozonation was improved with the addition of hydrogen peroxide. The total removal of LAS mixtures was achieved in less than 20 minutes. The optimal pH for the reaction was 9.3

which is a basic pH. Another research is done on the ozonation treatment of hospital laundry wastewater by Kern et al. [25]. After 180 minutes of reaction, the BOD removal of 32.2% and toxicity removal of 96.6% were achieved. Besides that, the surfactant concentration was reduced from 2.22mg/L to less than 0.10mg/L. The performance can be improved by different combination of ozonation processes such as UV/O_3/Fe.

Lechuga et al. [54] studied the combined use of biodegradation and ozonation degradation for the treatment of non-ionic surfactant, alkylpolyglucosides (APG) and anionic surfactant, linear alkylbenzene sulfonates (LAS). The ozonation process was able to destroy the aromatic rings presence in the surfactant structure which is unable to be breakdown by bacteria. In the case of LAS, the surfactant concentration decreases exponentially with ozonation time whereas alkylpolyglucosides decrease linearly with ozonation time. The structure of anionic surfactant with a benzene ring was oxidized faster than the non-ionic surfactant. The time needed for 50% degradation of the APG and LAS are 26 and 7 minutes respectively. The decomposition of both surfactants remained incomplete after 30 minutes.

Overall, the parameter of reaction time is also depending on principles of treatment design such as partial oxidation and full oxidation (complete mineralisation) of pollutants. However, it should be noted that longer reaction time means higher energy consumption to produce ozone. Therefore, optimisation of the ozonation treatment processes according to the treatment goals is needed to reduce the energy consumption to produce ozone. Besides that, determination of the degradation mechanism of the target pollutants along the reaction time of ozonation processes are needed. Moreover, efficiency of ozone-based AOPs such as catalytic ozonation and peroxone process can be investigated in order to achieve higher degradation of pollutants, shorter reaction time and lower ozone dosage. Table 9 shows some of the effects of reaction time on the effectiveness of ozonation treatment.

Table 9 Effect of reaction time on the efficiency of the ozonation treatment

Target Compound (s)	Experimental Conditions	Reaction Time	Results/ Remarks	References
Sodium dodecylsulfate (SDS)	Molecular ozone	Reaction time of 30 minutes, ozone dosage: 8g/h	SBS removal of 20%	[127]
	Ozone in basic pH		SBS removal of 70%	
tetraethylammonium dodecyl benzene sulfonate (DBS)	Molecular ozone		DBS removal of 50%	
	Ozone in basic pH		DBS removal of 80-100%	
Sodium dodecyl benzene sulfate (SDBS)	Regardless of pH	Reaction time of 60 minutes	COD and TOC removal decreased not more than 33% and 16% respectively	[128]
Benzalkonium chloride (BAC)	Without presence of nickel oxide nanoparticles,pH:8.5	Not mentioned.	More than 95% removal of BAC	[84]
	With presence of nickel oxide nanoparticles,pH:8.5		Only 12% removal of DOC	
16-BAC, 18-BAC	Ozonation	Reaction time of 90 minutes	TOC removal of 50%	[131]
	Photo-fenton reaction		TOC removal of 80%	
LAS mixtures (C_{10}, C_{11}, C_{13})	Ozonation at pH 9.3	Reaction time of 3 hours	TOC removal of 84%	[141]
	Addition of H_2O_2 to the ozonation process, pH 9.3	Reaction time of 20 minutes	Total removal of LAS mixtures achieved.	
Triton X-100 (with benzene ring)	Initial concentration of 120mg/dm^3	Reaction time of 60 minutes	28% reduction for Triton X-100 compared to 94.1% reduction of Tergitol TMN 10.	[132]
Tergitol TMN 10 (no benzene ring)	Initial concentration of 100mg/dm^3			
Tritons	3, 4, 8,10 mer, concentration: 20mmol/dm^3	Reaction time of 30 minutes	The molecules with higher number of mer were more easily degraded than those with shorter chain.	
Hospital Laundry wastewater		Reaction time of 180 minutes	BOD removal of 32.2% and toxicity removal of 96.6%, surfactant concentration is reduced from 2.22mg/L to less than 0.10mg/L	[25]
Alkylpolyglucosides (APG)	Initial concentration of 80mg/L, neutral pH.	Time needed for 50% of elimination: 25.8 minutes	The decomposition of surfactants remained incomplete after 30minutes.	[54]
Linear alkylbenzene sulfonates (LAS)		Time needed for 50% of elimination: 7.13 minutes		

4. Ozone-based AOP in laundry detergent wastewater treatment

Besides ozonation under basic condition, there are other methods to produce ROS such as combination of ozone and hydrogen peroxide, catalytic ozonation, combination of ozone and UV irradiation. Their applications in laundry detergent wastewater treatment are discussed in the following sections.

4.1 Combination of ozone and hydrogen peroxide (Peroxone process)

Hydrogen peroxide can combine with ozone in order to enhance the transformation of ozone to the hydroxyl radicals. This treatment system is also known as peroxone process [126]. The authors tested this method on the treatment of anionic surfactants and compare the results with the usage of iron (II) as a catalyst. Both addition of hydrogen peroxide and iron (II) induced the reduction of COD and TOC values. They also found that usage of hydrogen peroxide had a greater influence on the degradation efficiency than that of iron (II) for the surfactants. The degradation efficiency for the surfactant removal by addition of hydrogen peroxide to the ozonation process increased about 30% whereas only 10% for the case of iron (II) as catalyst [126]. These results showed that adding of hydrogen peroxide promotes the decomposition of ozone into hydroxyl radicals that effectively remove surfactant, TOC and COD values besides comparable with the homogeneous catalytic ozonation as shown in reactions (33-35).

The operational parameters such as pH, ozone dosage and ozone mass transfer effected the efficiency of the ozonation process and the peroxone process for the surfactant removal. For example, the degradation of polyethoxylated alcohol mixture (AEO) is more efficient at alkaline pH compared to acidic pH [142]. At the pH of 4.0 and after 60 minutes of reaction, the surfactant seems to be more modified than at the same pH without adding of hydrogen peroxide. The degradation efficiency is higher for the pH of 7.5 and 9.5. Ozonation

of AEO at low pH gives no reduction in TOC but there is modification in the chemical structure. The optimum degradation in terms of TOC was found to be at pH 7.5 with hydrogen peroxide addition [142].

The ozonation with addition of hydrogen peroxide (concentration of 0.001M) for the treatment of SDS and DBS are also tested and proved to be more efficient with 80 to 100% elimination of surfactants after 30 minutes of treatment if compared to single ozonation at neutral pH [127]. Kos et al. [143] evaluated the use of peroxone process for the treatment of real washing water of a textile industry in a flow system. The results show that the COD removal reached 64% which is higher compared to the COD removal of 48% achieved by ozonation process only.

Fernandez et al. [141] also studied the proxone process of linear alkylbenzene sulfonates (LAS) mixtures and commercial detergent and lastly compared the results with that of photocatalytic method. They found that the LAS mixtures was completely degraded in less than 20 minutes for the proxone process and it is faster than the photocatalytic method and also the ozonation reaction alone. The optimum pH for the degradation of LAS is 9.3. The reduction of total organic carbon (TOC) for commercial detergent was only 60% compared to higher TOC removal (84%) in 180 min for LAS standard mixtures. The treatment of commercial detergent is less efficient due to the content of various detergent additives in it and the commercial product is not completely soluble at high concentration too. The authors also identified the transformation products of LAS degradation by using LC-MS equipped with mass spectrometer and ion chromatography for the detection of low molecular short anionic species such as deprotonated formic acid, acetate, oxalate and sulfate ions during the ozonation treatment [141].

Mosteo et al. [144] studied the ozone-based processes such as single ozonation, catalytic ozonation with titanium dioxide (TiO_2) as catalyst, peroxone process and combination of $O_3/H_2O_2/TiO_2$ for the removal of nonylphenol, an endoctrine disrupting substances with the respect to chlorination in drinking water treatment. An average oxidation of 55% of nonylphenols achieved by peroxone processes and this is the highest achievement among the ozone-based processes.

Peroxone process is also capable to increase the biodegradability of surfactant wastewater and this process can be the pre-treatment of biological treatment. As such, the cost of treatment will be reduced because lower dosage of ozone are needed. Kitis et al.[145] reported the successfulness of peroxone process as pre-treatment for the aerobic biodegradation of non-ionic surfactants and polypropylene glycols (PPGs) including ethylene oxide/propylene oxide (EO/PO) block copolymers, linear secondary alcohol ethoxylates (LSAEs) and alkylphenolethoxylates (APEs). The polypropylated compounds (EO/PO block copolymers and PPGs) and LSAEs are biorecalcitrant whereas APEs were partially biodegradable. Increasing of ozone with the stoichiometric hydrogen peroxide increased the extent and rate of biodegradation of the compounds. The effectiveness of peroxone process for the biodegradability improvement is related to the modification of the structure of the pollutants to the more biodegradable one.

