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REVIEW

A comprehensive review on state-of-the-art photo-, sono-, and sonophotocatalytic treatments to degrade emerging contaminants

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Abstract

Emerging contaminants (ECs) are commonly originated from personal care products, cosmetics, pharmaceuticals, pesticides, dioxins, polycyclic aromatic hydrocarbons (PAHs), and alkylphenolic compounds. Due to the huge development of these industries, these ECs have been constantly detected in wastewater, groundwater, and surface water in hazardous quantity. The discharge of these ECs into the environment causes considerable non-esthetic pollution and could be a great threat to the entire ecosystem. The common wastewater treatment plants (WWTPs) which consist of biological, physical, and chemical treatments such as activated sludge, filtration, adsorption, and coagulation are found to be ineffective for desired removal of ECs. In turn, various emerging advanced oxidation processes (AOPs) such as ultrasonic and ultraviolet irradiation with or without the presence of catalyst have raised great attention due to their great potential in remediation of ECs. This paper presents a critical review on types, recent occurrence, sources, environmental impacts, and emerging treatment methods applicable to treat ECs. The current research and applications of ultrasonic, ultraviolet, and combination of both irradiations to treat ECs in wastewater are particularly reviewed. The effect of key parameters on photo-, sono- and, sonophotocatalytic degradation of ECs are commendably accessed such as ultrasonic power, ultrasonic frequency, light intensity, ultraviolet wavelength, solution pH, oxidizing agents, chemical additives, catalyst dosage, and modification of catalyst. The possible reaction mechanisms of ECs degradation process and kinetic model study are also elucidated in detail. Lastly, future research directions and conclusions are proposed to strengthen the understanding on their fate in water. All this information is vital to predict the negative impacts of ECs on the receiving environment effectively.

Keywords Emerging contaminants · Degradation · Photocatalysis · Sonocatalysis · Sonophotocatalysis

Introduction

In recent decades, water has become a scarce resource due to the rapid advancement in economy, agriculture, and industrialization. Water has been used in many sectors such as manufacturing industries, agriculture, domestics, and transportation (Pal et al. 2014; Tijani et al. 2016). Recently, concern has been raised on the urgent need of appropriate water treatment for water regeneration due to depletion of water resources. Today, the public are focusing even more on newly detected pollutants known as emerging contaminants (ECs). ECs are not new kind of pollutants, but they are being discharged to the environment since they have been discovered in 1800s due to the development of science and technology (Miraji et al. 2016).

The public awareness of the existence of ECs as significant water contaminants has been increasing. This is mainly

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related to the advancement of analytical instrumentation that enables the detection of ECs at low concentration and their potential toxicity to human and aquatic organisms even in nanogram per liter (ng/L) level (Glassmeyer et al. 2017; Miraji et al. 2016). These contaminants mainly constitute of pharmaceuticals, pesticides, and perfluorinated and endocrine disruptor chemicals which are commonly used in human daily life (Pal et al. 2014). It has been often reported that pharmaceutical compounds are commonly found at 0.15 ng/L–2.0 µg/L in recipient waters downstream of wastewater discharge and about 40–400 µg/L in an agricultural area in China (Norvill et al. 2016). The long-term ecological effects include the loss of habitats and development of microbiological resistance in aquatic ecosystem (Murgolo et al. 2017).

The sources of ECs are usually the effluents from wastewater treatment plants (WWTPs), runoff from agricultural fields, landfill leachate, manufacturing, industrial units, pharmaceutical, clinical, domestic usage and discharge (Murgolo et al. 2017; Norvill et al. 2016). It has been reported that WWTPs effluent is a main route to generate ECs since it is being collected and accumulated from industrial and urban discharge (Arzate et al. 2017; Sangion and Gramatica 2016). Most of the WWTPs comprise of biological, physical, and chemical treatments which are ineffective to treat ECs due to the complex molecular structure, non-biodegradable, and low concentration of the ECs in wastewater (Bolong et al. 2009; Sheng et al. 2016). ECs will not easily being decomposed which results in incomplete removal of the ECs by existing wastewater treatment systems. For instance, Chi et al. (2013)_ENREF_21 reported that about 64% of ECs were discharges by less than 50% while 9% were not removed at all by biological treatment processes. Meanwhile, Caldas et al. (2013) found that several compounds (biocide tebuconazole; pesticides carbofuran, atrazine, and epoxiconzole) were resistant to conventional WTP and shown higher concentration in drinking water than that observed in surface water prior to treatment. This indicated that ECs can easily escape from WTP or WWTP and discharge to environment via direct disposal of wastewater from the industry. Most ECs occurred in WWTP effluent in the concentration range between 1 ng/L and 1 µg/L (Kapelewska et al. 2018). Therefore, advanced technologies on the wastewater treatment process for removal of ECs to provide high-quality water for human and environmental is requisite.

Recently, advanced treatment technologies on advanced oxidation processes (AOPs) such as Fenton oxidation, ozonation, electrochemistry, photocatalysis, sonocatalysis, and sonophotocatalysis are well known to be effective wastewater treatment approaches. Among these technologies, ultrasonic and ultraviolet (UV) irradiations are very attractive due to their simple operation, reduced chemical usage, and a wide range of applications against various types of pollutants (Karaca et al. 2016; Xu et al. 2013). The generation of highly reactive hydroxyl radicals (·OH) and other oxygen species would react with the target ECs rapidly and non-selectively (Eren 2012). The presence of catalyst would enhance the amount of powerful oxidizing agents such as ·OH to initiate and accelerate the degradation rate of any pollutants. Therefore, AOPs have been investigated to eliminate ECs which are resistant to existing WWTPs.

Several reviews have been published with regard to the occurrence, monitoring, and toxicity of ECs (Tijani et al. 2016) as well as biological technologies for the removal of ECs (Ahmed et al. 2017; Ghattas et al. 2017). However, the information concerns to existing and state-of-the-art technologies on photo-, sono-, and sonophotocatalytic treatments to remove ECs in wastewater are still lacking. In this review, the occurrence and source of ECs in water are provided. Besides, summary on elimination of ECs using existing treatment methods via physical methods, i.e., adsorption, filtration; chemical methods, i.e., coagulation, flocculation, precipitation; and biological methods such as activated sludge are included. This review then demonstrates a comprehensive literature review on the performance of emerging AOPs (ultrasonic, ultraviolet, and combination irradiation) in the removal of ECs, their working principles, recent research progress and factors affecting the photo-, sono-, and sonophotocatalytic degradation processes are presented.

Background of emerging contaminants and their environmental impacts

ECs are regarded as the chemicals that had not been detected previously but have been recently discovered widely in water resources. ECs are natural or synthetic compounds that are not commonly regulated with no legal discharging limits are available but known to have potential risk to environmental (Meffe and de Bustamante 2014). ECs are more polar, acidic, and alkaline than natural chemicals when dissolved in water resources. Their hydrophobic properties enable them to accumulate in the lipid-rich tissues and thus being dynamic through food chain (Miraji et al. 2016).

ECs can enter the environment through two routes, i.e., point sources and non-point sources (Tijani et al. 2016). Point sources include effluent from WWTPs, wastewater discharged from household, industries, and hospitals, municipal wastewater and manufacturing of chemicals. Meanwhile, leakage from sewage system, landfills, and agriculture run-off are the examples of non-point sources (Pal et al. 2014). Most of the ECs remain unchanged or transformed into their metabolites and other transformation products after existing WWTPs treatments and will enter the environment depending on their persistency. Usually, their transformation or metabolite products are more harmful and toxic than their original compounds (Petrie et al. 2015).

Various kinds of chemicals that containing manufactured products are unavoidable to satisfy human needs. These products may range from pharmaceuticals, fragrances, cleansing agents, cosmetics, and personal care products. Although there are about 170 ECs from different group classes (Teijon et al. 2010), only priority class of pharmaceuticals, cosmetics and other types of ECs such as pesticides, dioxins, alkylphenolic compounds, and polycyclic aromatic hydrocarbons (PAHs) are discussed in this review paper.

Pharmaceutical products

Pharmaceutical products are manufactured with the purpose of curing and prevention of human diseases by direct ingestion. Besides, most of the pharmaceutical products have been extensively used in aquaculture to prevent parasitic and microbial infections and increase livestock production, respectively (Sangion and Gramatica 2016; Vandermeersch et al. 2015). Mompelat et al. (2009) classified pharmaceutical products into 24 different classes and the common types found in water were antibiotics, anticonvulsants, lipid regulators, and nonsteroidal anti-inflammatory drugs. More than 5000 pharmaceutical products are currently available in the market (Tijani et al. 2016). The largest annual drug consumption countries in 2016 were Brazil, Russia, India, China, and South Africa with about 100–200 thousand tons (Tijani et al. 2016).

Pharmaceutical compounds in the environment were detected due to the discharge of industrial effluents from the production plants, excretion, and waste from medical care centers and direct disposal from each household. The concentration of pharmaceutical compounds in river might reach up to mg/L level and can potentially cause undesirable effects to human and aquatic organisms (Baccar et al. 2012; Cruz-Morató et al. 2014). According to Hughes et al. (2013), 203 pharmaceutical products across 41 countries had been identified in freshwater ecosystem. For example, ciprofloxacin reached a maximum concentration of 6.5 mg/L in freshwater ecosystem. Meanwhile, the concentration of ciprofloxacin in a drug manufacturing factory effluent could reach up to 31 mg/L (El-Shafey et al. 2012).

Besides, carbamazepine, an anti-epileptic drug to control seizures has been frequently found in various environment sources including groundwater, soil, and sediment. Carbamazepine has been detected in groundwater at concentration up to 610 ng/L (Zhang et al. 2008). Generally, carbamazepine may undergo transformation to various aldehydes, ketones, hydroxyl-derivatives, and acridine (a known human carcinogen) (Jelic et al. 2013). It was also reported that another type of antibiotic, sulfamethoxazole, was widely present in surface water with a maximum concentration of about 2000 ng/L (Zhang et al. 2017). The occurrence of resistance bacteria is one of the major challenges which required extra effort in ECs elimination in order to minimize its effect to environment. This issue arises from the increasing of residual antibiotic detected in soil and aquatic environment. The bacterium's gaining resistance genes from antibiotic compound can potentially threaten other living creatures (Ou et al. 2016; Pal et al. 2014). In short, these pharmaceutical compounds might reach human through excretion, runoff from farm, or inappropriate disposal to aquatic environment (Meffe and de Bustamante 2014).

Personal care products

Personal care products and cosmetics are referred to the products used for enhancement of living standards. These products include active and inert substances such as fragrances, shampoo, sunscreen UV filter, cosmetics, antimicrobial agents, insect repellents, and disinfection products like triclosan and chloroprene (Pal et al. 2014; Tijani et al. 2016). Triclosan is one of the most common chemicals used in the production of toothpaste, lotion, deodorant, air freshener, hand soap, and mouth wash (Chen et al. 2011). It has been reported that 5% of triclosan is used in hospitals and 0.1–0.3% by weight concentration of triclosan is used in cosmetic products (Wang et al. 2013). Thus, concentrations in the range of μg/L level have been detected in wastewater effluent (Chen et al. 2011).

Besides, maximum contents of individual and combination of paraben species, which were the main ingredients in most of the cosmetic products, were 0.4% and 0.8%, respectively (Haman et al. 2015). Their reported concentrations were usually within 20–40 mg/L in the raw influent. Methyl-, propyl-, ethyl-, butyl-, and benzyl-parabens had also been discovered in various seafoods at maximum levels of 3600, 1100, 840, 70, and 2.5 ng/g, respectively (Vandermeersch et al. 2015). Wastewater samples collected from a treatment plant in Spain possessed concentrated paraben up to 2.9 μg/L and 0.06 μg/L in influent and effluent, respectively (Jonkers et al. 2009). The occurrence of sunscreen in influent and effluent stream wastewater has been detected at about 500 ng/L and 13.3 mg/L, respectively (Molins-Delgado et al. 2016). Such high concentration may often impoverish the wastewater treatment capacity for removing organic contaminants.

