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ELIMINATION OF RECALCITRANT POLLUTANTS FROM AQUACULTURE EFFLUENT VIA PHOTOLYSIS AND PHOTOCATALYSIS TREATMENT PROCESSES: A REVIEW PAPER

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Abstract

Aquaculture sectors are facing huge development worldwide. Consequently, aquaculture effluents come from various sources and affect the environment when released without treatment. Pollution and industrial waste are endangering water resources, damaging, and destroying the ecosystems of the entire world. Therefore, it is necessary to develop treatments for their removal in wastewater. Among the different possibilities, photolysis and photocatalysis treatment processes under the advanced oxidation processes (AOPs) may be a sustainable option. Thus, this review aims at providing survey on photolysis and photocatalysis in view of their application for degradation of recalcitrant pollutants from aquaculture effluent. Degradation scheme, experimental factors and employed photocatalyst were discussed in this review.

Keywords: advanced oxidation processes, aquaculture effluent, photolysis, photocatalysis

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1. Introduction

Production intensification of aquaculture would allow the usage of various and more inputs resulting in a rise of waste discharges of the systems. According to FAO (2019), over 91% of global aquaculture production currently being produced within the Asian region which is 102.9 million tonnes in 2017 and total global aquaculture production now

are exceeding the global capture fisheries production by over 18.32 million tons. Considering water used for the farming of aquaculture, the effluent produced from culture ponds after harvesting is also becoming a concern. The amount of water used for aquaculture is abundant, thus producing massive amounts of wastewater (Ghaly et al., 2005). Regrettably, several aquaculture systems contain elevated levels of wastewater that is overflowing with organic matter,

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suspended solids, and total phosphorus discharging into the rivers, lakes, and oceans (Hu et al., 2022). Before discharging the wastewater, a purification system or a pretreatment is crucial for the conservation.

Open and closed systems are the methods of aquaculture systems. For an open system, the methods are extensive, semi-intensive and intensive flow-through systems. Moreover, the method for a closed system is known as RAS which is intensive recirculating aquaculture systems. In RAS, the water is purified and reused continuously. In an open system, water flows once after being treated and finally discharged. These systems have a high intake of water. Even though water is remedied before discharge, the water quality will be decreased with adverse environmental effects (Tejido-núñez et al., 2019). According to Corcoran et al. (2010), the main components in wastewater are organic contaminants, toxic metals, micropollutants, nutrients from plants and pathogenic microorganisms, which are hazardous and cause severe health and environmental complications when released without treatment.

All parts of water treatment play an essential part in the intensive production of fish. Parameters such as total ammonia nitrogen (TAN), chemical oxygen demand (COD), biological oxygen demand (BOD), nitrite, and phosphate are crucial, that their excessive amounts will affect aquatic life. The ecosystem that receives high nitrogen effluent from aquaculture has limited capacity to incorporate and process these compounds into the biogeochemical cycles and the food chain (Lananan et al., 2014). These had caused a large amount of nutrients to continue to accumulate as well as causing ecological imbalance through eutrophication and hyper nitrification which needs to be remedied (Luo et al., 2018).

The decision in choosing the best wastewater recovery treatment is dependent on its subsequent purpose. The chemical procedures applied and the residual products resulting from the treatment should be considered. It can help in preventing contamination from affecting freshwater resources. Therefore, lower costs, robust and efficient wastewater decontamination and disinfection processes are needed without jeopardizing people's health or adding more environmental problems. According to Pellegrin et al. (2011), additional treatment steps are required, either removing the contaminants using membranes or oxidizing agents like chlorine or ozone. The order of the redox potential of these oxidants decreases from $\text{FeO}_2^{-4} > \text{O}_3 > \text{S}_2\text{O}_2^{-8} > \text{H}_2\text{O}_2$, $\text{Cl}_2 > \text{ClO}_2$, as shown in Table 1. Recently, amounts of organic, inorganic, and mineral components in the aquatic environment have escalated significantly. Increased and rapid urban invasion, industrialization, and improved agriculture operations are the cause of this problem (Cardenas et al., 2019). In this review, we summarize the current published work on the emerging field of photolysis and photocatalysis. The aims of this study are to discuss the fundamental aspects of the photolysis and photocatalysis treatment processes on aquaculture

effluent. Some data specific to the degradation of emerging contaminants such as antibiotics that may be present in water are also discussed.

2. Aquaculture effluents

In Malaysia, aquaculture is currently growing to be a significant economic sector. Despite its vast advantages, aquaculture in Malaysia is still strongly associated with environmental issues. Massive fish production requires land use modification to aquaculture sites (Polpanich et al., 2009). The expansion of freshwater and brackish water culture is closely related to both domestic and international markets for these commodities. From only marine culture techniques, brackish and freshwater techniques are now currently used in the culture technique. Brackish water and freshwater aquaculture techniques are done mainly in aquaculture ponds located in or around coastal areas to farm big tiger prawns, giant shrimp, bivalve molluscs, and marine fish (Sampantamit et al., 2020).

Aquaculture is the same as every other manufacturing business, where inputs produce goods. The waste generated by aquaculture systems is a public burden (Dauda et al., 2019). Fish farming is one of the aquaculture activities with the importance of economics. Fish excretion and feed leftovers caused a rise of phosphorus and nitrogen in the water, which can degrade other water bodies. According to Cyrino et al. (2010), these impacts are minimal compared to those soaring from domestic and industrial wastewater. Adequate wastewater treatment method is therefore crucial to prevent dire consequences on the aquatic environment. Effluent-released nutrients could lead to the eutrophication of the receiving water body and other diffuse forms of pollution. Omofunmi et al. (2016) previously investigated the effects of aquaculture effluents produced from catfish farming (*Clarias gariepinus*) in a river by determining the water quality through a few points of view such as chemical, physical, and biological aspects.