In addition, excess hydrogen peroxide can have scavenging effect towards the hydroxyl radicals and the dosage of hydrogen peroxide need to be optimised in order to achieve highest efficiency [69]. Uchiyama et al. [133] reported that the hydrogen peroxide can act as both promoter and a scavenger of hydroxyl radicals. The degradation rate of a non-ionic surfactant, Sannonic SS-90 increased with the increasing of the hydrogen peroxide concentration, reached a maximum value and then decrease with further addition of hydrogen

peroxide. The presence of hydrogen peroxide may depress the degradation process of pollutants.

Furthermore, besides the various factors such as optimum stoichiometry amount of hydrogen peroxide and ozone added and correct pH, Luis and Lombrana [24] reported that the correct timing for hydrogen peroxide addition are important for the application of peroxone process. The authors studied the enhanced process of LAS and dye (R6G) ozonation treatment by addition of hydrogen peroxide. Degradation of R6G was improved with the addition of H_2O_2 at the time after 10 minutes of the experiment. Lower efficiency for LAS removal at acidic pH was observed due to the foaming phenomenon and the predominant molecular mechanism. Adding hydrogen peroxide, the LAS can be completely degraded. Reduction of TOC over 65% was achieved in 10 minutes [24].

In terms of optimisation of the hydrogen peroxide usage in the peroxone process, Turkay et al. [138] investigated the combination of electro-peroxone process and ozonation (E-peroxone) in the laundry wastewater treatment. The E-peroxone process provides on-site hydrogen peroxide generation in controllable rates. The authors commented that the conventional peroxone process is non-feasible and unsafe for real scale application due to the external hydrogen peroxide addition. They compared the removal of COD and surfactants (MBAS) for the conventional ozonation, electro-oxidation (EOX) and combined ozonation with electro-oxidation (E-peroxone). The COD removal by E-peroxone after 120 minutes of process time is higher than ozonation. E-peroxone was found to be more cost-effective than ozonation whereas EOX was found to be the most cost-effective among the methods tested in this experiment. Table 10 shows the summary of laundry wastewater treatment by peroxone process.

Table 10 Summary of laundry wastewater treatment by peroxone process

Type of wastewater/ pollutant	Experimental Conditions/ Dosage of H_2O_2	Performance/ Remarks	References
SDS, DBS	0.001M and reaction time of 30 minutes.	80 to 100% elimination of the surfactants.	[127]
SDBS	Fe^{2+} and H_2O_2, using dielectric barrier discharge	Degradation of SDBS increase for 30% by addition of H_2O_2 whereas only 10% for the case of iron (II) as catalyst. H_2O_2 showed better results in COD reduction compared to iron (II) as catalyst.	[126]
SLS	Fe^{2+} and H_2O_2, using dielectric barrier discharge	H_2O_2 showed better results in COD reduction compared to iron (II) as catalyst. The iron salts performed better than the addition of H_2O_2 in TOC reduction.	
AEO (polyethoxylated alcohols)	Addition of H_2O_2 is in excess. The initial concentration of AEO mixtures is 50mg/L and the pH values are held constant at 4, 7.5 and 9.5.	Optimal degradation (TOC) was found at pH 7.5 with H_2O_2 addition.	[142]
Washing water from textile industry	Addition of 1.5cm^3 of 30% H_2O_2 to 1 dm^3 of wastewater	COD removal reached 64% with addition of H_2O_2 whereas only 48% of COD removal for solely ozonation.	[143]
Ethylene oxide/ propylene oxide (EO/PO) block copolymers, linear secondary alcohol ethoxylates (LSAEs) and alkylphenol ethoxylates (APEs)	Addition of H_2O_2 at a rate of 0.35mg per mg of absorbed ozone (stoichiometric ratio)	In order to achieve an 85% of dissolved organic carbon (DOC) removal, the dosages of 0.3, mgO_3/mg compound (including H_2O_2) required for LSAEs, 1.0 mgO_3/mg for EO/PO and PPGs and 5.0 mgO_3/mg for APEs respectively.	[145]
Sannonic, SS-90 (non-ionic surfactant)	0, 10, 123, 493, 740 mg/L	The oxidation rate increased with increasing concentration of H_2O_2, reached a maximum value and then decreasing with further addition of H_2O_2.	[133]

Table 10 (continued)

Summary of laundry wastewater treatment by peroxone process

Type of wastewater/ pollutant	Experimental Conditions/ Dosage of H_2O_2	Performance/ Remarks	References
LAS mixtures (C_{10}, C_{11}, C_{13})	50 litre of 10ppm LAS and the H_2O_2 used are 22.1μm, using airlift reactor and alkaline condition.	The LAS concentration was reduced to less than 1mg/L in the first 3 minutes. The pH of 9.3 is the optimum pH for LAS degradation. The TOC value only reached 13.6% after 30 minutes of reaction time.	[141]
Commercial detergent	Initial concentration of commercial detergent is 50ppm which is needed to have a total of LAS concentration of 3.5-7.5ppm.	Only 60% TOC removal compared to higher TOC removal (84%) in 180 min for LAS standard mixtures.	
Washing water from textile industry	Addition of 1.5cm^3 of 30% H_2O_2 to 1 dm^3 of wastewater	COD removal reached 64% with addition of H_2O_2 whereas only 48% of COD removal for solely ozonation.	[143]
Nonylphenol (NP), 4n-nonylphenol and technical nonylphenol	1.5mg/L of H_2O_2 with the ozone dosage of 3mg/L	O_3/ H_2O_2 achieve an average of 55% oxidation achieved. The use of post-chlorination after ozone-based processes is suggested for the total removal of NP compounds.	[144]
Real laundry wastewater (mainly consist of anionic surfactants)	E-Peroxone process and the H_2O_2 and ·OH was found to be ~10g/L and 0.1567Mm respectively.	E-Peroxone was not so effective in COD removal. The method removed about 77% of MBAS. It is more cost-effective than conventional ozonation.	[138]
R6G	1000mg of substrate in 20L of water, ozone concentration is 30mg/L, pH between 2.7 and 9.5	Degradation of R6G was improved with the addition of H_2O_2 at the time after 10minutes of the experiment. The mineralization in terms of TOC went from 47% with ozonation to 58% when H_2O_2 is added after 10 minutes.	[24]
LAS	Initial concentration of LAS: 50mg/L, Ozonation with different H_2O_2 / LAS molar ratios, r: 0,10,20 and 60, pH tested: 3.7 and 11.3.	At pH 3.7, LAS elimination is 80%, 90% and 100% for molar ratio, r of 0, 20, and 60 respectively. At pH 11.3, maximum LAS elimination was found at r=10 and 20. Greater TOC removal was obtained at r=20 for both pH values. TOC removal is 45% and 65% for acidic and alkaline pH respectively.	

4.2 Photocatalytic ozonation

In the photocatalytic ozonation, the ultra-violet light (UV) is used together with ozone alone (O_3/UV), in combination with hydrogen peroxide and ozone (O_3/UV/H_2O_2) or in photocatalytic ozonation (O_3/UV/TiO_2) with the goal to produce highly reactive hydroxyl radicals. UV light can help to mineralise the organic compounds that are not transforming in the absence of UV light and ozone can undergo photolysis at 254nm wavelength [114]. Several studies investigated these methods for the treatment of hospital laundry wastewater, industrial wastewater and removal of surfactants. In these processes, degradation of pollutants in laundry wastewater can be mediated by both hydroxyl radical reactions, photolysis or photocatalytic reaction by UV light.

Kist et al. [146] studied the photocatalytic ozonation (O_3/UV/TiO_2, O_3/UV, TiO_2/UV and O_3/TiO_2) for the treatment of hospital laundry wastewater that are polluted with various contaminants such as residues of sanitizers, disinfectants, antibiotics, wetting agents, surfactants and other soils. Ozonation has the important function of disinfection where the microorganism concentration can be reduced to non-infectious levels [146]. The authors reported that the COD removal efficiency is in the range of 20 to 30% and the percentage of COD removal are in the order of O_3/UV/TiO_2 > O_3/UV > O_3/TiO_2 > TiO_2/UV.