Unlike pharmaceutical products, personal care products are directly applied to human body instead of ingested to change taste, appearance, and odor. Since these products are being applied externally, they might be discharged and leaked easily to the environment without undergoing metabolic changes through washing, bathing, swimming, and domestic wastewater. This might lead to the endocrine system disruption due to the genotoxicity, cytotoxicity, and carcinogenicity of personal care products (Miraji et al. 2016).

Other ECs

Beside pharmaceuticals and cosmetics products, there are also existences of other types of ECs. These include pesticides (El-Shahawi et al. 2010), PAHs (Qiao et al. 2016), dioxins (Hajizadeh et al. 2011), and alkylphenolic compounds (McAdam et al. 2011). Pesticides are extensively used in agriculture to provide protection to crops from pests and diseases (Meffe and de Bustamante 2014; Pal et al. 2014). There are three categories of pesticides: insecticides, fungicides, and herbicides. Insecticides are widely used not only in agriculture but also in forestry, gardening, and landscaping (El-Shahawi et al. 2010). Meanwhile, fungicides are used against fungi and herbicides are mainly used to kill unwanted plants. They can easily leach to the ground through rain wash and irrigation water. Eventually, pesticides are being absorbed by soil and runoff to aquatic environment. Mosleh and Rahimi (2017) reported that more than 99.9% of the pesticides used in agricultural activities were actually dispersed to the environment with only a miniscule amount successfully targeted the pests. Consequently, this could pollute the water resources and bring adverse effects to both human and aquatic organisms.

On the other hand, dioxins and PAHs are produced from the incineration of municipal or industrial solid wastes or incomplete combustion of fossil fuel and petroleum refining (Hajizadeh et al. 2011; Qiao et al. 2016). Dioxins are generated at downstream of the combustion zone with a concentration ranging from 10 to 500 ng/m^3, and it is required to be removed to below 1 ng/m^3 before releasing to environment (Long and Yang 2001). PAHs are ubiquitous in natural environments, including air, water, soil, and sediment (Lu et al. 2011). The European Economic Community (EEC) adopted the second drinking water directive which restricted the discharge concentration of PAHs in water to less than 0.1 µg/L (Zeledón-Toruño et al. 2007).

Besides, alkylphenolic compounds are extensively used as surfactant in the production of paints, textiles, paper, and pulp industries. They also can be found as ingredients in manufacturing hair products, detergent, and pesticides. Alkylphenolic compounds are ubiquitous in wastewater discharges with reported concentrations ranging from 0.05 to 270 µg/L (McAdam et al. 2011). Approximately, 650,000 tons of alkylphenolic compounds had been produced globally every year (Siddique et al. 2016). The presence of alkylphenolic compounds in body can interact with estrogen receptors by mimicking the natural hormones and thus disrupting the endocrine function (McAdam et al. 2011).

Environmental impacts of ECs

The continuous disposal of a large amount ECs remains an unsolved matter. The adverse effects cause by the occurrence of ECs are not being well determined, but several reports have disclosed their potential to accumulate in ecosystem, causing health defects, as well as the ecological risk of ECs to aquatic environment and loss of habitats despite their low concentration (Murgolo et al. 2017). Some of the ECs are able to disrupt the process of synthesis, secretion, binding, transport, and elimination of certain hormones (Tijani et al. 2016). This can interfere with fertility, reproduction, and body homeostasis. Moreover, residence times and depth of water where ECs are present can significantly affect the natural attenuation. Hence, further investigation associated with exposure dose and period is essential to obtain a better insight on their impacts toward human, wildlife, and environment.

Nowadays, in order to improve welfare, pharmaceuticals, and cosmetics which are biologically active are often overused. The discharge of pharmaceutical products into the environment either intentionally or unintentionally might cause adverse impacts to human, wildlife, and ecosystem. The behavior and fate of ECs in water body highly depends on their properties such as adsorption characteristics, persistency, polarity, and volatility. Some of the ECs demonstrate the ability to survive in biological, chemical, or physical processes and remain unchanged after a long period of time. These compounds can accumulate within the body of living organisms and their toxicity and hazardous biological activities are subsequently emerging.

For instance, it has been reported that the prolong exposure to antibiotic contaminated groundwater could result in chronic effects on human beings and ecological systems (Ji et al. 2014). The existence of antibiotic in open water sources may also induce the spread of antibiotic resistance in microorganisms (Ji et al. 2014). Talib and Randhir (2017) reported approximately 4.6 µg/L of ibuprofen was detected in municipal WWTPs at Germany. Meanwhile, acetaminophen, diclofenac, and salicylic acid were found at concentrations ranging from 4.1 to 490 ng/g in marine and freshwater fish (Vandermeersch et al. 2015). Ibuprofen can inhibit the growth of bacteria and fungi, while diclofenac can accumulate in several organs such as liver and kidney (Michael et al. 2014). Hernando et al. (2006) and Michael et al. (2014) investigated the effect of pharmaceutical compounds upon aquatic organisms in term of short acute responses such as lethality in algae and fish. The lowest medium effective concentration (EC_{50}) values shown were highly hazardous to bacteria ($EC_{50} < 1$ mg/L), invertebrates, and algae ($EC_{50} = 1–10$ mg/L). The discharging of ciprofloxacin to environment can cause toxic effects toward human health and stimulate bacterial resistance, which can cause adverse effects to flora, fauna and aquatic ecosystems (Hassani et al. 2017).

Triclosan, 4-chloro-2-aminophenol, and methyl paraben are commonly found in the manufacture of personal care products. Triclosan have been identified in urine and breast milk at concentrations of 19.9 ng/g and 9.3 ng/mL, respectively (Siddique et al. 2016). The potential exposure of triclosan to human is through skin penetration during dermal application and thereby contributes to its presence in breast milk. The detection of triclosan in human breast milk might bring about adverse effects to human at a sensitive life stage and has implication for risk assessment for infants via oral ingestion. Meanwhile, 4-chloro-2-aminophenol may also cause hazardous effects such as fatality via inhalation, ingestion, or skin contact since it is also a well-known toxic organic compound (Barik and Gogate 2016). The presence of methyl- and propyl-parabens at concentrations of 2.92 mg/L and 2.43 mg/L, respectively, were detected in influent of WWTPs through measurements of intact esters of parabens in raw sewage (Velegraki et al. 2015). Methyl parabens have the lowest lipophilicity and thus easily absorbed through human's skin. Although paraben compounds have weak estrogen activity, their abilities to induce or promote MCF-7 breast cancer cells have been proven (Siddique et al. 2016). Molins-Delgado et al. (2016) had conducted acute/chronic toxicity tests of paraben family compounds using aquatic biota. The chronic toxicity hazard quotients of paraben compounds ranged from <0.5 to 3.32 in influent wastewater streams along Catalonia, Spain, indicating the potential risk to aquatic organisms.

Dioxins and related compounds such as biphenyls and polychlorinated dibenzofurans are likely human carcinogens. These compounds are able to induce adverse effects to the immunity systems and fetuses' development (Long and Yang 2001). The negative influence of dioxins related compounds on thyroid hormone metabolism has been widely recognized (Mandal 2005). It was estimated that approximately 60% of alkylphenolic compounds end up in the open water sources via sewage treatment plants (Siddique et al. 2016). Mcadam et al. (2011) reported that the toxicity of alkylphenol polyethoxylates increased with decreasing ethoxylate chain length. The LC_{50} values for long-chain ethoxylates and alkylphenol were 1.5 mg/L and 0.1 mg/L, respectively. In short, alkylphenolic compounds can be introduced into human body through oral intake of contaminated food or leaching from packaging material. As a result, it can increase the risk of female breast cancer and feminization of male.

Bisphenol A is one of the well-known endocrine-disrupting compounds. It is a chemical compound that displays estrogenic activity and commonly used in the production of polycarbonate plastic products (Heo et al. 2012). It has been shown that fetal exposure of 0.025–0.25 µg/kg body weight/day to bisphenol A might result in morphological alterations in both stroma and epithelium of the developing mammary gland (Siddique et al. 2016). Bilgin Simsek (2017) stated that bisphenol A concentration could increase up to 17.2 mg/L in industrial effluents and landfills leachates which containing liquid manures, compost materials and waste plastics. Vandermeersch et al. (2015) reported that the levels of bisphenol A could vary up to 213.1 ng/g for seafood and 224 ng/g for freshwater fish. They proved that bisphenol A could cause reproductive cancers (breast, uterine, testicular, prostate and ovarian), fertility problems (decreased sperm quality and low sperm count), and other endocrine-related consequences. Besides, it had been estimated that 0.55 kg/year of 17β-ethinylestradiol and 15.3 kg/year of 17β-estradiol were conveyed to the Finnish WWTPs (Välitalo et al. 2016). All these endocrine-disrupting compounds are hypothesized to cause an array of diseases, including breast and prostate cancer, neurobehavioral disorders, heart ailments and obesity.

Besides that, PAHs and their derivatives may contribute significant risk to human health as they are commonly known to be toxic, mutagenic and carcinogenic which can induce disruption to endocrine system (Vandermeersch et al. 2015). It has been reported that PAHs compounds are resistance toward biodegradation which result in the detection of their derivatives in the receiving river systems. The derivatives might be more hazardous than their corresponding original compounds (Qiao et al. 2016). These compounds show the potential to bioaccumulate and exhibit high carcinogenic activity. Meanwhile, pesticides can also result in adverse health effects in humans and other animals. The exposure to pesticides would induce chronic health issues such as neurotoxicity, carcinogenesis, and developmental effects (Remucal 2014).

Existing treatment methods

The common WWTPs consist of a primary treatment (physicochemical method) and a secondary treatment (activated sludge biological reactor). The removal efficiency of ECs usually depends on their physicochemical properties, chemical and biological persistency and applied operation conditions. Table 1 summarizes several treatment methods which are commonly reported and their significant findings using the common treatment processes.