Analyses for water and wastewater should be done following the Standard Methods for the examination of water and wastewater. The physical and chemical analysis are alkalinity (CaCO_3), ammoniacal nitrogen (NH_3), total phosphorus (TP), COD, BOD, total hardness (TH), nitrite (NO_2), nitrate (NO_3), Kjeldahl nitrogen, total nitrogen (TN), soluble orthophosphate (PO_4), sedimentable solids (SES), suspended solids (SS), and total solids (TS). According to Coldebella et al. (2018), if the appearance of calcium carbonate within the water is detected, it is related to alkalinity and total hardness. The decreasing amount of water during the fish removal process in fishponds and changes in the surrounding did not obstruct these concentrations. At the time of fish harvesting, pH did not change because of the high capability of alkalinity in maintaining a stable pH. The volume of suspended solids, total nitrogen and total phosphorus were frequently low in aquaculture effluents.

Table 1. Redox potentials of oxidizers used for wastewater treatment (Jiang and Lloyd, 2002)

Oxidizer	Reaction	E° [V]
FeO ₂ ⁻⁴	FeO ₂ ⁻⁴ (aq) + 8H ⁺ + 3e ⁻ ⇌ Fe ³⁺ (aq) + 4H ₂ O	2.200
O ₃	O ₃ (aq) + 2H ⁺ + 2e ⁻ ⇌ O ₂ (aq) + H ₂ O	2.076
S ₂ O ₂ ⁻⁸	S ₂ O ₂ ⁻⁸ (aq) + 2e ⁻ ⇌ 2SO ₄ ²⁻ (aq)	2.010
H ₂ O ₂	H ₂ O ₂ (aq) + 2H ⁺ ⇌ 2H ₂ O	1.776
Cl ₂	Cl ₂ (g) + 2e ⁻ ⇌ 2Cl ⁻	1.358
MnO ₂	MnO ₂ + 4H ⁺ + 2e ⁻ → Mn ²⁺ + 2H ₂ O	1.23
ClO ₂	ClO ⁻ (aq) + H ₂ O + 2e ⁻ ⇌ Cl ⁻ (aq) + 2OH ⁻	0.841
	ClO ₂ (aq) + e ⁻ ⇌ ClO ₂ ⁻ (aq)	0.954

Nitrogen is one of the crucial components for algae growth and its high concentrations lead to a eutrophication process. According to Hu et al., (2012), in aquatic environments, the supply of nutrients is mainly from industries, surface runoff, domestic sewage, animal wastes, aquaculture and leaching. Rivers are water sources for fish farming and rivers will also have high phosphorus and nitrogen concentrations due to other types of pollution. Comprehensive fish farming created effluents rich in nitrogen and phosphorus and will then cause eutrophic effluents. The effluents contain high BOD and high concentrations of SS, TN, and TP.

Waste from feed, antibiotics, and hormones are some additional substances found in aquaculture waste. Feed-derived waste contains components such as phosphorus and nitrogen-based nutrients, or that are in the solid phase such as suspended solid. These solids commonly contain 7%–32% of the TN and 30%–84% of TP in wastewater. Appropriate treatment technology and a proper way must be sufficient to ease the elimination of contaminants. Increasing the quality of feed is a critical matter, with greater bioavailability of phosphorus and proteins, decreasing the amount of fish sewage. Research by Hu et al. (2022) focuses on predicting greenhouse gas emissions from aquaculture wetlands, it is worth noting that these wetlands are often used to treat aquaculture effluent. Other treatment processes such as photolysis and photocatalysis can aid in the elimination of recalcitrant pollutants from aquaculture effluent.

2.1. Sources of waste from aquaculture

According to Ajani et al., (2011), feed contributes to waste from aquaculture. Feed has been reported to be the major source of waste in aquaculture systems (Dauda et al., 2017). The supply of feed to fish in an extensive system depends primarily on the environment while for semi-intensive culture, natural food production is not relied upon. Semi-intensive culture system uses high quality artificial feed targeted for fast growth. Over a large culture field, the fish are left at low density and feed mainly on organisms with support from pond fertilization. The consequence of the production of waste contributed by fish feed depends on the amount of additional feed (Dauda et al., 2017). Waste production from feed depends on so many factors including its nutrient composition, method of production (extruded vs pelleted), ratio of

feed size to fish size, quantity of feed per unit time, feeding method and storage time (Dauda et al., 2019).

Uneaten feed and fecal droppings of cultured fish are commonly in solids. These wastes are occasionally including those fish that do not survive the culture process. Solid waste can then be further classified into suspended solids and settled solids.

Chemicals are still used and needed in aquaculture systems. The application of chemicals in fish farms ponds is restricted but some chemicals are still applied for the purposes of medications, antifoulants, and disinfectants. Medicines are often added to fish farms for chemotherapy reasons, including the use of antibiotics for prophylaxis and curative purposes. For the purpose of microbial infections and for controlling parasites in aquaculture systems, anaesthetics, endoparasiticides, ectoparasiticides and vaccines are used (Ajadi et al., 2016). Among all types of pollutants, a group of recalcitrant compounds is formed by antibiotics (ABs), which are discharged into the aquatic environment. The accumulation of antibiotics in the environment constitutes risk for the aquatic flora and fauna and may cause resistance in some bacterial strains.

Excretion from aquatic species in the aquaculture system, degradation of microorganisms feed and feces are the major contributors of ammoniacal nitrogen in aquaculture wastewater. According to Pratipasen (2014), ammoniacal nitrogen may be able to be imported into aquaculture ponds through nitrogen fertilizers. The common nitrogen fertilizers used in aquaculture systems are ammonium sulfate and ammonium phosphate. Moreover, the urea hydrolyzed in ammoniacal nitrogen and watershed runoff are also the contributors of ammoniacal nitrogen.

Salts are used in aquaculture to decrease tension in fish. The use of lime is for acidity treatment of pond bottoms at the time for preparation of the pond. Although these chemicals are crucial for fish farming, they can be an environmental botheration (Boyd and McNevin, 2015). Water flowed from the ponds and natural water bodies. The consequences of these wastes on systems of natural water are dependent on the capacity of the farm including capacity of the receiving water bodies and the concentration of chemicals applied.

Pathogens are one of the wastes in the aquaculture system, which is seldom examined.

Release of pathogens containing wastewater causes disastrous effects on aquatic life in natural water bodies (Goldburg and Triplett, 1997). The discharge of pond wastewater is uncontrollable in semi-intensive ponds (FAO, 2009), where organic fertilizers such as pig and cow manure, applied in aquaculture with an extremely prominent levels of pathogens (Ansah, 2010). An increase in faecal streptococci levels is also caused by these organic fertilizers (Ampofo and Clerk, 2003).