In the study conducted by Arslan et al. [90] for the treatment of hospital laundry wastewater by using O_3/UV/H_2O_2 and O_3/UV. They reported that the ozone concentration and the dosage of H_2O_2 are the important factors that affect the efficiency of O_3/UV/H_2O_2 process and therefore the parameters can be optimised by using response surface methodology (RSM) based on central composite design (CCD). The hydrogen peroxide (H_2O_2) is added prior to ozonation. Based on the experiment, the O_3/UV/H_2O_2 process was

effective for COD removal whereas O_3/UV was effective for absorbance removal. Addition of H_2O_2 was indicated to have a negative effect on the COD removal and this may relate to the scavenger effect of excess H_2O_2. Initial pH and ozone concentration have negative interaction in the absorbance removal. This shows that the alkaline pH is needed for the decomposition of ozone into hydroxyl radicals as shown in peroxone process.

Another study done by Kern et al. [25] for the hospital laundry wastewater treatment by comparing the efficiency of UV alone, ozonation alone (O_3), O_3/UV, O_3/UV/Fe^{2+} (Fe^{2+} dosage of 50 and 150mg/L). The best performance is shown by the combination of O_3/UV/Fe^{2+} with the Fe^{2+} dosage of 150mg/L followed by the Fe^{2+} dosage of 50mg/L. However, the treatment needs to be facilitated with pH adjusted between 3.0 and 3.5 and neutralisation after oxidation and radiation [25]. This will involve high utilisation of chemical in order to change the high alkaline pH of laundry wastewater to acidic condition before the treatment [13,14].

UV light and ozone gas have the good property of disinfection and toxicity reduction. Significant reduction in terms of toxicity (EC50 with *D.magna*) were observed for the various processes in which, UV alone (EC50=50.6%), O_3(EC50=47.3%), O_3/UV (EC50=21.75%) and O_3/UV/Fe^{2+} dosage of 150mg/L (EC50=45.4%). The authors suggested that these methods were capable to treat the hospital laundry wastewater that causes toxicity and genotoxicity and future studies can be focused on the usage of them as post-treatment of biological processes and their COD/BOD_5 variations do not justify the pre-treatment for biological processes.

Mozia et al. [52] studied the application O_3/UV method as the intermediate process for the combination system of biological method and membrane separation in the industrial laundry wastewater treatment. The three-step system are in the sequence of biological method,

O_3/UV oxidation and membrane separation. The important finding is that the O_3/UV oxidation prior to membrane separation contributed for membrane fouling alleviation. However, the method needs to be optimised in order to reduce high energy consumption for irradiation and ozone generation. In general, the O_3/UV method is more effective for the compounds that can be degraded through the absorption of the UV irradiation together with the hydroxyl radical reaction that produced during indirect photolysis of the ozone [52].

Another disadvantages of photocatalytic is the generation of excess hydrogen peroxide that will act as hydroxyl radical scavenger. Matsuo et al. [117] studied the O_3/UV method for the treatment of laundry wastewater in the nuclear plant. In the O_3/UV method, the ozone was first dissolved in the wastewater and then the wastewater was irradiated with a mercury lamp. There are two parts of the reaction mechanism which are hydroxyl radical generation by UV-ozone reaction and the organic compounds oxidation by the hydroxyl radicals. The reduction of TOC drops sharply when the dissolved ozone is large in amount but it remained constant at ozone concentration greater than $1.0x10^{-3}mol/10^{-3}m^3$. The saturation of the dissolved ozone is due to the hydrogen peroxide generation according to the reaction (36) below mentioned in section 2.4.1.

$$O_3 + H_2O + hv \rightarrow H_2O_2 + O_2 \quad (36)$$

The excess production of hydrogen peroxide will interfere with the generated hydroxyl radical. Hence, the authors suggested several ways for improvement of the system such as enlargement of the UV light source output, using of mercury lamp with a vapour pressure of 10^5 Pa and run the treatment at the optimum condition where the dissolved ozone concentration is at $3.3x10^{-4}mol/10^{-3}m^3$ in the wastewater [117].

Vilve and Sillanpaa [147] also studied the treatment of nuclear laundry wastewater by O_3, O_3/H_2O_2 and $O_3/UV/H_2O_2$ and at different pH values of 3, 7 and 10 respectively. For the

case of O_3, the highest reduction of COD and TOC were obtained at pH 3 with the reaction time of 2 hours which is quite long. In this acidic pH, the reaction involved are mainly direct ozonation which involve oxidation-reduction reactions, dipolar cycloaddition reactions, electrophilic substitution reactions and nucleophilic addition [63,78]. The direct reaction of ozonation is selective and slow.

Significant improvements of COD, TOC and BOD reductions were detected when ozone was combined with hydrogen peroxide and UV radiation. The authors found that the $O_3/UV/H_2O_2$ is the most effective method with shortest reaction time of about 34 minutes at pH 7. The combination of $O_3/UV/H_2O_2$ obtained the COD, TOC and BOD reductions of 46%, 32% and 70% respectively. The method can produce radicals in many ways. The photolysis of dissolved ozone in water leads to the generation of hydrogen peroxide and then further decomposes to hydroxyl radicals. In addition, the hydroxyl radicals can be generated from the reaction of ozone and hydroxyl ions or by the photolytic decomposition of hydrogen peroxide [147].

Treatment of anionic surfactant (sodium alkylbenzene sulfonate) wastewater with the presence of impurities such as humic acid and natural organic matter (NOM) was done by Shvadchina et al. [70] using the ozonation processes such as O_3, O_3/UV and $O_3/UV/ TiO_2$ and photocatalytic oxidation with oxygen ($O_2/UV/ TiO_2$). The best result was shown by the combination of $O_3/UV/TiO_2$ with surfactant degradation of 86-93% and TOC removal of 74% for reaction time of 20 to 30 minutes. The ozonation and UV irradiation which used separately are less efficient methods for surfactant degradation.

Wang et al. [119] also studied the degradation of nonylphenol (NP) by using O_3/UV method and they found that shorter time and lower dosage of ozone are needed for total degradation of NP as shown in Table 11. The degradation of benzenesulfonate(BS) was done

by Zsilak et al [148] using the combinations such as O_3, O_3/UV, O_3/UV/TiO_2 and photocatalytic oxidation with air (air/UV/TiO_2). The best result is obtained by the combination of O_3/UV/TiO_2 with the initial mineralisation rate of $52.9x10^{-2}$ $mgdm^{-3}min^{-1}$. The optimum pH for the combination method is close to neutral or slightly acidic (6.25).

Zangeneh et al. [149] also study the series combination methods of UV/H_2O_2, O_3/UV, O_3/H_2O_2 and O_3/UV/H_2O_2 with optimisation by RSM for the treatment of linear alkylbenzene industrial wastewater. The four independent variables are initial concentration of H_2O_2, initial pH, ozonation and UV irradiation. The COD removal efficiency increased significantly for the combination methods compared to single method such as UV photolysis, oxidation by H_2O_2 and ozonation. The best result is shown by the combination of O_3/UV/H_2O_2 with the 58% COD removal, maximum ratio of BOD_5/COD (0.55) at neutral pH. However, the combination of O_3/UV/H_2O_2 showed an inhibitory effect at concentration of H_2O_2 higher than 10mM. The authors suggested that the photooxidation processes can be taken as pre-treatment method before biological treatment processes.

Fonagy et al. [150] studied the degradation of aromatic surfactant, namely 4-hydroxybenzenesulfonic acid (4-HBS) using photocatalytic ozonation with heterogeneous catalyst modified with silver (Ag-TiO_2). The modification of the catalyst surface with silver deposition can increase the photocatalytic activity by extension of light absorption, enhanced functions as electron trap and electron accumulator. However, there are high amount of intermediates are produced by the air/UV/Ag-TiO_2 system within the reaction time of 180 minutes if compared to that of ozonation, O_3/UV, heterogeneous photocatalysis with original TiO_2 (combined with both air and ozone respectively). The authors found that the degradation efficiency of O_3/UV/Ag-TiO_2 was just about the sum of the efficiencies of the individual procedures and the mineralization of the intermediates was not accelerated by it. In addition,

ozonation will cause an oxidative dissolution of the deposited silver from the catalyst surface [150]. The summary of various photochemical ozonation are shown in the following Table 11.

One of the challenges of this method are the high consumption of energy that hinder it to be operated in full scale. This is mainly because of the use of both ozone and UV requires high electrical energy and the corresponding high operation cost [56,88]. Besides that, the high turbidity of the wastewater will limits of the use of UV radiation as reported by Kist et al. [146]. Further researches are needed in order to develop more stable and efficient catalyst for the photocatalytic ozonation. As a summary, the important operating parameters such as ozone dosage, UV light intensity, operating pH, hydrogen peroxide dosage need to be optimized in order to achieve high efficiency of degradation besides high energy efficient.