Physical methods

Adsorption is a phenomenon of accumulation of substance to the adsorbent surface from a liquid or solid phase through either physical or chemical interaction. Recently, Sophia and Lima (2018) had reviewed a detailed adsorption performance of ECs and their mechanisms. They concluded

Table 1 Several examples of the common treatments applied to the removal of ECs

Treatment process	ECs	Significant findings	References
Physical and chemical methods			
PAC and coagulation	Triclosan	Triclosan was adsorbed by 50 mg/L PAC DETOX 1600 USP resulting in concentration reduction of 92% Resulted in relatively poor removal of less than 10% regardless of the concentration of coagulant (polyalumunium chloride) added from 5 to 20 mg/L	Sheng et al. (2016)
Waste-derived activated carbons	PCDDs	The average removal of 7 types PCDDs were 80, 84, and 84% by using activated carbon derived from fuel, textile and tire, respectively, after 4 days	Hajizadeh et al. (2011)
Waste-derived activated carbons	Atrazine Methyl parathion	The activated carbon was synthesized using ground tire granules It achieved moderately high removal efficiency of 82% and 72% for atrazine and methyl parathion, respectively	Gupta et al. (2011)
Nanofiltration	Atrazine	Three types of membranes were used: aromatic polyamide-based (ESNA), polypiperazine-based (NF200 and NF270) Higher removal of 30.12% was achieved using ESNA as compared to NF200 and NF270 due to stronger hydrophobic interaction	Plakas et al. (2006)
Ultrafiltration	Diuron	By using a polyethersulfate-based membrane, the removal efficiency achieved was between 46 and 55% Membrane made by thin film composite with layer of polyamide was able to remove 10–15% of diuron although it had low molecular cutoff	Acero et al. (2009)
Biological methods			
Biosorption and biodegradation	Triclosan	*Chlorella pyrenoidosa* acted as biosorption sites to triclosan 77.2% of initial concentration of 800 ng/L triclosan was removed in 96 h	Wang et al. (2013)
Biodegradation	Chlorpyrifos	Biodegradation using *Stenotrophomonas sp.G1* removed 42.6% of 50 mg/L chlorpyrifos in 20 h of incubation at 37 °C	Deng et al. (2015)
Biodegradation using fungal bioreactor	Acetominophen Ciprofloxacin Carbamazepine Iopromide	Removal efficiency of acetominophen and ciprofloxacin were 100% and 99%, respectively, using *T. versicolor* fungal bioreactor An increase in concentration of carbamazepine (0.16 µg/L) was observed Only 34% of 419.7 µg/L iopromide was removed as it was metabolically stable	Cruz-Morató et al. (2014)
Biotransformation/biodegradation	Ibuprofen Gemfibrozil Ofloxacin	A total removal of ibuprofen was achieved after 10 days using activated sludge It was found that biodegradation removal of gemfibrozil and ofloxacin were approximate 70% and 25%, respectively, after 35 days	Boix et al. (2016)
Aerobic biodegradation	Triclosan	Initial concentration of 3 mg/L of triclosan achieved 99% removal in 10 days using activated sludge under aerobic condition Intermediate product such as triclosan-methyl which was more persistent and lipophilic than parent compound was detected	Chen et al. (2011)
Biotransformation/biodegradation	APEOs	The removal of APEOs using carbonaceous activated sludge plant was 36.9% after 6 days Removal of 59% APEOs was achieved in nitrifying activated sludge plant after 20 days The lowest removal of 26.8% achieved in nitrifying/denitrifying activated sludge plant after 22 days	McAdam et al. (2011)

Table 1 (continued)

Treatment process	ECs	Significant findings	References
Aerobic biodegradation	Anthraquinone	Total removal of 0.5 µg/L anthraquinone using activated sludge under aerobic condition could be achieved after 7 days Meanwhile, the removal of 5.0 µg/L anthraquinone using activated sludge was about 93–97% after 7 days	Qiao et al. (2016)
Biodegradation	Naproxen	Two freshwater algae: *Cymbella sp.* culture and *S. quadricauda* culture were applied to naproxen biodegradation The removal rate of naproxen by *Cymbella sp.* culture and *S. quadricauda* culture were 97.1% and 58.8%, respectively, after 30 days of incubation	

that the adsorption process involved van der Waals forces of interaction, electrostatic attraction, π–π interactions, and other hydrophobic interactions. Commonly, the process of adsorption was regulated using physical interactions through polarity, steric interaction, π–π p electron donor–acceptor interaction, van der Waals forces, hydrogen bonds, hydrophobicity, and dipole–induced dipole interactions. The solute was first transported by the static fluid film surrounding the adsorbent through the bulk adsorbate movement, and this was followed by the pores diffusion–adsorbate diffusion to the adsorbent through the interaction between the porous surface and the adsorbate.

Adsorption can be considered as a well-established process, but the study of this adsorption method to remove ECs has not been widely explored. Most of the common carbon-based adsorbents include commercial activated carbon (Altmann et al. 2014) and carbon nanotubes (Heo et al. 2012). Adsorbents function as materials to adsorb the contaminant on their porous network surface and subsequently remove the contaminants from wastewater. These materials usually display characteristics of high porosity and large surface area (Rodriguez-Narvaez et al. 2017).

Activated carbon can be differentiated into two major groups with different properties which are powdered activated carbon (PAC) and granular activated carbon (GAC) (Altmann et al. 2014). PAC usually shows higher adsorption capacity than GAC as PAC can achieve higher adsorption capacity due to its higher surface area. PAC is suitable to adsorb majority of ECs, but it usually shows a general preference toward hydrophobic ECs than polar compounds due to the logarithm of octanol–water distribution coefficient (log K_{ow}) (Bolong et al. 2009). Pramanik et al. (2015) found that the adsorption of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) using PAC and GAC was driven by hydrophobic and electrostatic interactions. Higher adsorption performance of PAC compared to GAC was attributed to the smaller particles size of PAC which had greater surface area and shorter internal diffusion distances that favored the adsorption process. On the other hand, Real et al. (2017) explained that PAC with higher mesoporosity could act as transport pores to facilitate the diffusion through porous structure and finally being adsorbed by carbon materials. The lower performance of GAC could possibly relate to the rigidity of CF_2 backbone which hindered the sorption of PFOS and PFOA into the inner pores of GAC.

Meanwhile, the main drawbacks of commercial activated carbon were related to their high production and regeneration costs. According to Sophia and Lima (2018), less than 40% of activated carbon could be regenerated and the regenerated activated carbon was generally not effective for repeated usage. The interest for alternative low-cost adsorbents such as waste materials or by-products derived from agricultural or industrial processes was also growing. Biochar is a product derived from biomass rich in carbon content originating from agricultural by-products (Yao et al. 2012). It is usually produced by pyrolysis heat treatment at temperature from 300 to 1000 °C in the absence of oxygen ($\leq 2\%$). The pyrolysis temperature is a crucial factor that can significantly affect the performance of biochar to remove ECs. Pyrolysis is able to transfigure the biomass materials into high porosity carbon materials with large surface area. Biomass undergoes carbonization whereby the carbon content will increase gradually, while hydrogen, nitrogen and oxygen content decreased with the calcination temperature. At high calcination temperature, oxygen content is being reduced leading to increasing hydrophobicity of the biochar surface. At the same time, the biochar surface also becomes more basic since various acidic functional groups have been eliminated (Ahmad et al. 2012). Consequently, cost-effective biomass adsorbent can be synthesized compared to commercial activated carbon due to lower calcination temperature and do not require additional activating agent.

Biochar could also be applied to soil as conditioner to increase the fertility of the soil and creates a carbon sink to mitigate the severity of carbon emission in the atmosphere (Yao et al. 2012). Meanwhile, biochar has strong sorption ability to control the mobility and reactivity of contaminants in environment. The application of biochar amendment could limit pharmaceutical compounds such as antibiotic sulfamethoxazole and sulfamethoxazole leaching from soil into groundwater (Yao et al. 2012; Zheng et al. 2013). Li et al. (2018) reported that potassium hydroxide-modified biochar derived from potato stems and leaves to reduce the O-consisting functional groups favored the adsorption of ciprofloxacin by hydrophobic interaction and hydrogen bonding.

The removal efficiency is directly related to the adsorbent's properties such as porosity, pore diameter, specific surface area, functional groups, as well as the chemical nature of ECs. Besides, the presence of dissolved organic matter could also influence the effectiveness of adsorption. Pramanik et al. (2015) and Real et al. (2017) reported that the removal efficiency of ECs (PFOS, PFOA, and amitriptyline hydrochloride, methyl salicylate, and 2-phenoxyethanol) decreased in the presence of dissolved organic matters such as humic substances, proteins and acids. The presence of dissolved organic matters might compete the available adsorption site with target contaminant and consequently reduce the adsorption capacity of ECs. Although large volume of low cost biomass feedstock is easily available, it may not be economical as it requires high energy input in order to convert the feedstock to biochar. This adsorption method has poor economic feasibility as it requires the disposal of adsorbed absorbent and thus limits its industry application.

In addition, filtration with membrane technologies such as reverse osmosis, nano-, micro-, and ultrafiltration depending on the pore size and separation mechanism are commonly applied in WWTPs to eliminate ECs. The main benefit of membrane filtration technology is the high quality of the effluent without addition of other chemicals. Although membrane filtration technologies are well-known wastewater application for domestic and industrial wastewater, limited studies have been focused on the eliminating of ECs, especially reverse osmosis and nanofiltration. These techniques are able to reject most of the ECs, but they are energy-intensive processes, which might make them more unfavorable.

Bolong et al. (2009) demonstrated that the dominant mechanisms involved in the removal of ECs through filtration were hydrophobic adsorption between membrane and solutes, size exclusion, and charge repulsion. These three main mechanisms were interrelated to the ECs rejection which in turn affected the removal performances. Hydrophilic molecules showed better rejection rate as compared to hydrophilic molecules due to the hydration of the hydrophilic molecules by forming hydrogen bonds with water molecules. The steric exclusion rejection mechanism implied that the EC compounds with larger size or higher molecular weight than the membrane pore size were being rejected due to the sieving effect of the polymer matrix. Meanwhile, the electrostatic repulsion mechanism relied on relative charge interaction as well as molecular size.

The removal efficiency of atrazine and diuron using membrane filtration was depended on the nature of membrane and the size of ECs (Acero et al. 2009; Plakas et al. 2006). Heo et al. (2012) reported that the mechanisms involved in nanofiltration were hydrophobic adsorption and steric hindrance. Electrostatic repulsion during the filtration could increase the rejection of polar organic compounds. They reported that steric exclusion could be a vital factor if there was a reduction in ultrafiltration membrane pore size. When filtration membrane is governed by size exclusion, the performance of filtration is highly influenced by the molecular weight cutoff (MWCO), which refers to the pore size of the membrane (Acero et al. 2012; Xu et al. 2005). The contaminant with molecular weight above MWCO will retain on the membrane. The MWCO of nanofiltration membrane is larger than 200 Da, while reverse osmosis membrane has MWCO of 200 Da or less (Beier et al. 2010). Meanwhile, Secondes et al. (2014) found that ultrafiltration for removal of diclofenac (log $K_{ow} = 2.45$) was more significant compared to amoxicillin (log $K_{ow} = 0.61$). This indicated that hydrophobic adsorption was the main mechanism for the removal of diclofenac and amoxicillin. On the other hand, Zhu (2015) reported that the rejection mechanism by using nanofiltration membranes of dimethyl phthalate was dominated by steric hindrance instead of hydrophobic interaction due to the low hydrophobicity (log $K_{ow} = 1.6$).

Filtration has the ability to remove acidic and negatively charged contaminant effectively due to the electrostatic repulsion between membrane and ECs (Beier et al. 2010; Xu et al. 2005). However, high-pressure flow is required to force the wastewater to drive through the membrane. Membrane filtration experiences fouling problem easily due to the presence of colloidal particles and dissolved organic matters in wastewater. Therefore, it can be concluded that physical methods such as adsorption and filtration are ineffective in removing all types of ECs as it requires frequent replacement or regeneration of adsorbent and membrane especially when dealing with high concentration of ECs. Besides, the application of physical methods is not able to treat the continuous disposals of ECs load effectively and efficiently. These methods merely transform the contaminants from one form to another, resulting in generation of highly concentrated secondary waste which may pose further disposal issue.

Biological methods

Until now, biological treatment is the most commonly used technology for ECs removal. However, biological treatments can only eradicate about 30–75% of anti-inflammatories and antibiotics (Rivera-Utrilla et al. 2013). In general, biological treatment is separated into two categories: aerobic and anaerobic processes. Several examples of aerobic applications are aerobic bioreactor, membrane bioreactor, trickling filter, and sequencing batch reactor. Membrane bioreactors are activated sludge processes that include an aerobic compartment (for nitrification and carbon removal) and often an anoxic compartment for denitrification. Meanwhile, sequence batch reactors are activated sludge systems where all the steps occur in the same tank by changing the operational conditions. Anaerobic methods include anaerobic film reactors and sludge reactors.