3. Advanced Oxidation Processes (AOPs)

Advanced oxidation processes that generate powerful hydroxyl or sulfate radicals as its major oxidizing agent was first suggested in the 1980s for portable water treatment (Glaze, 1987). Since then, AOPs were often used to treat various categories of wastewater due to the strong oxidizers that can readily eliminate recalcitrant organic pollutants and degrade a particular inorganic pollutant in wastewater (Deng et al., 2015). Advanced oxidation processes are highly competing technologies for wastewater treatment in degrading organic contaminants. AOPs were first recommended for the use of drinking water treatments in 1980 (Camel and Bermond, 1998).

They were widely researched in later years as oxidizing treatments applied to various wastewaters. These techniques were widely acknowledged as competent in eliminating recalcitrant pollutants or used as a pre-treatment to turn pollutants into shorter-chain compounds which may be handled through conventional treatments such as biological processes. For the remediation of industrial or urban wastewater, one AOP or a combination should be chosen correctly, considering a few factors such as the characteristics of wastewater, technological applicability, regulatory criteria, economic aspects, and long-term environmental consequences (Cardenas et al., 2019).

AOPs are oxidation processes associated with the producing reactive oxygen species such as hydroxyl radicals in suitable quantities to generate recovered effluents. Hydroxyl radicals are non-selective and have high redox potential (2.8 eV) (Pignatello et al., 2006). Carbon radicals (R or R-HO) are produced by their reaction with organic contaminants that may be converted to organic peroxy radicals. Accompanied by the development of other reactive species, the radicals were then further react accompanied by the development of other reactive species resulting in chemical disintegration. In some situations, the mineralization of the target water pollutants. The AOPs demonstrate many advantages compared to conventional water remediation strategies, which include:

a) rather than merely coagulating and filtering from the water, the pollutants are directly eliminated, or decreased, so that secondary contamination could be prevented.

b) Due to their robust non-selective oxidizability, AOPs are applicable to a broad range of pollutants.

c) Since the final reduction of the AOPs is only carbon dioxide, water, or small inorganic ions, no hazardous products will be produced.

According to Bethi et al. (2016), the reaction of organic pollutants into simpler and biodegradable forms is by hydroxyl radicals. The degradation of contaminants may be affected by a few reasons such as pH, water turbidity, reaction time, the volume of organic compounds vulnerable to degradation as well as existence of hydroxyl radical scavengers or stimulator elements.

If AOP is applied in tertiary treatment, hydroxyl radicals are generated in situ due to their short lifetime by a range of procedures, including a combination of oxidizing agents, ultrasound or irradiation, and catalysts (Malato et al., 2009). Titanium dioxide (TiO₂) is the most frequently used catalyst. New systems using immobilized catalysis have been developed as an alternative (Al Dawery, 2013; Da Silva et al., 2015).

AOPs include numerous combinations of ozone, sonolysis, hydrogen peroxide, fenton-related AOPs, sulfate radicals, UV radiation, and photocatalytic treatments, which are effective in oxidizing various pollutants in water, air as well as contaminated soil. One of the characteristics of AOPs, when compared to conventional treatment of wastewater, is during mineralization the biodegradability of the wastewater intensifies when using AOP as the treatment. Another benefit of AOPs is the detoxification of wastewater due to their ability to degrade and eliminate highly toxic organic compounds into less toxic organic compounds (Roshini et al., 2017).

3.1. Photolysis

The use of UV irradiation without the presence of any catalyst to irradiate a polluted aqueous solution is known as photolysis. This process does not completely break down the contaminant molecules, but instead intermediates are generated from the pollutant molecules (Joseph et al., 2009).

According to Giannakis et al. (2015), ultraviolet water and wastewater treatment may imply two mechanisms which are direct or indirect photolysis. Zepp and Cline (1977) did research on direct and indirect photolysis and stated that the pollutants themselves absorb photons and then degrade in direct photolysis. Meanwhile, for indirect photolysis, decomposition happens through reaction of a compound with a reactive species produced by photosensitizers that can absorb radiation to reach an excited state. Dissolved organic matter and nitrate are two wastewater elements that have been identified to be the key role of photosensitizers and have a crucial part in the indirect photolysis of micropollutants in the aquatic system (Ryan et al., 2011). Moreover, humic substances are identified to have significant effects on the degradation of micropollutants, either by promoting their decomposition as a photosensitizer or by preventing their decomposition as a sunlight filter.

Hydroxyl radicals (OH), singlet oxygen ($^1\text{O}_2$), and triplet excited organic matter (^3OM) are examples of reactive species that can be produced during indirect photolysis to react with various compounds and are involved in the degradation of micropollutants such as pesticides and pharmaceuticals. For example, the degradation of cocaine in synthetic municipal wastewater effluent by direct sunlight exposure was higher than observed in distilled water without mineralization taking place in both investigated matrices (Riberio et al., 2019).

In addition to disinfection, UV can degrade organic contaminants via direct photolysis. However, due to the great variability in molecular structures and UV absorption coefficients of contaminants, UV irradiation alone usually cannot provide an effective way to remove the various contaminants present in water (Frangos et al., 2016). Thus, to enhance the degradation of pollutants, the use of UV in photolysis process can be combined with the addition of catalyst.

3.2. Photocatalysis

The word photocatalysis is a combination of photochemistry and catalysis. This indicates that light and a catalyst are essential in generating or accelerating a chemical transformation. Photocatalysis is described as “the acceleration of a photoreaction in the presence of a catalyst” (Serpone and Pelizzetti, 1989). This definition included photosensitization, a process when a photochemical amendment occurs in one chemical species due to the initial absorption of radiation by another chemical species called the photosensitizer (Gao et al., 2019). Photocatalysis is among the most powerful strategies for the AOPs. This process depends on the light irradiation on the photocatalysts to drive the oxidation reaction at the ambient condition. No additional energy is required during the entire reaction process and will produce no toxic by-products; thus, it is surely a green chemical technique (Ge et al., 2019).