Table 11 Summary of laundry wastewater treatment by photocatalytic ozonation

Type of wastewater/ pollutant	Methods	Experimental Conditions/ Catalyst used and dosage	Performance/ Remarks	References
Hospital laundry wastewater	O_3/UV/TiO_2	2.96mg/cm^2 of TiO_2, pH in the range of 8 and 9, 60 minutes of treatment time, radiated energy used 31.9 J/cm^2	COD removal (30.19%), BOD_5 removal (75.08%), turbidity (45NTU), surfactant (<0.02 ppm)	[146]
	O_3/UV		COD removal (29.76%), BOD_5 removal (41.63%), turbidity (61.1 NTU), surfactant (<0.02 ppm)	
	TiO_2/UV		COD removal (20.33%), BOD_5 removal (68.52%), turbidity (49.9 NTU), surfactant (<0.02 ppm)	
	O_3/TiO_2		COD removal (27.88%), BOD_5 removal (39.67%), turbidity (59.6 NTU), surfactant (<0.02 ppm)	
Hospital Wastewater	O_3/UV	Experimental factors for absorbance removal: pH of 2-11, 0-15mg/L of O_3 and reaction time of 20-60 min	Quadratic models were developed with 60% absorbance and 46% COD removal as the responses. According to the experiment, the O_3/UV/H_2O_2 process was effective for COD removal whereas O_3/UV was effective for absorbance removal.	[90]
	O_3/UV/H_2O_2	Experimental factors for COD removal: pH of 2-10, 0-20mg/L of O_3, reaction time of 30-60 min, and dosage of H_2O_2 of 0.5 -3.0ml.		
Industrial Laundry wastewater	O_3/UV	The method tested as intermediate process for biological and membrane filtration methods *In situ* ozone generation with 40 hours of UV irradiation	Reduction of 60% organic carbon. No effect to the conductivity reduction. Contribute to the alleviation of membrane fouling but optimisation is needed for reduction of energy consumption.	[52]
Nuclear plant laundry wastewater	O_3/UV	Initial temperature (20ºC), wastewater volume ($1.5x10^{-2}m^3$), reaction time of 0, 15, 30, 60, 120, 180, 240, 300 min.	The reduction of TOC drops sharply when the dissolved ozone is large but it remained constant at ozone concentration greater than $1.0x10^{-}$	[117]

			^{3}mol/10^{-3}m^{3}. The optimum condition of the system is when the ozone concentration 3.3x10^{-4}mol/10^{-3}m^{3} in the wastewater.	
Nuclear plant laundry wastewater	O_3	Ozone dose of 0.3mg/O_3/mgCOD at pH 10 for 90 minutes, 0.3mg/O_3/mgCOD at pH 7 for 75 minutes, 0.2mg/O_3/mgCOD at pH 3 for 2 hours	At pH 10, COD removal (30%), BOD removal (38%), TOC removal (24%); At pH 7, COD removal (43%), BOD removal (61%), TOC removal (34%); At pH 3, COD removal (52%), BOD removal (59%), TOC removal (40%)	[147]
	O_3/ H_2O_2 molar ratio of H_2O_2 and incoming ozone:1:2	Ozone dose of 0.6mg/O_3/mgCOD at pH 10 for 65 minutes, 0.4mg/O_3/mgCOD at pH 7.5 for 35 minutes	At pH 10, COD removal (32%), BOD removal (35%), TOC removal (20%); At pH 7.5, COD removal (35%), BOD removal (53%), TOC removal (27%)	
	O_3/UV/H_2O_2 molar ratio of H_2O_2 and incoming ozone:1:2	Ozone dose of 0.5mg/O_3/mgCOD at pH 10 for 30 minutes, 0.3mg/O_3/mgCOD at pH 7 for 34 minutes, UV radiation intensity of 24.4mW/cm^{2}	At pH 10, COD removal (35%), BOD removal (26%), TOC removal (85%); At pH 7, COD removal (46%), BOD removal (70%), TOC removal (32%)	

Table 11 (Continued)

Summary of laundry wastewater treatment by photocatalytic ozonation

Type of wastewater/ pollutant	Methods	Experimental Conditions/ Catalyst used and dosage	Performance/ Remarks	References
Anionic surfactant degradation (Sodium akylbenzene sulfonate)	O_3/UV	Initial concentration of surfactant is 5mg/dm^{3}whereas the concentration of humic acid tested is 10mg/dm^{3}. UV radiation intensity of 5.2mW/cm^{2}, ozone dosage of 0.8-2.0mg/dm^{3}/min.	Surfactant degradation (71-87%), TOC removal (47%) , reaction time of 20-30 min.	[70]
	O_3/UV/TiO_2		Surfactant degradation (94-95%), TOC removal (74%) , reaction time of 20-30 min. The specific ozone consumption per 1mg/dm^{3} of TOC.	
	O_2/UV/TiO_2		Surfactant degradation (86-93%), TOC removal (57%) , reaction time of 20-30 min.	
Nonylphenol(NP)	O_3	NP was dissolved in ethanol for 200mg/L mother solution.	2.4mg of ozone needed for total degradation of NP for the reaction time of 16 minutes.	[119]
	O_3/UV		1.8mg of ozone needed for total degradation of NP for the reaction time of 12 minutes.	
Benzenesulfonate (BS)	O_3	Initial concentration of BS is 10^{-3}M, TiO_2 dosage of 1g/dm^{3}, reaction time of 300minutes.	Initial rates of mineralization: 10.0x10^{-2} mgdm^{-3}min^{-1}, TOC removal of 31-32%	[148]
	O_3 /TiO_2		Initial rates of mineralization: 9.8x10^{-2} mgdm^{-3}min^{-}1, TOC removal of 31-32%	
	O_3/UV		Initial rates of mineralization: 11.2x10^{-2} mgdm^{-3}min^{-}1, TOC removal of 31-32%	

	air/UV/TiO_2		Initial rates of mineralization: $22.9x10^{-2}$ $mgdm^{-3}min^{-1}$	
	O_3/UV/TiO_2		Initial rates of mineralization: $52.9x10^{-2}$ $mgdm^{-3}min^{-1}$	
Linear alkylbenzene industrial wastewater	UV/H_2O_2	Initial pH of 7, initial concentration of H_2O_2 is 100mM, reaction time of 180 minutes. Optimization using RSM	TCOD reduction (49%), BOD/COD_5 ratio (0.46)	[149]
	O_3/UV		TCOD reduction (51%), BOD/COD_5 ratio (0.51)	
	O_3/H_2O_2		TCOD reduction (53%), BOD/COD_5 ratio (0.53), more economic among all the combination.	
	O_3/UV/H_2O_2		TCOD reduction (58%), BOD/COD_5 ratio (0.55)	

Table 11 (Continued)

Summary of laundry wastewater treatment by photocatalytic ozonation

Type of wastewater/ pollutant	Methods	Experimental Conditions/ Catalyst used and dosage	Performance/ Remarks	References
Hospital laundry wastewater	UV	15W germicidal lamp, UV radiation intensity of $0.980mW/cm^2$	COD removal (30.41%), BOD removal (31.8%), toxicity reduction (96.3%), surfactant reduced from 2.22 to <0.10mg/L	[25]
	O_3	Ozone dose of 1222mg/h	COD removal (43.62%), BOD removal (32.2%), toxicity reduction (96.6%), surfactant reduced from 2.22 to <0.10mg/L	
	O_3/UV	Ozone dose of 1222mg/h, UV radiation intensity of $0.980mW/cm^2$	COD removal (37.15%), BOD removal (26.9%), toxicity increased, surfactant reduced from 2.22 to <0.14mg/L	
	O_3/UV/Fe^{2+}	50mg/L of Fe^{2+}, UV radiation intensity of $0.980mW/cm^2$, Ozone dose of 1222mg/h, adjusted pH of 3-3.5	COD removal (53.92%), BOD removal (38.8%), toxicity reduction (96.2%), surfactant reduced from 2.22 to <0.10mg/L	
	O_3/UV/Fe^{2+}	150mg/L of Fe^{2+}, UV radiation intensity of $0.980mW/cm^2$, Ozone dose of 1222mg/h adjusted pH of 3-3.5	COD removal (59.08%), BOD removal (50.3%), toxicity reduction (96.2%), surfactant reduced from 2.22 to <0.10mg/L	
4-hydroxybenzene sulfonic acid (4-HBS)	O_3	Substrate concentration of 10^{-3}M, catalyst used was $1g/dm^3$, ozone concentration of 0.35Mm. total volume of reaction mixture was $3dm^3$	Initial rates of mineralization for TOC ($7.1x10^{-3}mgdm^{-3}min^{-1}$), Initial rates of mineralization for 4-HBS ($2.0x10^{-3}mMmin^{-1}$)	[150]
	O_3/UV		Initial rates of mineralization for TOC ($8.6x10^{-3}mgdm^{-3}min^{-1}$), Initial rates of mineralization for	