Basically, biological treatment refers to the involvement of microorganisms to degrade the targeted ECs into smaller molecules or further being biomineralized into simple inorganic compounds. It is advantageous over other physical treatments in terms of the operational cost and complexity, provided that the contaminants are readily to be biodegraded or oxidized by the microorganisms. In general, limited information is known about the biodegradation potential of ECs and mechanisms under environmental conditions. Liu et al. (2011) concluded that different electron acceptors present in the compounds might play a key role on the biodegradation process leading to specific biodegradability. Lacking of accurate analytical methodologies is one of the main difficulties for the researchers to identify and quantify the complex matrix. This creates a research opportunity for the exploration of isolation of ECs from biological process by-products.

It was noted that numerous non-biodegradable ECs are ineffective to be removed using biological treatment methods (Ahmed et al. 2017). Most of the ECs are highly hazardous and can disrupt microbial growth which may inhibit the biodegradation. Thus, wastewater characteristics are paramount in the selection of biological treatment. For instance, Deng et al. (2015) found that biological treatment was ineffective to treat pesticides-based pollutants (chlorpyrifos and methyl parathion) as less than 50% removal was recorded after 20 h. Meanwhile, Ternes et al. (2004) reported significant removal rates when ibuprofen was subjected to activated sludge system. However, under similar condition, it was found that carbamazepine was resistant in the system and thus remained as high concentration in effluent. This might be attributed to carbamazepine's poor biodegradability and poor attachment on sludge. Marco-Urrea et al. (2009) reported that the removal of ibuprofen by fungus was completely eliminated after 3 h of incubation and 1,2,-dihydroxy ibuprofen was the major metabolite after 7 d incubation period. However, it was found that 166-fold higher toxicity in water after being treated with white-rot fungi.

Vandermeersch et al. (2015) reported that alkylphenolic compounds exhibited high toxicity and resistance to biodegradation. A detection concentration of 1431 ng/g alkylphenolic compounds was recorded in aquatic environment. Alkylphenol polyethoxylates (APEOs) is one of the examples of alkylphenolic compounds. McAdam et al. (2011) found that nitrifying activated sludge plant to remove APEOs had recorded a biodegradation efficiency of 37%. They suggested that proliferation of microorganism was preferred under nitrification conditions.

Lately, Wang et al. (2017) studied on the sorption and biodegradation of fluoroquinolones, i.e., ciprofloxacin, enrofloxacin, lemofloxacin, norfloxacin, and ofloxacin through batch experiments. The results showed that the removal of fluoroquinolones in activated sludge was governed by sorption process involving bulk mass transfer, liquid film diffusion, and surface reaction which accounted for 78–90% removal. Meanwhile, only small portions (9–22%) of fluoroquinolones were removed via biodegradation process attributed to their poor biodegradability characteristics and long half-lives (> 100 h). Zhang et al. (2017) tried to apply manganese oxidizing bacteria (MOB) in the biological treatment. They reported that MOB was effective to remove sulfamethoxazole at approximately 90% removal rate. However, the application was constrained due to the high cost of manganese carbonate which increased the operating cost significantly. The unavoidable leaching of manganese ions may also cause contamination of the environment (Zhang et al. 2017).

In short, biological treatment is time consuming due to the slow growth rate of microorganism and long period of biological mineralization is required especially when fresh microorganism culture is applied (Mansour et al. 2014). Besides, relatively limited information on the microorganisms involved in the biological degradation process reflected the urgent need for additional fundamental research. Researchers should bear in mind that the generation of biosolids during the application of biological treatment such as activated sludge technologies is an important issue. Past studies had reported significant concentration of hydrophobic, recalcitrant ECs remained in the biosolids after biological treatment (Noguera-Oviedo and Aga 2016). Thus, these biosolids should be carefully handled as it might be another source of ECs released to the environments as the accumulated compounds may desorb after final disposal.

Besides, the usage of biological treatment is time consuming as cultivation of microorganisms is needed prior to the degradation process. Most of the ECs cannot be metabolized by microorganisms, prohibit microorganisms' activity and generate bioaccumulation in the food chain. As such, more effective treatments are required to comply with increasingly strict legislation and reduce the environmental or potential impact toward human beings. Emerging treatments such as AOPs to remove toxic biorefractory ECs are preferred as no sludge will be created. Hence, the following sections focus on the most important homogeneous and heterogeneous AOPs using photo-, sono-, and sonophototechnologies.

Emerging treatment methods: AOPs

In recent years, AOPs have gained much attention as their capabilities in treating wastewater have been proved. The production of non-selective reactive radical species by atoms or molecules containing at least one unpaired electron such as the hydroxyl radical (·OH), superoxide anion radical ($\cdot O^{2-}$), hydroperoxyl radical ($\cdot O_2H$) or alkoxyl radical (·OR). ·OH had attracted the most attention due to its high oxidizing power (oxidation potential, 2.8 V) which is an important phenomenon in elimination of ECs by AOPs (Altmann et al. 2014; Karaca et al. 2016). The degradation rate constants of ·OH to attack ECs through direct photolysis range from 10^7 to 10^{10} $M^{-1}s^{-1}$. For example, ozonation is able to degrade the ECs by direct oxidation with ozone or indirect reaction with the ·OH formed (Bolong et al. 2009). Electrochemical reduction of oxygen (O_2) at cathode could produce hydrogen peroxide (H_2O_2) in acidic medium. Meanwhile, the H_2O_2 produced could oxidize ferrous ion (Fe^{2+}) ions to generate ferric ion (Fe^{3+}) and ·OH, which able to degrade the target contaminants (Mansour et al. 2014). These processes are also known as Fenton and Fenton-like reactions. However, further discussion regarding these treatment methods are not provided in this review paper as the scope is to study the roles of photo-, sono-, and sonophototechnologies to degrade ECs. Such a focus is relatively rare to be reviewed in the literature. Table 2 lists several significant findings of ECs removal by photo-, sono-, and sonophototechnologies.

Ultraviolet (UV) irradiation

UV irradiation is one of the AOPs that have considerable potential in degradation of ECs. This application involves the usage of light with appropriate wavelength and transmits the light photons to attack and degrade the undesired ECs (Burrows et al. 2002). A series of photoinduced radicals are created during photochemical processes (Burrows et al. 2002). Basically, UV spectrum has wavelength range from 100 to 400 nm. It can be further classified into at least 3 groups: UV-A (315–380 nm), UV-B (280–315 nm), and UV-C (200–280 nm) (Zoschke et al. 2014).

UV irradiation using UV light can be considered as potential wastewater treatment due to suitability to treat wide range of ECs, less affected by drastic pH changes and addition of chemical reagents was not required. However, there are some drawbacks on the use of UV light in the application. These include high consumption of energy which leads to high power requirement and frequent replacement of light bulb or tube (Ou et al. 2016). Besides, UV irradiation might be ineffective to treat large volume of system due to limited exposure and penetration of light which limited to several millimeters in water medium (Pang et al. 2011b).

Photolysis

Photolysis is an application involving interaction between UV light and target ECs which induce a series of chemical reactions (Matilainen and Sillanpää 2010). It has been testified that photolysis is a potential remediation method in wastewater treatment plant especially in treating ECs which are recalcitrant pollutants (He et al. 2016). There are two types of UV irradiation, namely direct and indirect irradiations (Rubio-Clemente et al. 2014; Santos et al. 2016). Direct irradiation involves photon absorption by ECs before any photochemical reactions. This process is defined as photosensitized photodegradation (Burrows et al. 2002). As a consequence of being excited by light photons, the ground-state target contaminant (X) is converted to excited state (X*) with higher potential energy [Eq. (1)] by direct irradiation. Subsequently, excited state compound can return back to ground state by releasing the energy [Eq. (2)] or converted into intermediates (radical species) by photochemical reactions [Eq. (3)] (Santos et al. 2016).

Meanwhile, indirect irradiation takes place under the presence of UV-sensitive materials (Lester et al. 2013; Norvill et al. 2016; Remucal 2014). In this process, the degradation is highly depends on the absorption of light by UV-sensitive materials instead of ECs itself (Remucal 2014). Photo-oxidative degradation may happen in the presence of oxygen and react with the intermediates (Yousif and Haddad 2013). Further oxidation process could lead to the degradation of target contaminants into simple products such as CO_2 and H_2O [Eq. (4)] (Rubio-Clemente et al. 2014, Santos et al. 2016).

$$X + h\nu \rightarrow X^* \tag{1}$$

$$X^* \rightarrow X \tag{2}$$

$$X^* \rightarrow \text{Intermediates (Radicals)} \tag{3}$$

$$\text{Intermediates} \rightarrow \rightarrow \text{Photo-degraded products} + CO_2 + H_2O \tag{4}$$

The effectiveness of UV processes for the removal of ECs depends strongly on the UV absorption by ECs. Most of the ECs are photoactive to absorb luminous radiation due to the presence of aromatic rings, heteroatoms, and other functional groups that allowed the absorption of solar radiation and produce photosensitizing species (Rivera-Utrilla et al. 2013). However, when oxidizing agent, i.e., H_2O_2, was

Table 2 Several examples of photo-, sono-, sonophotodegradation of ECs in the absence and presence of catalysts

ECs	Catalyst	Optimum conditions	Significant findings	References
Photolysis and photocatalysis				
Ciprofloxacin	Ag/TiO_2 Au/TiO_2 Cu/TiO_2	[ciprofloxacin]: 3 mg/L [Ag/TiO_2]: 0.5 g/L Light source: UV-C Duration: 3 h Volume: 250 mL	The optimal loading of Ag, Au and Cu on TiO_2 were 1.5, 1.5 and 1.0 wt%, respectively Total removal of ciprofloxacin was achieved by photocatalysis Photocatalytic degradation in the presence of Ag/TiO_2 showed the best result as it achieved total removal in 30 min	Durán-Álvarez et al. (2016)
Acetominophen Diclofenac Ibuprofen	TiO_2	[Acetominophen]: 20 mg/L [Diclofenac]: 20 mg/L [Ibuprofen]: 20 mg/L [TiO_2]: 0.5 g/L Light source: UV-C Duration: 3 h Volume: 150 mL	The photolytic degradation rates of acetaminophen, diclofenac and ibuprofen were 22, 50, and 36%, respectively Photocatalytic degradation rates of acetaminophen, diclofenac and ibuprofen were approximately 42, 92, and 58%, respectively	Eskandarian et al. (2016)
2,4-dichlorophenol Ibuprofen Flurbiprofen	TiO_2 B-TiO_2	[2,4-dichlorophenol]: 20 mg/L [Ibuprofen]: 20 mg/L [Flurbiprofen]: 20 mg/L [TiO_2 or B-TiO_2]: 1 g/L Light source: UV-A Duration: 5 h Volume: 100 mL	The photocatalytic degradation of 2,4-dichlorophenol, ibuprofen and flurbiprofen in the presence of TiO_2 were 75.7, 89.1, and 93%, respectively The photocatalytic degradation of 2,4-dichlorophenol in the presence of with B-TiO_2 for ibuprofen and flurbiprofen were 89.7, 96, and 98%, respectively	Bilgin Simsek (2017)
Acetominophen	TiO_2	[Acetominophen]: 50 mg/L [TiO_2]: 2 g/L Light source: UV-C Duration: 5 h Volume: 250 mL	The photolytic degradation efficiency of acetaminophen was approximately 15% Total removal of acetaminophen was achieved under photocatalytic process in 4 h	Moctezuma et al. (2012)
Ibuprofen	Graphene quantum dot/ $AgVO_3$	[Ibuprofen]: 10 mg/L [TiO_2]: 0.2 g/L Light source: Xenon lamp (420 nm) Duration: 3 h Volume: 50 mL	The graphene quantum dot (GQD) loading were varied from 0, 1, 3, 5 to 7 wt% The photocatalytic degradation efficiencies of ibuprofen in the presence of 0 wt%, 1 wt%, 3 wt%, 5 wt%, and 7 wt%-GQD/ $AgVO_3$ were approximately 20, 70, 90, 60, and 50%, respectively	Lei et al. (2016)

added during photooxidation, the reactivity of ·OH toward ECs would be the predominant mechanism. This indicated that direct photolysis and photochemical processes play important roles in the removal of ECs from water.