Photocatalysts are used in the photocatalysis process. There are various types of photocatalysts for example TiO_2 anatase type or ZnO in the presence of

UV radiation to eliminate contaminants (Zao et al., 2004). Numerous research focus primarily on semiconductor-based photocatalysts for the degradation of contaminants and the water splitting under illumination ultraviolet or visible light. Among numerous photocatalysts of oxide and non-oxide semiconductor, TiO_2 is identified to be the best photocatalyst in various environmental and energy treatment applications. This is due to its properties, suitable valence and the position of the conduction band, long-term stability, nontoxic, low cost, readily available, and strong oxidizing power (Tay et al., 2013).

Several compounds of metal oxides such as titanium dioxide TiO_2 , zinc oxide ZnO , vanadium(V) oxide V_2O_5 , zirconia ZrO_2 , iron (III) oxide Fe_2O_3 , and metal sulfides (CdS , ZnS) have been demonstrated to be a suitable semiconductor photocatalysts in photocatalysis process (Malato et al., 2013). Titanium dioxide is the common photocatalyst used in water treatment. TiO_2 is a wideband semiconductor with anatase and band gap energy of 3.2 eV which means titania needs UV excitation. One of the characteristics of TiO_2 is that it has three polymorphs: anatase, rutile and brookite. There are differences from each other which is the phase. Rutile is the stable phase, while the anatase and brookite are metastable phases. Brookite is less studied as a photocatalyst as it has difficulty in synthesis (Beltran et al., 2006). Commonly, anatase exhibits greater photocatalytic activity than rutile and brookite. Fig. 1 shows the schematic cells for the polymorph of titanium dioxide. The green spheres stand for Ti atoms and, the small red spheres represent O atoms.

The photochemical reaction involved the presence of photocatalyst at the surface of metal oxide. Two simultaneous reactions must occur at the same time. The first reaction will involve the oxidation process from photo-induced positive holes, and the second reaction involves reduction from photo-induced negative electrons (Fujishima et al., 2008).

Numerous photocatalysts have been successfully implemented in the environmental waste management system.

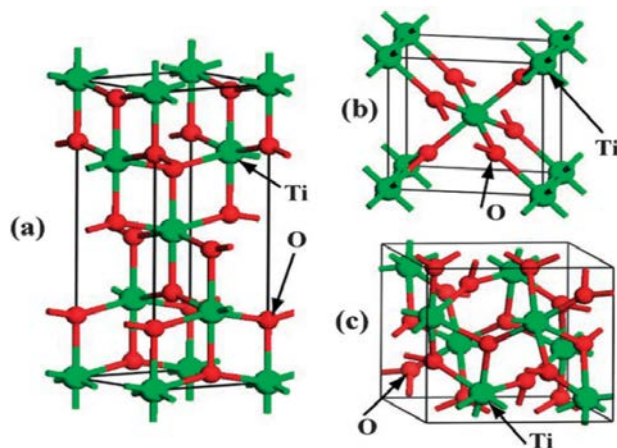


Fig. 1. The schematic conventional cells for the three polymorphs of TiO_2 , (a) anatase, (b) rutile and (c) brookite. The green spheres represent Ti atoms, and the small red spheres represent O atoms. They were adapted from (Zhang et al., 2014)

Major benefit of application of semiconductor-based photoactive catalysts materials in the elimination of contaminants is the complete mineralization of the contaminants into environmentally safe compounds without generating wastes. A perfect photocatalyst must be chemically and photochemically stable, have a high surface area for the adsorption of the reacting species, a high photon absorption coefficient, a small scattering albedo, readily available and low expenses (Ribeiro et al., 2019).

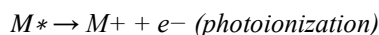
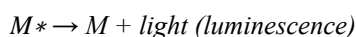
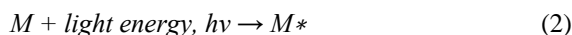
3.3. General mechanism and reaction for photolysis

According to Brightman (1954), photolysis is the use of energy of radiation to generate chemical changes and according to Rowland (1989), light is composed of discrete particles known as quanta or photons that carry an amount of energy (E) determined by its wavelength (λ) or frequency by referring to quantum theory with the equation as shown in Eq. (1).

$$E = h c / \lambda = h \nu \quad (1)$$

where: E = energy; h = Plank's constant (6.63×10^{-34} Js); C = velocity of light (3.0×10^8 ms $^{-1}$); λ = wavelength of the absorbed radiation, ν = frequency

The energy of a photon is inversely proportional to the wavelength, thus the electromagnetic waves can also initiate photolysis reactions with the energy of visible light or higher, such as UV radiations, X-rays, and gamma rays. (James et al., 2018). The absorption step is represented by Eq. (2):



3.4. General mechanism and reaction for photocatalysis

The schematic diagram of the general mechanism of photocatalysis illustrated in Fig. 2.

Hoffman et al. (1995), described photocatalysis as "the acceleration of a photoreaction by the presence of a catalyst". If a semiconductor is in contact with water and then radiated with a wavelength greater or equal than its band gap, the radiant energy will be absorbed and the electrons are promoted from the valence band to the conduction band, which then contribute to the development of electron-hole pairs. These charge carriers recombine with energy and are reemitted as light or heat or migrate to the catalyst's surface. If they have lived long enough to approach the semiconductor or water interface, they can participate in redox reactions. Reaction 3 to 10 below shows the reaction in photocatalysis:

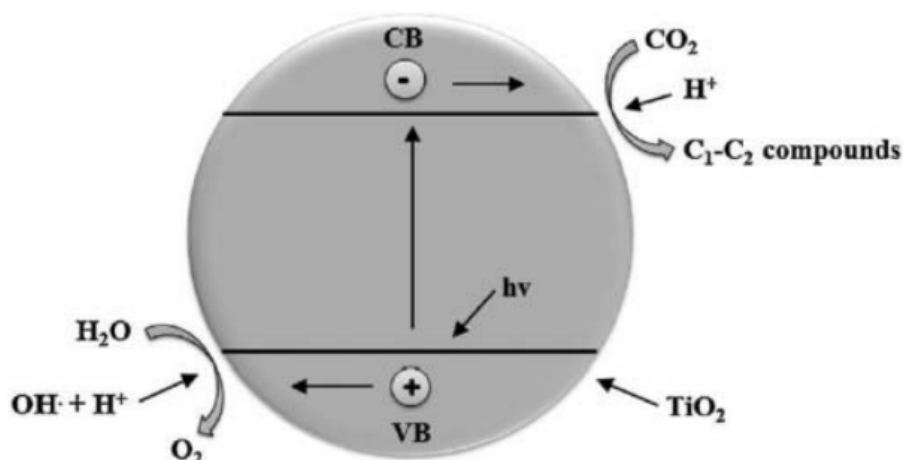
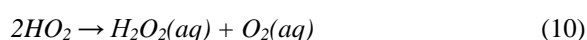
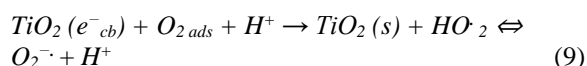
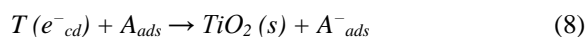
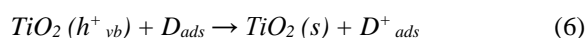
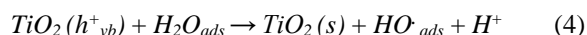
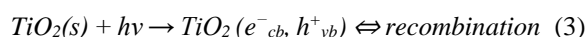


Fig. 2. Schematic diagram of the mechanism of photocatalysis (Updated upon Ola and Morato-Valer, 2015)

The reactive oxygen species produced are very reactive. ROS is not only able to damage a wide range of pollutants in water, but it also causes disastrous effects to the microorganisms (Malato et al., 2013). In a number of situations, the final products of the photocatalytic process of organic compounds are carbon dioxide and water, as well as their respective inorganic acids or salts. Table 2 shows past research on wastewater treatment including in aquaculture effluents using photolysis or photocatalysis treatment process. Nowadays, there are many research using doping of photocatalyst ZnO and TiO₂. Various methods have been established to improve the photocatalytic performance of common photocatalyst such as ZnO and TiO₂, such as doping with metallic (Cr, Fe) and nonmetallic elements (C, N) (Ismael, 2021). In a study conducted by Lin et al. (2019),

photocatalysis of ammonium nitrogen which is one of the pollutants that can be found in aquaculture wastewater.

Compared to the pure ZnO powder, proper doping of Chitin is the most conducive way to enhance UV light's utilization and broaden the ZnO's responsive range. It was observed that 88.64% of NH₄⁺-N could be removed in a short period of irradiation time of 2 h when using a 0.5 g/L Chitin/ZnO (2:3) photocatalyst. Moreover, okadaic acid is classified in diarrhetic shellfish poisoning (DSP) toxins. Okadaic acid has been widely used in biochemistry research as it is an effective specific inhibitor of Ser/Thr protein phosphate. Consequently, OA exhibits tumor-promoting activities and causes cell cycle alterations and cytoskeleton disruption (Munday, 2013).

Table 2. Studies utilizing photocatalysis and photolysis for the degradation of pollutants in aquaculture wastewater

<i>Degradation scheme</i>	<i>Target pollutant</i>	<i>Result</i>	<i>Reference</i>
<p>Bench-scale UV-TiO₂ reactor</p> <p>2 T5 8-watt fluorescent GE Blacklight Blue bulbs</p> <p>Provided irradiance at UVA spectral range of 350 to 400 nm.</p> <p>Plates prepared by spray coat method, with a target immobilized</p> <p>Catalyst concentration 0.25 mg cm⁻²</p>	<p>Geosmin (GSM) and 2-methylisoborneol (MIB)</p>	<p>Batch reactor at full-scale RAS resulted in average GSM and MIB removal efficiencies of 61% and 72%, respectively, with 6 h reaction time.</p> <p>No significant removal of GSM and MIB observed in continuous flow mode</p> <p>No negative effects observed on downstream biological wastewater treatment processes, indicated the proposed UV-TiO₂ process can be safely integrated into RAS</p>	<p>Rodriguez-Gonzalez et al. (2019)</p>
<p>A batch photoreactor was used</p> <p>Quartz tube with a closed base installed concentrically to the reservoir and used to support a UVA lamp (8 W, Philips)</p> <p>5mL samples collected between 0 & 30 min</p>	<p>Benzimidazoles</p>	<p>Photonic flux emitted by UVA₃₆₀ nm lamp was 2.8 J s⁻¹</p> <p>Accumulated UV energy, resulting in a value of 4.8 kJ L⁻¹ after 30 min of irradiation.</p> <p>Benzimidazoles remained stable under illumination of UVA dose for 30 min of exposure</p> <p>Photolysis by UVA radiation was not efficient in removing albendazole and ricobendazole from the effluent samples.</p>	<p>Porto et al. (2019)</p>
<p>Photocatalyst Chitin/ZnO, prepared via sol-gel method.</p> <p>pH water 8.12</p> <p>Irradiated by uv light for 2h.</p>	<p>Ammonium nitrogen</p>	<p>88.73% NH₄⁺-N degradation from 60 mg/L wastewater achieved by direct radiation for 2h</p> <p>Purification rate of wastewater can be improved by addition of chitin with different proportions</p>	<p>Lin et al. (2019)</p>
<p>Nano-TiO₂ photocatalyst prepared via sol-gel method.</p> <p>λ_{max} NH₄⁺-N is 640nm</p> <p>100mg/L NH₄⁺-N stock solution pH values of 4, 5, 6, 7, 8, and 9.</p>	<p>Ammonium nitrogen</p>	<p>1.2 g/L TiO₂ dosage, 20 mg/L NH₄⁺-N initial concentration, pH value 5, 3h reaction time, 92.1% NH₄⁺-N removal was achieved.</p> <p>Crystal structure of photocatalysts remained in the anatase form</p>	<p>Zhu et al. (2011)</p>