	4-HBS ($2.7x10^{-3}$mMmin^{-1})
air/UV/TiO_2	Initial rates of mineralization for TOC ($9.6x10^{-2}$mgdm^{-3}min^{-1}), Initial rates of mineralization for 4-HBS ($3.5x10^{-3}$mMmin^{-1})
air/UV/Ag-TiO_2	Initial rates of mineralization for TOC ($1.1x10^{-1}$mgdm^{-3}min^{-1}), Initial rates of mineralization for 4-HBS ($6.6x10^{-3}$mMmin^{-1})
O_3/UV/TiO_2	Initial rates of mineralization for TOC ($2.5x10^{-1}$mgdm^{-3}min^{-1}), Initial rates of mineralization for 4-HBS ($6.7x10^{-3}$mMmin^{-1})
O_3/UV/Ag-TiO_2	Initial rates of mineralization for TOC ($4.4x10^{-1}$mgdm^{-3}min^{-1}), Initial rates of mineralization for 4-HBS ($1.4x10^{-2}$mMmin^{-1})

4.3 Catalytic ozonation

Among ozone-based oxidation technologies, catalytic ozonation have been widely studied and developed in order to overcome the limitations of ozonation alone such as low solubility of ozone in water and the short lifetime of generated ozone [92,116]. The application of various catalysts on the course of ozonation produces more reactive hydroxyl radicals with lower ozone demands compared to ozonation alone [56], helps to reduce the operational cost since it does not need other energy cost (e.g. UV generation) or cost for pH alteration as it can be applied in wide range of pH values [63]. Homogenous catalytic ozonation employs metal ions as the catalyst whereas heterogenous catalytic ozonation uses metal oxide or metal oxide (or metal) on supports as the main catalysts [93].

Martins et al. [151] evaluated the use of heterogenous catalysts such as Fe_2-O_3-MnO_x and Mn-Ce-O mesoporous catalyst for ozonation treatment of detergent industry wastewater. The catalytic ozonation with Mn-Ce-O catalyst slightly improve the treatment efficiency at

lower pH and the colour removed in less than 3 minutes for all the experimental conditions. About 20% of COD was removed at pH 3.0 and only 3% of COD removal was achieved at pH 9.5. This indicate that the effect of pH on the catalytic ozonation is different from the non-catalytic ozonation system. The ozone/Mn (II) system works better in the acidic condition compared to alkaline condition [78]. The catalytic ozonation with Mn-Ce-O catalyst involved adsorption of both ozone and pollutants followed by surface reaction as shown in the Fig.3. The limitation with the catalyst is high level of carbonyl compounds adsorbed on the recovered solid. Better performance is shown by catalytic ozonation with Fe_2-O_3-MnO_x catalyst which involved 72% COD removal compared to the non-catalytic ozonation system which achieved only 6% COD removal after 60 minutes of ozonation. However, the intermediates formed with this catalytic system were found to have an elevated ecotoxicity. Therefore, the treated effluent cannot be discharged directly to the environment and still can be further treated with biological treatment [151].

Mosteo et al. [144] compared the efficiency of catalytic ozonation with titanium dioxide (TiO_2) as catalyst with the single ozonation, peroxone process and combination of $O_3/H_2O_2/TiO_2$ for the removal of nonylphenol (NP). The use of titanium dioxide as a catalyst did not improve the ozonation process because the ions present in the natural water could be absorbed on the surface of the catalyst. The highest removal of 55% was achieved by peroxone processes. In contrary, Kist et al. [146] studied the effect of titanium dioxide addition to the ozonation treatment of hospital laundry wastewater. After the treatment, the reduction of COD value is from 477 to 344 mg/L (28%), reduction of BOD_5 are from 305 to 184 mg/L (40%) and turbidity reduced from 87.9 to 59.6 NTU.

Rivera-Utrilla et. al. [152] reported that the removal of sodium dodecylbenzenesulfonate (SDBS) from water by combination of ozone and powdered

activated carbon (PAC) achieved removal percentage of 70% whereas 18% and 30% achieved for single ozonation and peroxone process respectively in 5 minutes of reaction time [152]. The author claimed that these results are due to the combined effect of SDBS adsorption on the activated carbon surface and transformation of the dissolved ozone into hydroxyl radicals [152]. Their results are in accordance with the study done by Mendez-Diaz et al. who also used activated carbon as catalyst [153]. They compared the degradation of SDBS by using conventional oxidants (sodium hypochlorite (NaClO), chlorine dioxide (ClO_2), potassium permanganate ($KMnO_4$) and ozone), the peroxone process and combination of ozone and activated carbon. The conventional oxidants are not able to degrade SDBS to an acceptable degree and the use of sodium hypochlorite will cause toxicity increase. The use of peroxone process and combination of ozonation with activated carbon (AC) are much more effective than the conventional oxidants. The combination of ozonation with activated carbon (AC) showed the best result with 80% of SDBS removal and TOC reduction of 50%.

Activated carbon acts as both catalyst and adsorbent in the ozonation processes [69]. Activated carbon acts as initiator of ozone decomposition into hydroxyl radicals because of the modification of its surface chemical properties by ozone [78]. Catalytic ozonation with activated carbon is a pH dependant process and it leads to different reaction mechanisms in acidic and alkaline conditions as shown in the following reactions (39-45) [69].

i) At pH of 2 to 6:

$$O_3 + AC \leftrightarrow O_3 - AC \quad (39)$$

$$O_3 - AC \leftrightarrow O - AC + O_2 \quad (40)$$

$$O_3 + O - AC \leftrightarrow O - AC + 2O_2 \quad (41)$$

ii) At pH (>6):

$$AC + OH^- \leftrightarrow O_3 - AC \quad (42)$$

$$O_3 + OH - AC \leftrightarrow \cdot O_3 - AC + \cdot HO \quad (43)$$

$$\cdot O_3 - AC \leftrightarrow \cdot O - AC + O_2 \quad (44)$$

$$O_3 + \cdot O - AC \leftrightarrow \cdot O_2^- + AC + O_2 \quad (45)$$

The hydroxyl radicals are not formed when the pH is 2 to 6 whereas the generation of hydroxyl radicals is the result of adsorption of ozonide and atomic oxygen on the catalyst surface. Adsorption, desorption and surface chemical reaction are involved in the surface chemical reaction [69]. Catalytic ozonation with carbon is receiving attention for the treatment of industrial wastewater. However, some authors claimed that the use of activated carbon has some disadvantages such as low mechanical strength [78], get saturated easily and replacement is needed for another effective removal of pollutants [69].

Rivera-Utrilla et al. [154] studied the catalytic ozonation to treat sodium dodecylbenzene sulfonate (SDBS) solution using the base treated zeolites, namely zeolite Z-2, zeolite ZSM-5, zeolite Z-13X, zeolite ZSM-5-NaOH. The presence of zeolites with high silicon content not only improves the adsorption of ozone but also ozone stabilization. The basicity of treated zeolites functioned as promoters for the ozone decomposition. The optimum concentration of zeolite for ozone decomposition rate is 100mg/L. SDBS oxidation rate were solely depends on the hydroxyl radicals generation but SDBS adsorption was not observed on the zeolite surface. SDBS oxidation efficiency followed the order of O_3/Z-13X > O_3/Z-2 > O_3 > O_3/ZSM-5. The highest SDBS oxidation rate was obtained using zeolite Z-13X because this catalyst had the largest surface area and the basicity character. The O_3/ZSM-5 obtained the lowest rate because of its acidic character and higher ability to stabilize dissolved molecular ozone. Stabilization of ozone on the catalyst surface hinders generation of hydroxyl radical. The author found that catalytic potential of zeolite ZSM-5 can be

activated by NaOH to produce zeolite ZSM-5-NaOH that has improved SDBS oxidation rate as shown in Table 12.

More recently, Patil et al. [155] studied the treatment of laundry wastewater containing surfactant residues by using conventional ozonation, ultrasound (US), catalytic ozonation and combination of cavitation and ozonation. The catalysts used for catalytic ozonation were solid zinc oxide (ZnO) and copper oxide (CuO). The catalytic ozonation by using solid zinc oxide (ZnO) was very effective in treating the synthetic wastewater (2.5g/L) with the COD removal of 85.8% compared to non-catalytic ozonation in the absence of catalyst which yielded 76.8% COD reduction. The ZnO perform better than CuO because of the smaller particle size of ZnO than CuO, corresponding to greater active surface are and greater catalytic activity. Another important parameters for catalytic ozonation is pH values and the catalyst loading. Maximum degradation was obtained at initial pH of 9 with catalyst loading of 0.1g/L.