Photocatalysis

In order to enhance the performance of photolysis degradation, semiconductors have often been employed as

Table 2 (continued)

ECs	Catalyst	Optimum conditions	Significant findings	References
Sonolysis and sonocatalysis				
Paracetamol	TiO_2 Iron(III) nitrate nanohydrate	[Paracetamol]: 0.09 mM [TiO_2]: 1.0 g/L [Fe^{3+}]: 0.05 mM Duration: 3 h Volume: 250 mL	The removal rates for sonolytic and sonocatalytic degradation of paracetamol were 8.2×10^{-7} M/min and 6.0×10^{-7} M/min, respectively Sonocatalytic degradation of paracetamol obtained a reaction rate of approximately 26.0×10^{-7} M/min	Jagannathan et al. (2013)
Naproxen	ZnO	[Naproxen]: 10 mg/L [ZnO]: 0.5 g/L Ultrasound power: 60 W/L Duration: 2 h Volume: 100 mL	The sonolytic and sonocatalytic degradation rates of naproxen were 20% and 60%, respectively	Karaca et al. (2016)
Ciprofloxacin	Immobilized TiO_2 on MMT	[Ciprofloxacin]: 10 mg/L [TiO_2/MMT]: 0.2 g/L Ultrasound power: 650 W/L Duration: 2 h Volume: 100 mL	The sonolytic degradation efficiency of ciprofloxacin was less than 10% The sonocatalytic degradation efficiency of ciprofloxacin in the presence of TiO_2 and TiO_2/MMT were 40% and 60%, respectively	Hassani et al. (2017)
Sonophotolysis and sonophotocatalysis				
Paracetamol	Iron(III) nitrate nanohydrate	[Paracetamol]: 0.09 mM [Fe^{3+}]: 0.05 mM Duration: 3 h Volume: 250 mL	The degradation efficiencies for sono-Fenton, photo-Fenton and sonophoto-Fenton were 26.1×10^{-7}, 18.6×10^{-7} and 46.7×10^{-7} M/min, respectively	Jagannathan et al. (2013)
	TiO_2	[TiO_2]: 1.0 g/L Duration: 3 h Volume: 250 mL	The degradation efficiencies for sonocatalytic, photocatalytic, and sonophotocatalytic degradation were 6.0×10^{-7}, 30.2×10^{-7} and 40.2×10^{-7} M min^{-1}, respectively.	
Diclofenac Ibuprofen	TiO_2	[Diclofenac]: unknown [Ibuprofen]: unknown [TiO_2]: 500 mg/L Duration: 2 h UV source: UV-A	Sonolytic degradation efficiencies of diclofenac and ibuprofen were 80% and 10%, respectively Photocatalytic degradation efficiencies for diclofenac and ibuprofen were 21% and 65%, respectively Sonophotocatalytic degradation enhanced both diclofenac and ibuprofen to 96% and 85%, respectively	Michael et al. (2014)
Carbamazepine	TiO_2	[Carbamazepine]: 10 mg/L [TiO_2]: 100 mg/L Duration: 2 h UV source: UV-A Volume: 350 mL	The sonolytic, photocatalytic, and combined sonolytic and photocatalytic degradation of carbamazepine were 50, 75, and 82%, respectively	Jelic et al. (2013)

Table 2 (continued)

ECs	Catalyst	Optimum conditions	Significant findings	References
Abamectin	$Cu_2(OH)PO_4$-HKUST-1 MOF	[Abamectin]: 30 mg/L [$Cu_2(OH)PO_4$-HKUST-1 MOF]: 0.4 g/L Light source: LED light (465–470 nm) Duration: 30 min Reactant volume: 500 mL	The sonolytic, photolytic, sonocatalytic, photocatalytic, and sonophotocatalytic degradation of abamectin were 4.38, 10.20, 17.88, 57.46, and 99.93%, respectively	Mosleh and Rahimi (2017)
Ofloxacin	TiO_2	[Ofloxacin]: 10 mg L^{-1} [TiO_2]: 1 g/L Duration: 2 h UV source: UV-A Volume: 300 mL	The sonocatalytic, photocatalytic, and sonophotocatalytic degradation efficiencies of ofloxacin were 62, 85, and 100%, respectively	Hapeshi et al. (2013)

photocatalysts to achieve complete mineralization in the photocatalytic reaction (Bhatkhande et al. 2002; He et al. 2016; Zangeneh et al. 2015). The common photocatalysts contain either single component, i.e., titanium dioxide (TiO_2), zinc oxide (ZnO), tungsten oxide (WO_3), graphene oxide (GO), carbon nanotubes (CNT), graphitic carbon nitride (g-C_3N_4), or combined components. TiO_2 has emerged as one of the most efficient and widely employed photocatalysts due to its high catalytic activity, low toxicity, and good chemical and thermal stability properties. The photocatalysts could absorb light photons to induce chemical reactions for the generation of reactive radicals (Rubio-Clemente et al. 2014).

Figure 1 illustrates the mechanism involved in the formation of electrons (e^-) and holes (h^+) with the aid of catalyst under UV irradiation. Photocatalysis reaction is initiated by the light photons absorption by the target contaminant as well as by the catalyst added into the system (Rubio-Clemente et al. 2014). The energy carried by light photons must exceed the band gap energy of a semiconductor to stimulate the e^- from valence band to conduction band (Leong et al. 2014; Linsebigler et al. 1995). Upon excitation, the separated conduction band e^- and valence band h^+ are created [Eq. (5)] (Matilainen and Sillanpää 2010; Rubio-Clemente et al. 2014). Subsequently, ·OH radical is formed by h^+ trapping via reaction (6) either on catalyst surface or by direct photolysis of target contaminant. Valence band h^+ possesses high positive oxidation potentials (slightly less than 3 V vs. SHE for TiO_2) and therefore can oxidize nearly all chemicals, including the water molecules. Meanwhile, dissolved oxygen is being reduced by conduction band e^- to form superoxide radical ($\cdot O_2^-$) [Eq. (7)] (Chong et al. 2010; Leong et al. 2014; Zangeneh et al. 2015). Both ·OH and $\cdot O_2^-$ are responsible for the degradation of target contaminant [Eqs. (8)–(10)]. Apart from this, $\cdot O_2^-$ can further react with ·OH to form H_2O_2 as shown in reactions (11) to (13) (Chong et al. 2010; Fujishima and Rao 1997).

$$\text{Catalyst} + hv \rightarrow {h_{vb}}^+ + e_{cb}^- \tag{5}$$

$$H_2O + h_{vb}^+ \rightarrow \cdot OH + H^+ \tag{6}$$

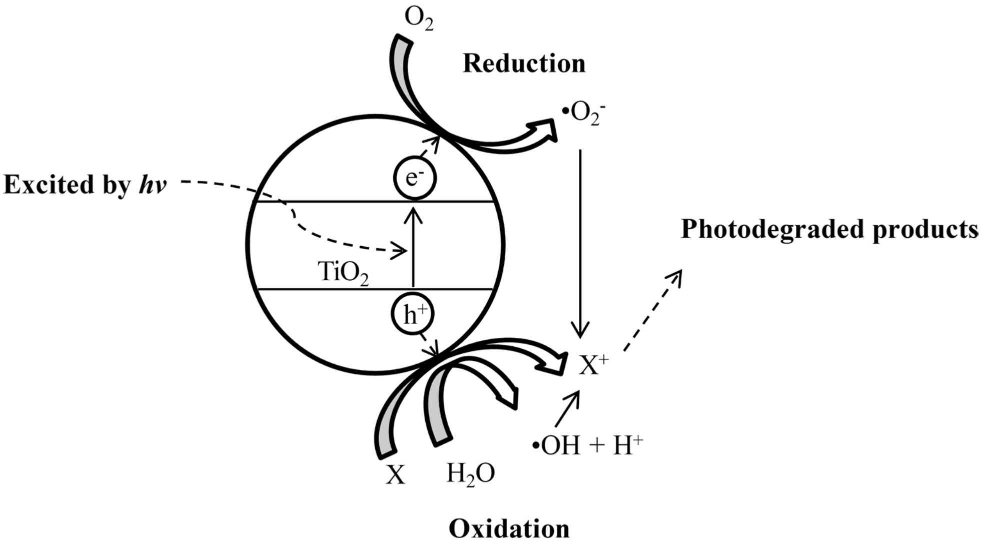

Fig. 1 Mechanism for the photoinduced formation of electron–hole pair in the presence of TiO_2 (Chong et al. 2010)

$$O_2 + e^-_{cb} \rightarrow \cdot O_2^- \quad (7)$$

$$\cdot OH + X \rightarrow \text{Photo-degraded products} + CO_2 + H_2O \quad (8)$$

$$\cdot O_2^- + X \rightarrow \text{Photo-degraded products} + CO_2 + H_2O \quad (9)$$

$$\cdot O_2^- + \cdot OH \rightarrow \cdot OOH \quad (10)$$

$$\cdot OOH + e^-_{cb} \rightarrow HOO^- \quad (11)$$

$$HOO^- + H^+ \rightarrow H_2O_2 \quad (12)$$

The photocatalytic degradation efficiency usually increases with increasing concentration of catalyst until it reaches an optimum concentration (Rubio-Clemente et al. 2014; Zangeneh et al. 2015). The increment in the amount of catalyst would increase the number of active sites on the catalyst surface, which in turn increases the number of reactive free radicals such as hydroxyl and superoxide radicals. Excessive amount of catalyst would inhibit light penetration into the solution which caused catalyst surface to be unavailable for photon absorption. Furthermore, the high concentration of catalyst can also cause reduction in active surface sites due to the particle agglomeration and reduce the homogeneity of suspension and unfavorable light scattering due to high turbidity of the reactants (Leong et al. 2014; Rubio-Clemente et al. 2014; Zangeneh et al. 2015). The optimum amount of catalyst may be different for each type of ECs based on their natural characteristics. Other identified operational parameters that can affect the photocatalytic efficiency include light wavelength, light intensity, catalyst modification, solution pH, and the presence of oxidant.

In literature, Villegas-Guzman and coworkers (Villegas-Guzman et al. 2015a, b) had reported that antibiotic dicloxacillin was persistence to photolysis, slightly degradable by photocatalysis, but highly sensitive to sonochemical degradation. Later, Serna-Galvis et al. (2016) found that high-frequency ultrasound-assisted degradation of the penicillinic antibiotic oxacillin in the presence of mannitol or calcium carbonate additives exhibited the highest performance as compared to photo-Fenton, TiO_2 photocatalysis, and electrochemical oxidation processes. Both phenomena indicated the fact that long wavelength UV radiation may not be able to penetrate the liquid medium and cause any change in the chemical structure of the ECs.