<p>Using TiO₂ as photocatalyst</p> <p>Radiated using UVA, UV-B, UVC and solar light initial dye concentration (2-10 ppm) -initial pH (4–10), total reaction time of 1 h.</p>	<p>Methylene blue</p>	<p>Shortest wavelength irradiation (UV-C) proved to be the most effective yielding 100% degradation of MB was achieved within 14 min.</p>	<p>Joseph et al., (2016)</p>
<p>Chlorination conducted in batch reactors; free available chlorine (FAC) stock solution added into the samples containing ETA.</p> <p>UV irradiation conducted in a quasi-collimated beam system, low-pressure mercury lamp 254 nm.</p> <p>Samples prepared in a crystallization dish</p>	<p>Ethanolamine (ETA)</p>	<p>Photolysis rate constant (kUV) was 1.0x 10⁻³ cm²/mJ for N-Cl ETA and 1.3x 10⁻³ cm²/mJ for N-Cl₂ ETA</p> <p>UV/chlorine was efficient in treating ETA under wide range of conditions for FAC</p> <p>Photolysis of N-Cl and N-Cl₂ ETA responsible for ETA degradation (≥75%)</p> <p>Efficient abatement for ETA by UV/chlorine with kUV of up to 1.5x10⁻³ cm²/mJ</p>	<p>Ra and Lee, (2021)</p>
<p>Experiment performed in Atlas Suntest XLS+ solar simulator system equipped with a Xenon arc lamp, irradiance 500 W m⁻²</p> <p>Ultrapure water and 2 wastewaters from a lagoon treatment system</p>	<p>Amisulpride and desipramine</p>	<p>Amisulpride short irradiation times (t_{1/2} approximately 3h in pure water and 4h in wastewater) were adequate to degrade the parent compound.</p> <p>Longer exposure period required for desipramine (t_{1/2} of approximately 36 h in pure water),degradation of despiramine enhanced almost 3 times by indirect photolysis</p>	<p>Gros et al. (2015)</p>
<p>Fe₂O₃/S-doped C₃N₄ nanocomposites were for photocatalytic ozonation treatment.</p> <p>Reactor with ozone generator and UVA/ UVC/LED lamps.</p> <p>Different light irradiation wavelengths, 80 W UVC, 80 W UVA and 3 W LED</p>	<p>Bisphenol A (BPA) and carbamazepine (CBZ)</p>	<p>Fe(4.3) CNS/O₃/LED technique showed high mineralization degree for the BPA and the CBZ by changing the wavelength from k = 254 nm to k = 460nm</p> <p>Photo-assisted ozonation process the BPA and CBZ degradations were not significantly changed by replacing irradiation, BPA degradation was increased in presence of Fe(4.3)CNS photocatalyst.</p> <p>Pseudo-first order kinetic coefficients for the degradation of BP</p> <p>Highest BPA mineralization (97.8%) performed by the FeCNS/O₃/LED method, irradiated under LED radiation for 3h</p>	<p>Jourshabani et al. (2019)</p>
<p>Photocatalysis experiment conducted in lab-scale photoreactor, 4 UV lamps (Philips PL-L 36W/09/ 4P UVA)</p> <p>Experiments carried out in triplicate in the dark and irradiated by UV lamp with the presence and absence of the catalyst TiO₂.</p> <p>Performed at pH 6.9 in DW, 7.3 in AOW and 7.8 in SW</p> <p>10 µg/mL OA prepared in DW), were maintained under magnetic stirring at 20–22°C for 2h.</p>	<p>Okadaic acid (OA)</p>	<p>Complete degradation of OA was determined in sw after 30 min and 7.5 min in dw</p> <p>The rate constants fit the pseudo-first order kinetic model (R² >0.96)</p> <p>Detoxification was achieved in parallel with the degradation of OA in dw and aow (≤20 min) but not for sw</p> <p>Results suggest that UV/TiO₂ photocatalysis can be an effective approach for degradation of OA.</p>	<p>Camacho-Muñoz, et al. 2020)</p>

<p>Batch reactor for 40 mg L⁻¹ MTZ, recirculation reactor with immobilized catalyst for 10 mg L⁻¹ of MTZ</p> <p>3h reaction time for photocatalytic Philips Solarium lamp fitted with 4×15W Cleo fluorescent tubes, maximum emission at 365 nm, irradiated power of 9 W m⁻²</p>	<p>Metronidazole (MTZ)</p>	<p>Photolysis demonstrated no mineralization in MTZ degradation</p> <p>Best configuration catalyst immobilized on the outer with the internal UV-C lamp & H₂O₂ (k_{E,UV-C}; 0.055 min⁻¹)</p> <p>Degradation efficiency processes with catalyst immobilized on outer tube with internal UV-C lamp (k_{E,UV-C}; 0.042 min⁻¹), compared UV-A lamp (k_{E,UV-A}; 0.022 min⁻¹)</p> <p>Considering TiO₂ photocatalysis, the best results were obtained for the catalyst Evonik-P90 and Evonik-P25</p> <p>100% MTZ elimination successfully achieved after 2h and up to 6 all re-runs.</p>	<p>Santana et al. (2017)</p>
<p>OTC (4 mg/L) aqueous solutions performed using quartz tubes.</p> <p>The irradiations were carried out in a sunlight simulator, illumination of 55 W/m² (290–400 nm), corresponding to 550 W/m² in all spectral range. OTC were irradiated for 5 different times (2–20 minutes)</p>	<p>Oxytetracycline</p>	<p>Ca²⁺ enhances the kinetic rate constant of OTC but, when Mg²⁺ is also in solution, an inhibition of the accelerating effect of Ca²⁺ occurs.</p> <p>Similar binding patterns of OTC with Ca²⁺ and Mg²⁺ and a lower quantum yield of the direct photolysis of the complexes with Mg²⁺</p> <p>Ca²⁺ and Mg²⁺ induce different mechanisms of OTC photodegradation which also affect the kinetics of formation/degradation of some OTC photoproducts.</p>	<p>Leal et al. (2019)</p>
<p>20 mL OXA solutions in ultrapure and natural water irradiated, in triplicate, in quartz tubes covered with Parafilm.</p> <p>500 µL irradiated OXA and dark controls withdrawn from quartz tubes, stored at 4 °C and analyzed within 24h</p>	<p>Oxolinic acid</p>	<p>T_{1/2} increased from 0.9h (ultrapure water) to 1.65–4.03 h (natural water).</p> <p>Dissolved organic matter (DOM) explain the decrease of OXA photodegradation observed in freshwater.</p> <p>Sharp decrease of OXA photodegradation observed in solutions containing NaCl and in synthetic sea salts</p>	<p>Louros et al. (2020)</p>