The general mechanism of heterogenous catalytic ozonation employing metal oxides such as MgO, ZnO, Al_2O_3 and so forth are as follows [63].

$$O_3 + Me - OH_2^+ \rightarrow \cdot Me - OH^+ + \cdot HO_3 \quad (46)$$

$$2O_3 + Me - OH \rightarrow \cdot Me - O_2^- + \cdot HO_3 + O_2 \quad (47)$$

$$\cdot Me - OH^+ + H_2O \rightarrow Me - OH_2^+ + \cdot HO \quad (48)$$

$$\cdot Me - O_2^- + O_3 + H_2O \rightarrow Me - OH + O_2 + \cdot HO_3 \quad (49)$$

$$\cdot HO_3 \rightarrow \cdot OH + O_2 \quad (50)$$

The dissolved ozone is firstly transferred from the liquid to the catalyst surface. Next, the organic compounds and ozone are adsorbed onto the catalyst surface. Then, the adsorbed ozone will combined with the Me-OH groups and continued with the initiation of radical reactions at the surface active sites (Lewis acid sites or basic sites) of the metal oxide. The

active species such as hydrogen trioxide ($\cdot HO_3$) and hydroxyl radicals can react with ozone to produce more hydroxy radicals or directly degrade the pollutants [63].

It should be noted that the application of zeolite and activated carbon as the catalyst for catalytic ozonation shows that the pollutants removal in laundry wastewater is not merely depends on the attack of hydroxyl radicals, but also include indirect or non-radical mechanisms. The important operating parameters such as pH, ozone dosage, catalyst loading, temperature and reaction time need to be optimised in order to increase the treatment efficiency [78]. The review of literature on the usage of catalytic ozonation technology for the laundry detergent wastewater treatment is limited and the its application is a possible future research for the laundry detergent wastewater remediation. The summary of catalytic ozonation experiments used for laundry detergent wastewater treatment are shown in Table 12.

Table 12 Summary of laundry wastewater treatment by catalytic ozonation

Type of wastewater/ pollutant	Catalyst used/ system	Experimental Conditions/ Dosage	Performance/ Remarks	References
Detergent industry wastewater	Fe_2-O_3-MnO_x	5g of the catalyst is used, inlet ozone concentration of 40gO_3/Nm^3, 500ml of effluent	COD removal of 20% and 3% obtained at pH 3 and 9.5 respectively after 60 minutes. The treatment is limited at alkaline condition.	[151]
	Mn-Ce-O		COD removal of 72% obtained after 60 minutes. The intermediates formed have an elevated ecotoxicity.	
Nonylphenol (NP), 4n-nonylphenol and technical nonylphenol	O_3	Ozone dosage of 3mg/L, pH of solution is 8	30-40% removal of NP	[144]
	O_3/H_2O_2	Ozone dosage of 3mg/L, 1mg H_2O_2/L or with 1g TiO_2/L	55% removal of NP	
	O_3/TiO_2		No effect on the ozonation process	
	O_3/ H_2O_2/TiO_2		35-40% removal of NP	
Sodium dodecylbenzenesulfonate (SDBS)	O_3/PAC	Initial concentration of SDBS ($2.8x10^{-5}$M) in 1 litre solution, pH7 and 5 minute of treatment.	70% removal of SDBS	[152]
	O_3/H_2O_2		30% removal of SDBS	
	O_3		18% removal of SDBS	
Laundry wastewater	Ultrasound (US)	Detergent solution with concentration of 2.5 and 5g/L, volume of solution is 100ml, sonication at 120W	COD reduction of 12.6% and 23.1% for 2.5g/L and 5g/L respectively during initial 30minutes. COD start to increase after 30 minutes.	[155]
	O_3	Detergent solution concentration varying from 0.7 to 5g/L, initial pH9, volume of	COD reduction rapidly in the initial 30 minutes and then slowed down. COD reduction of 81%, 76.8% and 66% for	

		solution is 100ml, sonication at 120W	0.7g/L, 2.5g/L and 5.0g/L respectively.	
	O_3/HC	Detergent solution with concentration of 2.5g/L, volume of solution is 5L, 4 bar pressure, initial pH9,	COD reduction of 39.51%	
	O_3/ZnO	Detergent solution with concentration of 2.5g/L, initial pH9, catalyst loading of 0.05mg/L	COD reduction of 85.8% with catalyst compared to COD reduction of 76.8 % in the absence of catalyst.	
	O_3/CuO		COD reduction nearly similar to that of ozonation in the absence of catalyst	
Sodium dodecylbenzene sulfonate (SDBS)	O_3 without catalyst	Initial concentration of SDBS ($2.8x10^{-5}$M) in 1 litre solution, pH7, ozone concentration of $2x10^{-5}$M	SDBS oxidation rate constant: $2.75±0.66x10^{4}$ s^{-1}	[154]
	O_3/Z-2	Initial concentration of SDBS ($2.8x10^{-5}$M) in 1 litre solution, pH7, ozone concentration of $2x10^{-5}$M, catalyst dosage of 100mg/L	SDBS oxidation rate constant: $4.05±1.16x10^{4}$ s^{-1}	
	O_3/ZSM-5		SDBS oxidation rate constant: $2.69±0.37x10^{4}$ s^{-1}	
	O_3/Z-13X		SDBS oxidation rate constant: $7.36±1.16x10^{4}$ s^{-1}	
	O_3/ZSM-5-NaOH		SDBS oxidation rate constant: $6.25±1.66x10^{4}$ s^{-1}	

Table 12 (Continued)

Summary of laundry wastewater treatment by catalytic ozonation

Type of wastewater/ pollutant	Catalyst used/ system	Experimental Conditions/ Dosage	Performance/ Remarks	References
Sodium dodecylbenzene sulfonate (SDBS)	ClO^-	Initial concentration of SDBS ($2.8x10^{-5}$M), 8mg/L, pH7	SDBS has very low or no reactivity to chlorine after 30 minutes of reaction. Increase in toxicity for longer reaction time.	[153]
	ClO_2	Initial concentration of SDBS ($2.8x10^{-5}$M)	No reduction in in SDBS	
	$KMnO_4$	Initial concentration of SDBS ($2.8x10^{-5}$M), $3x10^{-5}$ and $6x10^{-5}$M, test at pH 2 and 7.	SDBS reduction about 5% after 30 minutes of reaction at pH7, SDBS reduction about 14% after 30 minutes of reaction at pH2	
	O_3	Initial concentration of SDBS ($2.8x10^{-5}$M), ozone concentration of $2x10^{-5}$M	SDBS reduction about 30% after 30 minutes of reaction	
	O_3/H_2O_2	Initial concentration of SDBS ($2.8x10^{-5}$M), ratio of $[O_3]/[H_2O_2]$ is 2:1	SDBS reduction about 50% after 30 minutes of reaction	
	O_3/PAC	Initial concentration of SDBS ($2.8x10^{-5}$M), catalyst dosage of 100mg/L	SDBS reduction about 80% after 30 minutes of reaction	
	O_3/GAC	Initial concentration of SDBS ($2.8x10^{-5}$M), catalyst dosage of 100mg/L	SDBS reduction about 80% after 30 minutes of reaction	

5. Economical Evaluation

Energy consumption is one of the important factors that should be taken into consideration when selecting the best method for wastewater treatment besides treatment efficiency and fulfilment of the stringent environmental regulations. There are two categories of ozonation processes which are non-catalytic ozonation and catalytic ozonation. However, the comparisons with the energy consumption data for the treatment of laundry wastewater by ozonation processes from the data reported in literature is rather difficult because of the wide variety of reactor configurations, ozone generator capacity and the results can differ depending on the organic compound and its concentration. Moreover, there are different removal or treatment results such as detection of surfactant using methylene blue active substances (MBAS), reduction in organic content measured by total organic carbon (TOC) or chemical oxygen demand (COD), partial or complete mineralization to carbon dioxide and water.