Ultrasonic irradiation

Ultrasound is defined as sound wave which possesses higher frequency outside of human hearing ability. It usually has frequency which varies from 18 kHz to 500 MHz with successive wavelengths of 10–0.01 cm (Thompson and Doraiswamy 1999). The acoustic cavitation is a main phenomenon to cause chemical and mechanical changes in order to accelerate the destruction of the contaminants in liquid (Adewu-Yi 2005). First of all, the propagation of ultrasound involves cycles of compression and rarefaction as shown in Fig. 2 (Adewuyi 2005; Mahvi 2009; Pang et al. 2011a). Acoustic cavitation is created during rarefaction cycle. This event happens when the magnitude of negative pressure is sufficient to pull the water molecules apart. As a result, nucleation sites are formed with trapped gases (Thompson and Doraiswamy 1999). During the compression cycle, the acoustic pressure is positive. Under the influence of positive pressure, the cavitation bubbles grow gradually. Eventually when they reach their critical size, the cavitation bubbles collapse and implosions take place (Mahvi 2009; Pang et al. 2011a). In short, the phenomenon of acoustic cavitation can be represented by a series of nucleation, growth and violent collapse of gas-filled microbubbles produced during compression–rarefaction cycles in the liquid.

For ultrasonic irradiation, no additives or additional chemicals are required to aid this wastewater treatment. Besides, ultrasound can produce wavelength with high penetration distance which can typically stretch up to 20–30 cm from its source and therefore enhances the degradation efficiency (Pang et al. 2011b). It can also be considered as clean and safe process. However, high energy consumption might

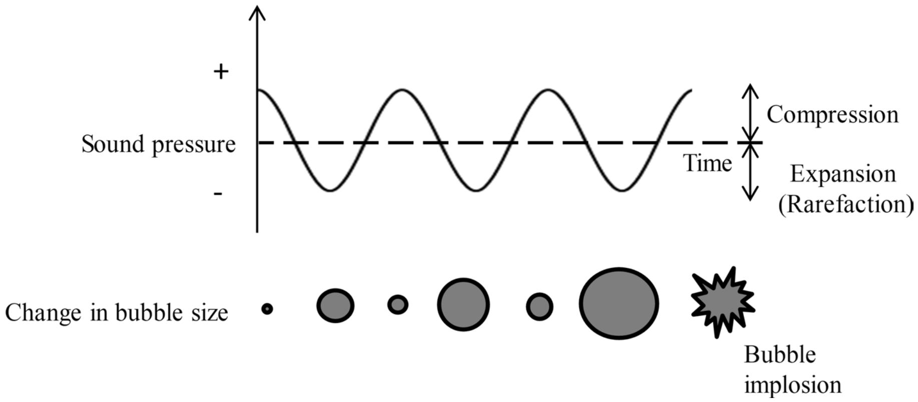

Fig. 2 Cycles of compression and rarefaction in ultrasound

be its main drawback in commercialize ultrasonic irradiation treatment as it requires high operating cost.

Sonolysis

Sonochemistry is the utilization of ultrasound to perform chemical processes with the aid of cavitation bubbles (Adewuyi 2005; Mahvi 2009). The degradation of ECs takes place in three reaction zones (Fig. 3): inside the cavitation bubble, in the interfacial region between cavitation bubble and bulk solution, and in the bulk solution (Eren 2012; Mahvi 2009; Pang et al. 2011a; Thompson and Doraiswamy 1999). When the cavities experience a sudden collapse, the localized hot spot (zone 1) is created. In this region, the temperature and pressure are extremely high. It can reach up to 5000 K and 1000 atm (Eren 2012; Karaca et al. 2016). Under such high temperature and pressure, thermal dissociation occurs to convert water molecules and oxygen into highly reactive species such as ·H and ·OH (Eren 2012; Pang et al. 2011a). The acoustic cavitation enhances the sonochemical degradation by either pyrolysis or radical attack upon implosion [Eqs. (13)–(17)].

$$H_2O +))) \rightarrow \cdot OH + \cdot H \tag{13}$$

$$O_2 +))) \rightarrow 2 \cdot O \tag{14}$$

$$\cdot OH + \cdot O \rightarrow \cdot OOH \tag{15}$$

$$\cdot O + H_2O \rightarrow 2 \cdot OH \tag{16}$$

$$\cdot H + O_2 \rightarrow \cdot OOH \tag{17}$$

The second region is the interfacial region between gaseous- or vapor-phase cavitation bubble and liquid-phase bulk solution (zone 2). Temperature in zone 2 is usually lower than zone 1 which is in the range of 1000–2000 K (Eren 2012). This temperature gradient is due to the difference in temperature between zone 1 and zone 2 which enables the diffusion of radical species from zone 1 to zone 2. Thus, radical attack is the dominating reaction and pyrolysis may occur to a lesser extent in zone 2. In this region, H_2O_2 can be generated as a result of self-recombination of ·OH and ·OOH as described in reactions (18) and (19). The generated H_2O_2 can further react with radicals that diffuse from zone 1 to increase the amount of reactive free radicals such as ·OH and ·OOH in order to enhance degradation efficiency [Eqs. (20)–(21)].

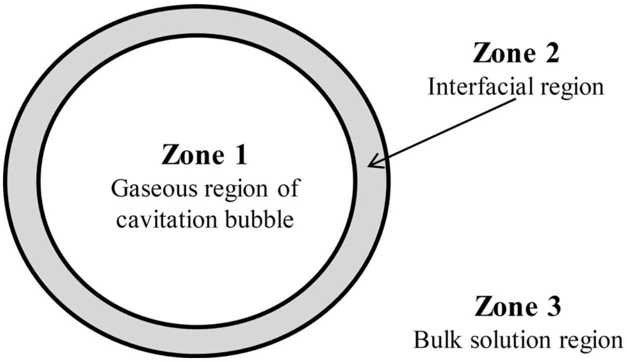

Fig. 3 Three reaction zones in ultrasonically irradiated liquid solution (Thompson and Doraiswamy 1999)

$$2 \cdot OH \rightarrow H_2O_2 \tag{18}$$

$$2 \cdot OOH \rightarrow H_2O_2 + O_2 \tag{19}$$

$$\cdot H + H_2O_2 \rightarrow \cdot OH + H_2O \tag{20}$$

$$\cdot OH + H_2O_2 \rightarrow \cdot OOH + H_2O \tag{21}$$

The third region is the bulk solution (zone 3) at ambient temperature. Under ambient temperature, pyrolysis will not occur but only radicals attack can take place. It has been reported that only approximately 10% of radicals created from interfacial region will diffuse or escape to zone 3 (Thompson and Doraiswamy 1999). Hydrophilic and nonvolatile species are not easily degraded inside the cavitation bubbles. Thus, these compounds are most probably degraded in the bulk solution through the reaction with ·OH radical (Eren 2012; Pang et al. 2011a). Other than that, the unreacted radicals may recombine and release to this region as presented in Eqs. (22)−(25).

$$\cdot OH + \cdot H \rightarrow H_2O \tag{22}$$

$$2 \cdot OH \rightarrow H_2O + \cdot O \tag{23}$$

$$\cdot OOH + \cdot OH \rightarrow O_2 + H_2O \tag{24}$$

$$2 \cdot H \rightarrow H_2 \tag{25}$$

Figure 4 illustrates the generation, growth and implosion of cavitation bubbles to generate radicals to decompose the ECs (Shimizu et al. 2010). The reaction mechanism of sonodegradation of ECs was mainly determined by the physical properties of the ECs through either thermal decomposition (pyrolysis) inside cavitation bubble or free radical reaction mechanism in gas–liquid interfacial region or bulk solution. Thermal dissociation of water, oxygen molecules and related radicals are described in reactions (5) to (9). In these reactions, the ultrasound could be denoted as symbol ')))'. Most of the volatile organic compounds can decompose inside cavitation bubbles either by pyrolysis or by reacting with radical species. Meanwhile, the hydrophilic and nonvolatile compounds are being decomposed mainly in the bulk solution region by reaction with radical species (Eren 2012; Karaca et al. 2016). Thus, sonolysis can treat both hydrophobic and hydrophilic ECs. Chu et al. (2017) reported that hydrophobicity of ECs such as 17α-ethinyl estradiol and bisphenol A significantly influenced the accumulation of ECs at

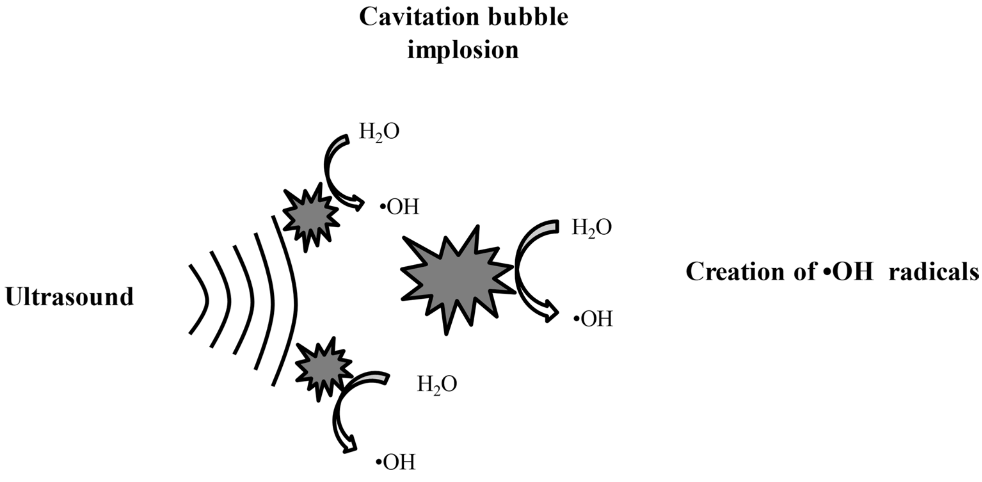

Fig. 4 Schematic diagram of radicals creation from the implosion of cavitation bubbles

the bubble gas–liquid interface to enable the sonochemical degradation of hydrophobic compounds.

Sonocatalysis

Besides the generation of acoustic cavitation, sonochemistry also involved the phenomenon of sonoluminescence (Adewu-Yi 2005). Sonoluminescence is a phenomenon of light flash formation (200–500 nm) with an average photon energy of 6 eV could be produced (Pang et al. 2010). Sonoluminescence has a short lifespan of approximately 100 ps, and it is released from the center core of cavitation bubbles. The origin of light can be caused by plasma emission or luminescence of ·OH depending on acoustic pressure and ultrasonic frequency (Gielen et al. 2016). The emission of light by sonolusminescence acts as the energy source to trigger the catalyst. This light flash is able to provide sufficient energy to excite the e^- from the valence band to the conduction band (Thompson and Doraiswamy 1999). The formation of conduction band e^- reduces the dissolved oxygen while valence band h^+ oxidizes water molecules in order to produce radical species.

In recent studies, the addition of semiconductor as sonocatalyst had been considered as an alternative approach to improve the performance of ultrasonic degradation (Wang et al. 2006, 2013). When the cavitation bubbles implodes in the presence of sonocatalyst, turbulent microjet flow will be created in the system (Adewu-Yi 2005). This turbulent microjet can break down the sonocatalyst into smaller size particles. As a result, it increases the surface contact area, minimizes fouling effect of catalyst and mass transfer resistance, and increases the number of nuclei (Fig. 5). The surface area of catalyst acts as the weak point for the nucleation of cavitation bubbles (Karaca et al. 2016). Subsequently, it increases the number of cavities and radicals created upon implosion of cavities. Larger number of radicals generated can help to promote the transfer of radicals from cavitation bubbles to the bulk solution. High concentration of radicals can promote the degradation of hydrophilic and nonvolatile compounds since they are highly soluble in water (Wang et al. 2008). Hence, the involvement of sonocatalyst has the ability to accelerate the process of ultrasonic irradiation.