In the photocatalytic process, the presence of ions has been reported as an adverse effects at concentration as low as 10⁻³ M due to the competitive adsorption at the active sites of the catalyst (Mills and Le Hunte, 1997) or acting as radical scavengers (Bennedsen et al., 2012). However, this effect was not observed in this study. There are no previous reports about the photocatalysis effectiveness of UV/TiO₂ on OA in seawater for comparison. The slower rate constant in SW compared to AOW might be due to the presence of organic matter in the matrix. As it has been reported previously, the nature and concentration of organic matter in the matrix can inhibit or promote photocatalytic reactions (Li and Hu, 2016). By using photocatalysis and TiO₂ as the photocatalyst, OA was degraded and their TPs in the marine environment. This research has shown that this is the first report on the use of photocatalysis to degrade marine toxins and its promising potential to protect shellfish harvesting sites (Camacho-Muñoz, et al., 2020).

Emerging AOPs such as the combination of photocatalysis and ozonation had shown positive results in degrading persistent pollutants. It has been

reported that persistent contaminants such as carbamazepine (CBZ) and bisphenol A (BPA) remained unchanged through secondary treatment in wastewater treatment plants. As such, in research using integrated photocatalytic ozonation treatment by Jourshabani et al., 2019, modified graphitic carbon nitride photocatalyst is used for both degradation and mineralization of bisphenol A (BPA, 50 mg/L) as well as carbamazepine (CBZ, 10 mg/L), which is a visible-light insensitive pollutant. The highest BPA mineralization (97.8%) was performed by the FeCNS/O₃/LED method and photocatalytic ozonation using FeCNS photocatalyst showed high mineralization degree.

Recalcitrant pollutants in aquaculture such as antibiotics are the main contributors of pollutants in aquaculture effluent and are commonly administered as medicated feed. A study conducted by Leal *et al.*, 2019, for the photodegradation of oxytetracycline with the complexation metal ions namely Ca²⁺ and Mg²⁺ proved that calcium promotes a faster OTC photodegradation in aqueous solution, while the same does not occur with Mg²⁺.

4. Economic and cost benefits of AOPs

In recent years, much research about advanced oxidation processes in wastewater treatment is focused on their usefulness in lessening hazardous compounds from a chemical point of view. Nonetheless, the energy efficiency for the removal of contaminants and the cost of elimination processes are correlated. Numerous studies have shown these technologies' potential to oxidize and eliminate numerous organic and inorganic compounds from the air, water and wastewater (Mehrojuei et al., 2014). Recalcitrant contaminants and microorganisms which demonstrated high persistence against conventional method are mainly considered as suitable targets for advanced oxidation processes (Robertson et al., 2012).

Cost and high energy involved when using advanced oxidation processes, such as ozone generation, light sources, and pumps caused them to be classified as costly processes. Thus, economic feasibility research is required in each treatment case, other than determining the specialized values of these technologies in remediating and tackling environmental issues. Unfortunately, this significant viewpoint that can advance the commercialization of AOPs is often neglected (Chong et al., 2012).

A review by Mahamuni and Adewuyi (2011) on estimation cost involving ultrasound on trichloroethylene (TCE), phenol, and reactive azo dyes concluded that combining ultrasound with different AOPs was economically more alluring than the application of only ultrasound for wastewater treatment. Table 3 shows the cost and treatment for three different pollutants and their expenses.

The amount of energy needed to accomplish the degradation of the target pollutant was determined from the energy density (watt/ml). Consideration was taken for the capital and operating costs to determine the treatment process cost. These treatment expenses were then analyzed and contrasted with other AOPs.

In the treatability tests, high energy densities were used for US wastewater treatment, and this shows the reason for the prohibitive cost and expense of US wastewater treatment. Till this day, the cost of wastewater treatment using hybrid ultrasonic processes are one to two orders of magnitude more than currently applied AOPs such as O_3/H_2O_2 ,

ozonation and UV/ H_2O_2 (Mahamuni and Adewuyi, 2011). It can be observed that cost for treatment of TCE using US only is magnitude lower than the other 2 pollutants treated.

The energy density for TCE is the lowest which can be seen in Table 3. Moreover, TCE is a hydrophobic contaminant thus the mechanism of degradation is degraded inside the cavitation bubbles. Hence the mass transport of OH- radicals to the contaminant becomes the controlling step of the degradation mechanism.

Several processes have been developed such as the high-temperature Fenton process, O_3/H_2O_2 , H_2O_2/UV , and $O_3/catalyst$. However, their costly expenses indicated that their application is restricted mainly on the treatment of effluents with a low flow rate. The practical application of AOPs is as a pre-treatment stage to intensify biodegradability as a primary process for particular product flows or work as the finishing stage. More cost-effective processes are needed to enable the development of a wide scale for these water treatments (Suty et al., 2004).

Photolytic methods for the degradation of pollutants in water are based on providing dissolved energy to chemical compounds in the form of radiation. Among the main advantages of photolysis treatment process is it is a chemical free treatment that requires relatively modest maintenance and operational costs.

Research by Cuerda (2019), stated that using photolysis, wide variety of antibiotics used in aquaculture were able to be degraded.

5. Development of UV radiation

According to Ahmad (2017), ultraviolet (UV) radiation is electromagnetic radiation with the wavelength ranging from 100–400 nm. The two main origins of the UV radiations are from the sun and human-made UV lamps. The ultraviolet radiation is classified into three bands according to its wavelength. Visible light, which is longer than 400 nm starts with blue light, and on the other is x-rays which are shorter than 100 nm. Table 4 shows the wavelength and types of ultraviolet radiation. The terms UV light or UV rays are also used as synonyms for UV radiation. (Ahmad and Christensen, 2017).