The main cost involved in the ozonation processes is the electricity required to produced ozone. The production of ozone gas either from pure oxygen gas or dried air for is an electrical energy-intensive process. The theoretical electricity consumption to produce 1220g of ozone is 1kWh of electrical power corresponding to the enthalpy of formation of ozone which is 0.82kWh/kg ozone [156]. Using the Boltzmann equation, a maximum ozone yield efficiency of 33% (400g O_3/kWh) and 17% (200g O_3/kWh) can be obtained for oxygen feed and dried air feed respectively. However, the ozone yield by an actual ozone generator is far lower than the theoretical calculation, and has reported as about 130g O_3/kWh for pure oxygen feed [76]. During the process, only 10% of electrical energy is used for ozone generation, whereas the remaining 90% of the electrical energy is dissipated as heat depending on the operation conditions of ozone generator [75]. In addition, the photocatalytic

ozonation system may represent an additional cost to the treatment process due to the high energy demands for the UV lamps [56,68].

The cost assessment of the oxidation techniques can be determined by calculating the specific energy consumption (SEC). The energy consumed during the oxidation process is apportioned to the amount of degraded materials [69]. Turkay et al. [138] compared the removal of COD and surfactants (MBAS) for the conventional ozonation, electro-oxidation (EOX) and combined ozonation with electro-oxidation (E-peroxone). The specific energy consumption (SEC) values for EOX, ozonation and E-peroxone were 0.003, 0.079 and 0.067 kWh^{-1} respectively. E-peroxone was found to be more cost-effective than ozonation whereas EOX was found to be the most cost effective among the methods tested in this experiment [138].

On the other hand, another method to determine the cost assessment of the oxidation techniques which highly rely on energy consumption is calculation of the electrical energy per order (EE/O). Electrical energy per order (EE/O) is the electric energy required to degrade a contaminant by one order of magnitude in one unit of volume of contaminated water or air and it is expresses in the unit of KWh/m^3/order [157]. The EE/O is given by the equation (41)

$$EE/O = \frac{P(kW) \times t(h) \times 1000}{V(L) \times \log(\frac{C_i}{C_f})} \quad (41)$$

Where P is the rated power (kW), V is the volume (L) water treated in time t (h), and C_i and C_f are the initial and final concentration of the target compound (mol/L). It should be note that the calculation of EE/O values require optimization of main operating parameters and if possible estimated based on full-scale process [158].

Jimenez et al. [158] use this method to compare the electric energy consumption per order of target pollutant destruction measured by total organic carbon (TOC) for different AOPs such as photocatalysis, Fenton-based processes and ozonation. The calculated E_{EO} for photocatalysis, photo-Fenton, Fenton, sonoFenton and ozonation processes are 505KWh/m^3, 245KWh/m^3, 1634KWh/m^3, 723KWh/m^3 and 188KWh/m^3 respectively. From their results, the ozonation process require low electrical energy to degrade the TOC value compared to other AOPs besides achieving the highest TOC removal [158]. In other word, ozonation is found to be most energy efficient among the other AOPs.

Miklos et al. [56] conducted a comprehensive comparison of EE/O values for different types of AOPs as shown in **Fig. 4**. They classified the different AOPs into 3 main groups based on their median values of EE/O. Group 1 with the EE/O median values less than 1KWh/m^3 is the group of treatment technologies that show their potential to be applied full-scale. Ozonation and ozone based AOPs are in group 1. Photo-Fenton, plasma and eAOP with median value of 2.6, 3.3 and 38.1 KWh/m^3 are in group 2 which indicate that they are too energy intensive for practical purposes but still can be applied for the degradation of specific target pollutants. Lastly, the AOPs such as UV-based photocatalysis, ultrasound and microwave-based AOPs with median value greater than 100 KWh/m^3 are categorized in group 3 with the indication of extremely high energy consumption and inefficient AOPs [56].

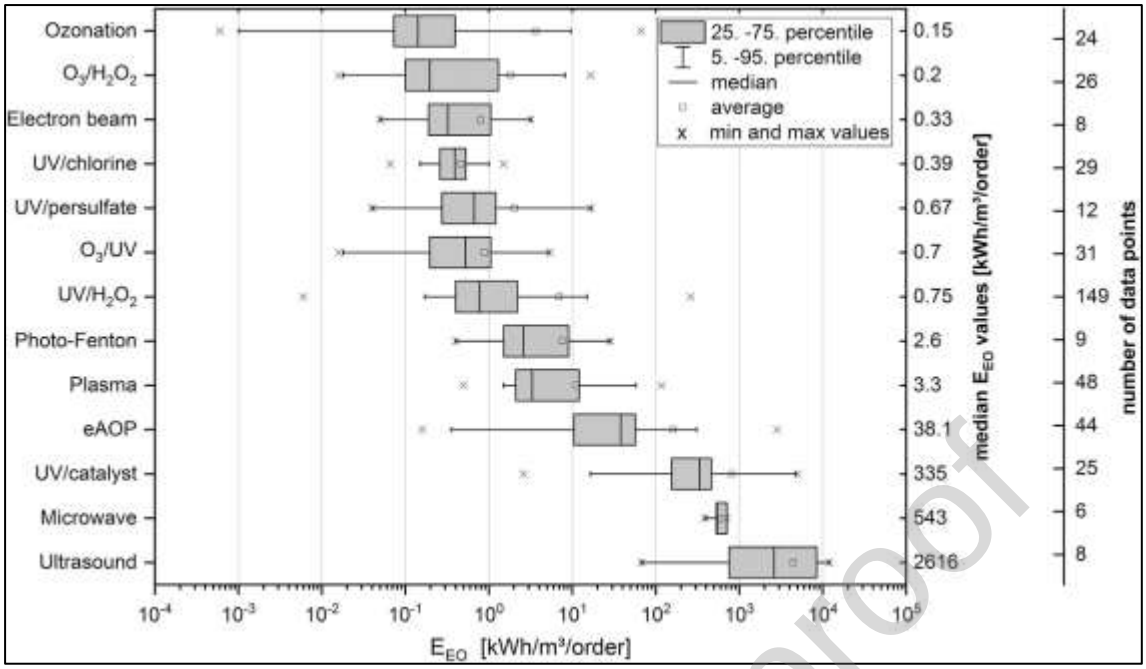

Fig.4: The overview of published EE/O values for different AOPs according to median values [56].

Therefore, ozonation is a suitable option that can be applied commercially. However, the high electricity consumption by ozonation need to be overcome by other substitution of energy supply such as with the using renewable energy sources such as solar energy. This can be one of the important topics of future research. The costs involved in the treatment and reclamation of laundry detergent wastewater by using ozonation or ozone-based treatment are depending on principles of treatment design such as full oxidation (complete mineralization) and partial oxidation. The cost involved can remain rather high and not economically if mineralization is required. However, partial oxidation of water contaminants is feasible since it requires lower ozone dose. Application of ozonation processes as pre-treatment or post-treatment processes can be implemented to reduce the treatment cost after process optimization.

6. Future perspectives and recommendations

Domestic, industrial and institutional laundry wastewater can be recycled and reused in many applications depending on the treated water quality standards. The main goals of laundry detergent wastewater ozonation treatment are summarized as: 1) water reuse and recycling, 2) improve biodegradability of organic substances 3) disinfection and toxicity reduction and 4) complete mineralisation of bio-recalcitrant or toxic organic pollutants. The above mentioned goals can be achieved by varying the ozonation processes according to the characteristics of the laundry detergent wastewater. Hence, more detailed characterisation of physical and chemical components such as combining size fractionation, chemical decomposition and biodegradability in wastewater is important for design and operation of wastewater treatment processes [159].

Depending on the final reuse purpose of the treated laundry wastewater such as for facility process, toilet flushing or irrigation [14], the extent of degradation or oxidation of surfactants can be altered by varying the ozone dosage, ozonation time and also using various types of ozone-based treatments. It is not economically justifiable if the ozonation processes are used for the removal of biodegradable pollutants from wastewater [112]. However, ozonation can serve as supplementary method for biological method such as pre-treatment or post-treatment method. LAS concentrations in municipal wastewater treatment plant treating only domestic wastewater ranges from 1 to 10mg/L. The problems with biological treatment of wastewater with high LAS concentrations include long retention of time for complete LAS removal, excess foaming and large pH adjustment can be mitigated by ozonation [128]. The study done by Beltran et al. [128] using real domestic wastewater and synthetic wastewater with 15mg/L of SDBS show that ozonation process alters the molecular structures of the organic matter initially presented in the wastewater and improve the biodegradability of the organic pollutants.

Ozonation as post-treatment method after biological treatment are widely used as disinfection and pathogen inactivation. Untreated grey water that contains pathogenic bacteria are not suitable for food crops watering, fields irrigation and household use. Common methods for water disinfection are UV lights and chlorination [160]. Ozone is another effective and promising disinfectant that shows better performance than chlorine disinfectant. The problem with chlorine is the generation of halogenated disinfection by-products. Ozonation is also good in colour removal besides the taste and odour removal [77]. An important issue that should be concerned with ozone disinfection or ozonation is the formation of bromate through the reaction of ozone and bromide (Br^-). There are several strategies to reduce the formation of bromate such as decreasing ozone concentration or pH values (<6.0), addition of ammonia or hydrogen peroxide, combination with UV-radiation [63].