The enhancement performance of sonocatalytic degradation of ECs may be due to several factors including the

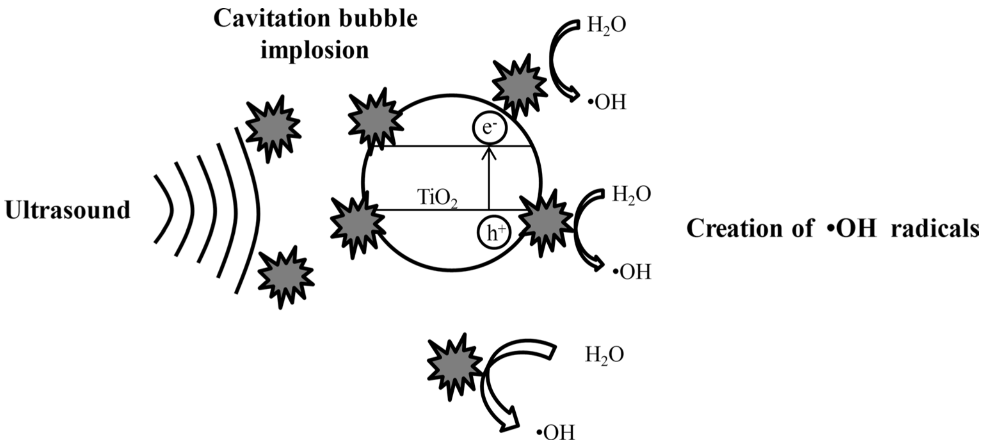

Fig. 5 Schematic diagram of radicals creation under ultrasonic irradiation in the presence of TiO_2

positive characteristic of the semiconductor surface at pH < 7 which resulted to the attractive forces between the positively charged solid surfaces and EC molecules, the improvement in chemical reactivity and mass transfer processes of ECs toward catalyst surfaces and enhanced number of active reaction sites (Chu et al. 2017). However, a complete mineralization is hard to be achieved in most of the cases although sonocatalytic degradation reactions were quite efficient and effective for degradation of ECs. This might be related to higher polarity of the ECs, low availability of OH radicals or lack of dissipated power. Other factors that may affect the degradation efficiency include ultrasonic power, sonication frequency, hydrophobicity of ECs, and ECs volatility and its concentration. Although sonocatalytic degradation does not require additional chemicals, this technique is rather energy demanding which results in low mineralization and is limited to laboratory-scale investigation for now. To overcome all these drawbacks, a combination of AOPs was proposed to obtain benefit from the synergistic effects of the AOPs such as addition source of ·OH while at the same time reduce the operating cost.

Combination of ultrasonic and ultraviolet irradiation

A lot of attention has been concentrating on the application of purely sonolysis and photolysis in wastewater treatment system. However, individual treatment may not be effective in eliminating hydrophilic ECs. Therefore, the combination of AOPs such as photolysis and sonolysis can become a novel and hybrid technique to be explored in the future (Jagannathan et al. 2013; Na et al. 2012b; Park et al. 2014; Xu et al. 2013). Sonophotochemical degradation of ECs can be achieved through simultaneous application of ultrasound and ultraviolet irradiation with or without catalyst. Combination of two oxidation processes advantageously eliminates the shortcoming of both irradiations when applied it solely in the reaction system. To maximize the degradation efficiency, AOPs are either treated sequentially or simultaneously for real or synthetic wastewater.

Sonophotolysis

The combination of ultrasonic and ultraviolet irradiation is commonly known as sonophotolysis. This technique has been widely investigated in eliminating organic contaminants especially coloring dyes and phenolic compounds in wastewater (Park et al. 2014). The studies on sonophotolysis of ECs are still limited. Although ultrasonic and ultraviolet irradiations produce wave energy from different sources, their combination can bring about high synergistic effect. This can be achieved by continuous production of ·OH which are available for chemical reaction. Therefore, by maintaining high concentration of ·OH, the rate of degradation can be significantly increased (Khokhawala and Gogate 2010; Park et al. 2014).

In sonolysis, ·OH radical is produced when the cavitation bubbles experience a sudden collapse. As mentioned earlier, ·OH radicals have short lifetime and easily recombine to produce H_2O_2 (Hamdaoui and Naffrechoux 2008; Park et al. 2014; Xu et al. 2013). By applying hybrid technique of sonophotolysis, H_2O_2 produced could dissociate to ·OH easily when it is exposed to photolysis as compared to sonolysis (Na et al. 2012a, b; Xu et al. 2013). This statement was proven by Na et al. (2012b) in the sonophotolytic degradation of diethyl phthalate which was a source of endocrine disruption in human. They reported that H_2O_2 accumulated during sonolysis and was being consumed rapidly once underwent photolysis process. As a result, the amount of H_2O_2 remained in low concentration under sonophotolysis. This can be explained by generation and consumption of H_2O_2 at the same time by sonolysis and photolysis, respectively. Thus, application of sonophotolysis is advantageously beneficial in generation oxygen reactive radicals for degradation of ECs. As the majority of hybrid and integrated techniques perform superiorly compared to single treatment as shown in Table 2, more studies are being directed toward this research area. The coupling of AOPs treatment in the presence of catalyst has been suggested since it is regarded to be more effective and efficient due to the acceleration of degradation process.

Sonophotocatalysis

Sonophotocatalysis is a combined operation of sonolysis and photolysis in the presence of catalyst to improve and enhance the performance of individual treatment method (Adewu-Yi 2005; Gogate and Pandit 2004). The sonophotocatalysis process includes various AOPs such as photolysis, photocatalysis, sonolysis, sonocatalysis and sonophotolysis. The main aim of the combination of AOPs is to reduce the operational cost and shorten treatment duration. There are three factors to accelerate the sonophotocatalysis which include promotion of mass transfer ECs between the liquid phase and the catalyst surface, stimulation of the ·OH production, and additional nuclei for the formation of bubbles in the presence of catalyst particles. In the application of photocatalysis, catalyst deactivation owning by adsorption of ECs is one of the major concerns. This may be caused by fouling of contaminants on catalyst surface after long operation period and blocking of light activated surface, which in turn reduces the availability of active sites for the degradation of ECs (Khokhawala and Gogate 2010). In order to counter this problem, sonolysis is simultaneously applied with the aim to eliminate the unfavorable activity of blockage by constant cleaning of catalyst surface during photocatalysis

(Adewu-Yi 2005; Gogate and Pandit 2004; Khokhawala and Gogate 2010). This would avoid the accumulation of ECs and their intermediates produced during the degradation.

In addition, mass transfer of target contaminant to active sites especially in the case of immobilized type of catalytic reactors would limit the performance of photocatalysis (Adewu-Yi 2005; Gogate and Pandit 2004). With the aids of sonolysis, a sudden collapse of cavitation bubble creates high temperature and pressure. Under such extreme condition, catalyst will experience fragmentation into smaller particles (Adewu-Yi 2005). This favors the degradation reaction as it provides additional active sites for adsorption–degradation of target contaminant (Adewu-Yi 2005; Joseph et al. 2009). Moreover, both sonolysis and photolysis create highly reactive radicals followed by the reaction between free radicals and the target ECs. Thus, it is anticipated that the coupled operation of sonolysis and photocatalysis provides a superior performance of degradation efficiency (Gogate and Pandit 2004). This is because simultaneous operation of sonolysis and photolysis can concurrently produce ·OH, and therefore, more radicals are available for degradation purpose.

Jelic et al. (2013) explained that the synergistic effect of photocatalysis with sonolysis might contributed by the enhancement of mass transfer rate between the liquid phase of ECs and the solid phase of catalyst. Based on their findings, sonophotocatalytic treatment had improved the degradation of carbamazepine from 7 (photolysis) up to 82% after 120 min. Jagannathan et al. (2013) reported that the combination of ultrasound and ultraviolet enhanced the removal rate of paracetamol due to the increased generation of ·OH from cleavage of H_2O_2. Michael et al. (2014) found that ultrasonic irradiation was ineffective to treat ibuprofen while ultraviolet irradiation was not efficient in degradation of diclofenac. However, when diclofenac and ibuprofen were subjected to simultaneous operation of ultrasonic irradiation and ultraviolet irradiation, the degradation rate greatly enhanced as compared to individual operation. The analysis of toxicity was performed using freshwater species *Daphnia magna* as the control subject. The acute toxicity of the diclofenac and ibuprofen to *Daphnia magna* was monitored and sonophotocatalysis was able to decrease toxicity to 20% after 48 h of exposure. All the results revealed that the combined technology has a superior performance than the individual treatment technology.

In short, drawbacks of catalyst fouling and limited mass transfer in photocatalysis can be improved by application of sonophotocatalysis. It is predicted that sonophotocatalysis has great potential over existing WWTP methods in removal of recalcitrant ECs (Adewu-Yi 2005; Gogate and Pandit 2004). The extent of degradation efficiency of ECs can be affected by a change in the optimal operating parameters. Thus, the fundamental understanding on the basic effect and usefulness of essential operating parameters are vital.

Photo-, sono-, and sonophotochemical degradation under various parameter studies

The photo-, sono-, and sonophotochemical degradation can significantly influence by several factors such as ultrasonic power, ultrasonic frequency, ultraviolet intensity, ultraviolet wavelength, solution pH, addition of oxidizing agents, catalyst dosage, and modification of catalyst.

First of all, the effectiveness of cavitational activity is depends on the dissipation of ultrasonic power. It has been reported that more energy is being released when increasing the ultrasonic power, leading in more violent collapse of cavitation bubbles. This would produce higher solution temperature and pressure during bubble collapse. High ultrasonic power can cause turbulent flow within the system and thus enhances the mass transfer of target EC to the surface of catalyst. Hassani et al. (2017) found that sonocatalytic degradation of ciprofloxacin increased from 53 to 65% when varying the ultrasonic power ranged from 350 to 650 W/L. Similarly, Villaroel et al. (2014) reported that by varying the ultrasonic power from 20 to 60 W could enhance the degradation efficiency of acetaminophen from 20 to 55%.

In another separate study, Teh et al. (2017) proposed that cavitational activity might decreased when the ultrasonic power was adjusted exceeding the optimum value which might due to the coalescence of the cavitation bubbles. Adityosulindro et al. (2017) explained that this situation was due to the scattering effect of sound waves by increment formation of cavitation bubbles and resulting in lower energy released. Besides, this might also due to the increment in the size of cavitation bubble which will cause the incomplete bubble collapse during a single compression cycle. The produced enlarged bubbles might float to the liquid surface and will not affect the degradation process (Ramandi et al. 2017). In fact, a minimum ultrasonic power is required to reach the cavitational threshold which is depends on the ultrasonic frequency (Teh et al. 2017). Unfortunately, very little information on optimum ultrasonic power was being discussed during sonophotochemical degradation of ECs.