Table 3. Expenses for different type of wastewater treatment (Mahamuni and Adewuyi, 2011)

<i>Pollutant</i>	<i>Type of treatment</i>	<i>Cost (\$, per 1000 gallons)</i>
Phenol	UV/US/ O_3	\$89
	US only	\$15,536
Trichloroethylene (TCE)	US + UV	\$25
	US only	\$91
Azo dyes	US + UV + H_2O_2	\$65
	US only	\$14,203

Table 4. Type of ultraviolet radiations and the wavelength

Type of UV	Wavelength (nm)
Ultraviolet A	320–400
Ultraviolet B	290–320
Ultraviolet C	100–280

UV radiations have serious health effects on humans. Solar radiation is essential for vitamin D synthesis. However, prolonged exposure to radiation for a long time may cause some serious health conditions such as edema, erythema, burns, immunosuppression, and skin cancer (Gordon et al., 2013). In addition, UV radiation causes direct or indirect biological harm through the generation of ROS. When UVB is absorbed by DNA it causes severe damage to the DNA, which are cyclobutane-pyrimidine dimers and (6-4) pyrimidine-pyrimidone photoproducts. Meanwhile, UVA generates ROS that causes oxidative damage to DNA such as 8-oxo-deoxyguanosine (Rastogi et al., 2010).

Since sunlight is pervasive, it is potentially exposed to humans, and the most often exposed organ is the skin. According to Gabet *et al.*, (2021), a few research have concluded the ability of sulfate radicals ($\text{SO}_4^{\cdot-}$) generated by persulfate UVA or UVC photoactivation to degrade micropollutants and estrogens in wastewater (Angkaew et al., 2019). Using UVC illumination, treatment efficacy for recalcitrant organic contaminants in wastewater effluents was established using oxidant species catalysts such as hydrogen peroxide (H_2O_2) and persulfate ($\text{S}_2\text{O}_8^{2-}$).

5.1. Ultraviolet lamps (UV Lamps)

UVC lamps are fragile, formed from mercury and need extra energy compared to UVA and UVB lamps. UVC can be replaced by LED to reduce the operational expenses (Beck et al., 2017). However, the performance of UVC-LED performance in wastewater treatment was less than 3%. This demonstrates that their utilization is not suitable for long term wastewater treatments. Due to these circumstances, the implementation of UVA and UVB lamps to improvise water treatment processes can provide various positive outcomes (Huang et al., 2018).

In most cases, the photoactivation applied in advanced oxidation processes is performed with radiation from UVC. Subsequently, they also have their own drawbacks. UVC lamps are expensive and, most importantly, hazardous to use and manipulate. Applying lamps other than UVC, such as UVA or UVB lamps aims for more sustainable processes. Benefits like less energy needed, being nonhazardous when manipulated and can be substituted by solar light for wide-scale applications are the characteristics of UVA and UVB lamps. Lamps for research purposes are easier to use and manage than solar light (Brauer et al., 2021). Continuous-wave mercury vapor lamps are commonly used in photochemical AOPs, in which the lamps are arc discharge instruments that emit UV

radiation by creating an electric arc between the two electrodes in specifically constructed lamps containing mercury vapor or a combination of mercury and another gas [40]. The energy released by the excitation of the mercury vapor results in the emission of UV radiation. However, there are concerns over mercury toxicity, high energy needs, mechanical instability, and the cleaning costs of the application of mercury lamps, thus emphasizing the need to find alternative UV sources.

5.2. Ultraviolet Light-Emitting Diodes (UV-LEDs)

UV-LEDs are LEDs that emit ultraviolet light with wavelengths of 400 nm or less. However, it is crucial to adhere to the conventional classification of UV wavelength ranges and identify UVA, UVB and UVC LEDs when focused on UV LED application in AOPs (Matafonova and Batoev 2018). UV-LEDs are superior to UV lamps in that they are mercury-free, as well as having superior energy efficiency, a longer lifespan, more consistent light output, and easy temperature and heat management (Muramoto et al., 2014). Recently investigations had shown good disinfection performance in wastewater containing PPCPs, dyestuffs, and pesticides by using UV-LED/PS process. However, most of them focused on continuous illumination of LEDs in process.

Based on all pros of the newly investigated UV-LEDs and recent developments of semiconductor technology and outstanding improvement of UV-LEDs' performance and its capability in coupling with advanced oxidation processes, UV-LEDs are considered to be an efficient technology for water purification approaches.

5.3. Pulsed UV lamps

Pulsed UV (PUV) lamps are a non-mercury alternative to mercury vapor lamps. PUV lamps is a method initially developed for microbial inactivation based on the use of xenon lamps that emit wide spectrum light pulses of exceedingly high intensity and short duration, with a continuous spectrum ranging from ultraviolet light to infrared radiation. The food industry was the first to use PL technology, but in recent years, its application domains have broadened (Baranda et al., 2017).

It is considered a safe and environmentally friendly technology due to the use of mercury free xenon flash lamps and the lack of applied chemicals disinfectants or preservatives as well as waste effluent. As PUV contains a significant amount of UV light, a potential application of this technology might be its

usage as a part of an AOP for the breakdown of polluting dyes, pesticides, inactivation of a wide broad of waterborne microorganism and organic contaminants (Bohrerova et al., 2008; Navarro et al., 2017).

6. Conclusions

Photolysis and photocatalysis has proven that pollutants in aquaculture effluents such as ammonium nitrogen can be degraded by using these processes. It was confirmed that AOPs that facilitate the combination of ultraviolet radiation, catalyst, and oxidant to produce hydroxyl radicals (OH) in solution can be used as effective technology for the degradation of aquaculture wastewater. The study summarized that photocatalysis could be used for the complete mineralization of various pollutants as well as recalcitrant pollutants present in the water using light and a photocatalyst by simultaneous occurrence of oxidation and reduction reactions.

The information gathered from the literature could lead to various areas where future improvements could be made. The conceptual proof of advanced oxidation processes has already been presented for wastewater treatment yet the application of AOPs on a wide- scale is less studied.

Scaling up an energy efficient, cost effective and sustainable technique for the complete mineralization of diverse types of recalcitrant organic pollutants from water by using a reusable energy source sunlight and industrial AOPs or hybrid AOPs is the challenging future aspect. The generation of wastes such as poisoned catalyst in photocatalysis should be minimized and possible alternatives for the valorization of such waste should be explored.

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