The application of catalytic ozonation and peroxone processes not only effectively remove several emerging contaminants (EC) but also help to mitigate the bromate formation [113]. Among ozone-based oxidation technologies, catalytic ozonation technology have been widely studied and developed in order to overcome the limitations of ozonation alone such as low solubility of ozone in water and the short lifetime of generated ozone [116]. The review of literature on the application of catalytic ozonation technology for laundry detergent wastewater treatment indicates that only very few studies have been performed thus further investigation should be performed. Deeper study of the peroxone process and catalytic ozonation not only at laboratory scale but also in practical situation for laundry detergent wastewater treatment remediation are needed.

During the treatment of laundry detergent wastewater by ozonation, the pollutants such as surfactant residues undergo a series of oxidation which results in intermediate

transformation products (TPs) [84]. Ozonation process not only reacts with the primary molecule but also with the intermediate compound formed during ozonation [118]. Depending on the condition of treatment such as ozone dosage used, water pH, chemical structure of pollutant, treatment time, water matrices and so forth, incomplete treatment of surfactants by conventional ozonation generally will produce the intermediate products of organic acid such as carboxylic acid, some homologues with shorter chain length, low molecular products, aldehydes and ketones [83–85,118,134,161]. The degradation by-products after ozonation can be characterized in terms of their chemical structure, toxicity and biodegradability. Hence, it is important to understand and study the degradation pathway and mechanism of the contaminants in laundry wastewater for the optimization of the ozonation processes. The reaction kinetics between the target pollutant and molecular ozone or hydroxyl radicals are important for the engineering design of ozonation processes [139]. Toxicity test should be conducted to ensure that the intermediate products are not harmful than the parent compound [161].

Another important role of ozonation treatment is to destroy poorly biodegradable or toxic organic contaminants in laundry detergent wastewater. However, complete degradation of organic contaminants by using molecular ozone is not economical [63]. Hence, further investigation and optimisation of the ozone-based systems such as catalytic ozonation, peroxone process and so forth are needed for the efficient treatment and recycling of laundry wastewater. The operational parameters for ozonation treatment such as temperature, pH, initial concentration, ozone dosage and reaction time need to be optimised. Review of literature also shows that there is a lack of optimization studies on the ozonation treatment of laundry detergent wastewater. Some example includes the use of response surface methodology (RSM) which allows the optimum utilization of ozone, removal efficiency of

contaminants and operation cost reduction of the system, however more fundamental techno-economical and optimization studied are needed.

Currently, the number of hotels or self-service laundry premises is increasing especially in tourism hot spots and also urban and rural areas. The rapid growing of the laundry service industries will result in mass volume of laundry wastewater produced [162]. This is one of the least address issues in water pollution with the introduction of massive cleaning agents such as detergents into the water source. Therefore, water management and remediation of wastewater from the hotel industry or the self-service laundry shops need special consideration due to the enormous water consumption for personal hygiene and laundry services. Seneviratne [163] reported that a typical commercial laundry can use as much as 400-500m^3/ day and over 85% of this water used in the washing process whereas the remaining water is for steam generation, cooling and amenities. Another study done by Deng and Burnett [164] on the water use characteristics in a hotel (450 guest rooms) with an in-house laundry showed that there are 47% of the total water usage are for laundry services. There are about 70 to 90% of the laundry wastewater discharges to the public sewage system. In addition, the effluent may contain toxic and flammable chemicals that incapable removed by the public treatment facilities before they are discharged to the environment [163]. Ozone treatment method which employed the principles of green chemistry (if operated with renewable electricity) and advanced oxidation process can be used to remediate the wastewater generated by the hotel industry or the self-service laundry shops. The treated water can be reuse for continuous consumption by the hotel or other applications such as wash water, fountain water or irrigation purposes [165].

The recent studies of the ozonation treatment of synthetic surfactant wastewater are based on individual or respective commercial surfactants such as anionic surfactants, cationic

surfactants and non-ionic surfactants. Palmer and Hatley [19] highlighted that there are presence of multiple types of surfactant in wastewater. For instance, anionic surfactant was present together with the other class of surfactant such as cationic surfactant in the same wastewater. This may provide an additional complexity to the treatment method. In addition, the efficiency of biodegradation method is limited by a low concentration of surfactant and concentration of above 1,000,000μg/L will depolarises the bacterial cell wall and therefore destroys function and structure of the bacteria [19]. So, mixtures of surfactants can be another potential research area for ozonation treatment method.

Besides that, ozonation treatment of the amphoteric surfactant are not been well studied if compared to that of anionic and non-ionic surfactants. This may be due to the low application of this surfactant type in laundry detergent [166]. However, based on the recent market report, the market projection for amphoteric surfactants is expected to grow from $3.52 billion in 2018 to $4.94 billion by 2023 due to the rising demand for personal care products and high-performance of amphoteric surfactants [167]. Review of biodegradation treatment for amine-oxide-based surfactants shows that the percentage of ultimate biodegradation was 70% after 28 days [168]. So, it is suggested that more studies should be performed in order to study the feasibility of pre-treatment of amphoteric surfactants with ozonation prior the biological method in order to increase the surfactants' biodegradability.

The detections and occurrence of alkyl phenoxy-benzenesulfonate surfactants (APBS) in municipal wastewater is of concern [169]. The APBS comprised of homologue mixtures of monoalkyldiphenylether sulfonates (MAMS), dialkyldiphenyether sulfonates (DAMS), monoalkyldiphenylether disulfonates (MADS) and dialkyldiphenylether disulfonates (DADS). This type of surfactant is widely used as surfactants, detergents, emulsifiers, solubilizers, wetting agents, dispersants and also industrial processes. However, there is still lacking of

information on their persistence after conventional treatment, ecotoxicity and the environment impact [169]. Hence, there is another opportunity for the study of ozonation processes on this type of surfactant.

Ozone-based treatment is not only can be applied as single treatment method but also can combine with the other treatment methods. O_3/UV method can be applied for example to improve the quality of the effluent in the treatment of industrial laundry wastewater [52] in a three-steps sequential system including biological oxidation, O_3/UV oxidation and membrane separation. The ozone-based method (O_3/UV) reduces membrane fouling and decrease the concentration of organic contaminants. However, in such combined systems process optimization is key to achieve efficient energy consumption [52]. The same process train system can be suggested for the treatment of domestic laundry wastewater or even smaller scale grey water promoting water reuse [170]. Hence, future studies can focus on the development of small scale ozonation wastewater treatment systems for domestic use integrated with electricity produced from renewables. This suggestion can help to facilitate the application of ozonation treatment in practical real-world situation.

7. Conclusion

Ozonation as one of the most effective advanced oxidation processes (AOPs) have shown tremendous potential in treatment and reclamation of laundry detergent wastewater. In this comprehensive paper, the ozonation treatment of laundry detergent wastewater and the effects of various parameters are reviewed. In addition, the important remarks such as potentials and goals of ozonation treatment, economic evaluation and suggested future work scopes on the laundry detergent wastewater treatment are also discussed in detailed. In order to develop the ozonation technologies with higher efficiency, functionality and with low consumption of energy, there are a lot more studies needed in terms of engineering design

and modeling for successful application of the laboratory scale techniques to real-scale operation.

Acknowledgement

This research was supported by the Research Management Centre of Universiti Malaysia Sabah (Grant No. SDK0016-2017) and is gratefully acknowledged.

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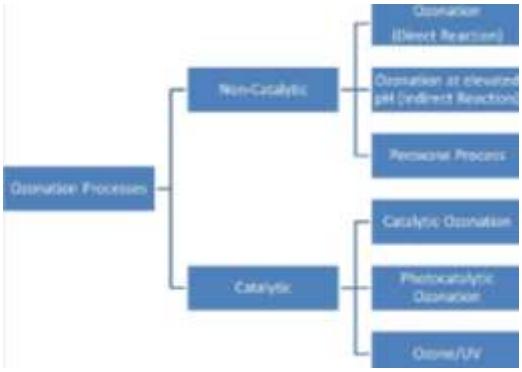
Graphical Abstract



Schematic diagram of various ozonation processes.

The SWOT analysis for ozone technologies

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Research Highlights

- Detergent wastewater is a potential renewable resource for commercialization
- The removal of pollutants in detergent wastewater by ozonation and ozone based AOPs is reviewed
- Catalytic ozonation and peroxone processes are promising technology for detergent wastewater treatment.