In addition, ultrasonic frequency may affect the cavitational effects such as temperature inside bubble, bubble population, lifetime of the bubble and cavitation threshold (Teh et al. 2017). An increase in ultrasonic frequency would result in a decrease in the cavitation bubble size and lower bubble temperature. Eren and Ince (2010) reported that larger critical bubble size, higher collapse temperature and longer bubble lifetime at ultrasonic frequency of 20 kHz as compared to 861 kHz. At low frequency, long bubble collapse duration results in low incidence of bubble collapse. This lead to the reduction of ·OH generation rate and limits the oxidation process. On the other hand, at high ultrasonic frequency, larger number of occurrence bubble collapse even the cavitation bubbles created is small. This improved the bubble collapsing (25-fold) at high ultrasonic frequency of 861 kHz than 20 kHz due to the energy released upon the implosion of cavitation which created larger amount of free radicals (Eren and Ince 2010). However, high ultrasonic frequency will reduce the cavitational effect. This may be attributed to the insufficient duration or the intensity to initiate cavitation during rarefaction cycle of sound wave, or the time for the bubbles to collapse occurs slower than the compression cycle (Adewu-Yi 2005). Consequently, the degradation rate of ECs will decrease if too high ultrasonic frequency is applied due to the reduction generation of reactive radicals. For example, Capocelli et al. (2012) found that sonochemical treatment of estradiols at different ultrasonic frequencies had different efficiency following the order of 850 kH z > 380 kHz > 1000 kHz > 40 kHz. Furthermore, Papadopoulos et al. (2016) had recommended that violent cavitation could be produced at low ultrasonic frequency. This helps to improve the degradation efficiency as the collapse of violent cavitation can increase the localized temperature and pressure. This indicated that determination of optimum frequency is needed to achieve maximum degradation and optimum frequency may vary at different ultrasonic power.

The light intensity is a significant factor that affect to the photo and sonophotodegradation process. Higher light energy could improve the separation of electron–hole pairs, results in production of larger amount of radical species for degradation process. Macawile et al. (2011) had investigated photodegradation of polychlorinated biphenyls under UV light with different power. The results showed that increasing the light intensity from 17 to 24 W would improve the degradation efficiency from 90 to 100%. Similarly, Gar Alalm et al. (2016) reported enhancement results of photocatalytic degradation efficiency of ampicillin and carbofuran with the aid of Ru/WO_3/ZrO_2 catalyst under different light intensities. In the absence of light, less than 10% of both ECs were degraded. The photocatalytic degradation efficiency of ampicillin and carbofuran when subjected to UV flux of 25 W/m^2 were achieved 69% and 76%, respectively. Increasing the UV flux to 50 W/m^2, the degradation performance of ampicillin and carbofuran were further improved to 96% and 100%, respectively. The results proved that the catalyst played a significant role during photocatalytic reaction.

Besides, photo and sonophotodegradation efficiency can also be affected by the wavelength of different light source. In general, different types of light sources release different wavelengths and energies. The common applied light wavelength can be classified as vacuum ultraviolet (185 nm), ultraviolet (254 nm), visible (400 nm) and the combination of them. This is a crucial factor in order to degrade targeted ECs by direct photolysis or to activate photocatalyst. For the direct photolysis without photocatalyst, ECs have excellent light absorption from deep UV region to 460 nm. Eskandarian et al. (2016) reported the direct photolytic degradation efficiency of sulfamethoxazole. The photolytic degradation of sulfamethoxazole was approximately 60, 80 and 100% under UV-A (315 nm), UV-B (280 nm) and UV-C (100 nm), respectively, after 180 min. Meanwhile, Cizmic et al. (2017) found that UV-C displayed better performance than UV-A in the degradation of an praziquantel (anthelmintic drug). Full degradation of praziquantel was completed within 15 min under UV-C irradiation. However, negligible degradation was observed when praziquantel was subjected to photolysis under UV-A irradiation, this might relate to the insufficient energy provided by UV-A. Besides, Eskandarian et al. (2016) found that the effectiveness order for UV-LEDs was UV-C (250–280 nm) > UV-B (295–305 nm) > UVAH (360–370 nm) > UV-A (355–375 nm). This proved that light source with shorter wavelength can provide higher energy and, therefore, enhance the degradation process.

Solution pH is a crucial parameter that plays an important role in determining the charge state of catalyst, size of catalyst aggregate, and availability of ·OH and $\cdot O_2^-$. Meanwhile, the change of solution pH during reaction is highly dependent on the acid strength (pKa) of target ECs and the zero point charge (pH_{zpc}) of catalyst. Rao et al. (2016) had studied the effect of solution pH on sonolytic treatment of carbamazepine. Carbamazepine is a nonvolatile compound and has two pKa values of 2.3 (ketone group) and 13.9 (amino group). They concluded that there was no significant effect to the sonolytic degradation of carbamazepine in the solution pH range from 4.5 to 11. At solution pH 2, carbamazepine exists in ionic form with high hydrophilicity and solubility. Small amount of ·OH was readily available reacted with the carbamazepine in bulk solution, thereby, contributed to a lower degradation performance. Similarly, a lower pseudo-first-order reaction rate was observed when ibuprofen ($pKa = 4.7$) exist in ionic form under alkaline solution (0.0020 min^{-1}) than in molecular form under acidic medium (0.0035 min^{-1}). Therefore, sonochemical degradation of ibuprofen performed better in solution pH 2.6 than pH 8 (Adityosulindro et al. 2017).

Yuan et al. (2016) reported that the N-doping carbon nanotubes/TiO_2 ($pH_{zpc} = 4.4$) exhibited high photocatalytic activity at low solution pH. The catalyst displayed positive surface charge (Ti-OH_2^+) when operating $pH < pH_{pzc}$ and negative surface charge (TiO^-) when operating $pH > pH_{pzc}$, respectively. Ibuprofen tends to dissociate to its conjugate base with negative charge at low solution pH. The negatively charge ibuprofen preferably adsorbed and reacted on the positively charge catalyst and hence promoted the photo-degradation efficiency from 53 to 89%. Similar result obtained from Zhang et al. (2015) mentioned that photocatalytic degradation of ibuprofen favor under acidic condition with the presence of P25-TiO_2/tetraethyl orthosilicate film as photocatalyst ($pH_{zpc}=6.66$). It was found that the initial solution pH usually drops slightly after the oxidation reaction might be related to the formation of intermediate products that may possess various functional groups.

The addition of oxidizing agents or chemical additives is one of the vital parameter in sono-, photo- and sonophotocatalytic degradation of ECs. As demonstrated by Ziylan-Yavas et al. (2015), the addition of persulfate ($S_2O_8^{2-}$) could led to the formation of sulfate radical ($SO_4^{-\cdot}$) by breaking of peroxide bond [Eq. (26)]. Subsequently, $SO_4^{-\cdot}$ reacts with H_2O and OH^- to form ·OH which responsible to degrade the target ECs [Eqs. (27) and (28)]. The positive effect of persulfate was supported by Hassani et al. (2015). They used potassium persulfate in the photodegradation of ciprofloxacin and reported significant improvement in the degradation efficiency from 61 to 97%.

$$S_2O_8^{2-} +))) \quad \text{or} \quad h\nu \rightarrow 2SO_4^{-\cdot} \tag{26}$$

$$SO_4^{-\cdot} + H_2O \rightarrow \cdot OH + H^+ + SO_4^{2-} \tag{27}$$

$$SO_4^{-\cdot} + OH^- \rightarrow SO_4^{2-} + \cdot OH \tag{28}$$

A recent study by Markic et al. (2018) revealed that the addition of $S_2O_8^{2-}$ resulted in a better performance as compared to H_2O_2 during photolysis degradation of 17a-ethinyl estradiol, 17b-estradiol, azithromycin, carbamazepine, dexamethasone, erythromycin, and oxytetracycline in a mixture. Wei et al. (2015) concluded that the reaction rate of pharmaceutical substances levofloxacin followed the order of US/H_2O_2/Fe_3O_4 magnetic nanoparticles $>H_2O_2/Fe_3O_4>$ US/$H_2O_2>H_2O_2>$ US. The mineralization was found to increase with H_2O_2 concentration as more ·OH were formed until reached a maximum concentration [Eq. (29)]. Reactions (30)–(33) may also lead to depletion of H_2O_2 and/or ·OH at the elevated pH levels. An excess of H_2O_2 would cause scavenger effect where H_2O_2 reacted with ·OH to reduce the amount of radicals available to degrade levofloxacin and decrease the mineralization activity.

$$2H_2O_2 +))) \quad \text{or} \quad h\nu \rightarrow 2 \cdot OH \tag{29}$$

$$2H_2O_2 +))) \quad \text{or} \quad h\nu \rightarrow 2H_2O + O_2 \tag{30}$$

$$H_2O_2 +))) \quad \text{or} \quad h\nu \rightarrow OH^{2-} + H^+ \tag{31}$$

$$OH^{2-} + H_2O_2 \rightarrow H_2O + O_2 + OH^- \tag{32}$$

$$\cdot OH + OH^{2-} \rightarrow H_2O + \cdot O_2^- \tag{33}$$

On the other hand, Bagal and Gogate (2014) reported that sodium bicarbonate was an effective hydroxyl radical scavenger to reduce the availability of ·OH for the degradation of diclofenac. Bolobajev and Goi (Bolobajev and Goi 2017) discussed the influence of chemical additives such as deoxyribose, tert-butanol and tetrachloromethane on the sonodegradation of chlorophene. Based on their results, the presence of ·OH scavengers such as nonvolatile deoxyribose and volatile tert-butanol significantly decreased the pseudo-first-order reaction rate from 0.0345 to 0.0024 and 0.0048 min^{-1}, respectively. This situation indicated that deoxyribose and tert-butanol had suppressed the availability of ·OH in the reaction system and indicated the vital role of ·OH in the oxidation process. Meanwhile, enhancement on the sonolytic degradation rate was observed when tetrachloromethane was added. The positive effect relied on the reaction between tetrachloromethane and ·H in which reduced the possibility of recombination of ·H and ·OH.

Davididou et al. (2017) discussed the effect of addition of ferrous ion (Fe^{2+}) and oxalic acid to the photo-degradation of antipyrine. Based on their findings, both Fe^{2+} and oxalic acid exhibited positive effect especially at molar ratio of [oxalic acid]/[Fe^{2+}] maintaining at 3. Reaction between Fe^{3+} and oxalic acid formed ferrioxalate and extra ·OH are generated by ferrioxalate photochemistry. Appropriate amount of Fe^{2+} could form ferric ion (Fe^{3+}), ·OH and $\cdot O_2^-$ via oxidation and Fenton reaction. Meanwhile, excessive amount of Fe^{2+} compete with antipyrine to react with ·OH, which was unfavorable to the degradation of antipyrine.

In addition, the catalyst dosage is also paramount in affecting the overall sono-, photo-, and sonophotodegradation performance. Generally, increasing the catalyst dosage will increase the total available surface area which lead to a more prominent degradation performance and thereby enhance the creation of radical species. Hassani et al. (2015) carried out sonocatalytic and photocatalytic degradation on ciprofloxacin with the presence of TiO_2/montmorillonite catalyst from 0.05 to 0.4 g/L. Sonocatalytic degradation efficiency was enhanced with increasing catalyst dosage up to 0.2 g/L and remained constant thereafter. The highest degradation efficiency of 65% was achieved at optimum dosage of 0.2 g/L catalyst. Meanwhile, photodegradation of ciprofloxacin using 0.1 g/L of TiO_2/montmorillonite catalyst recorded the highest degradation efficiency of 61%. Beyond that, a decreased in degradation efficiency was observed. The adverse effect was expected due to the agglomeration of catalyst by excessive catalyst dosage which scattering the light waves and increase turbidity of solution.

Metal oxide such as TiO_2 and ZnO are commonly being employed as the catalyst in the oxidation degradation process since they have high availability, relatively cheaper, long durability and chemical stability (Lin et al. 2017; Ramandi et al. 2017; Yuan et al. 2016). However, the application of these types of metal oxide is not common due to the fast recombination rate of electron–hole pairs and high band gap energy of 3.2 eV, which are only efficient under

UV light and a small range of solar irradiation (Lin et al. 2017; Ramandi et al. 2017). Therefore, several modification techniques on metal oxide catalyst have been investigated in order to enhance its catalytic activity and reduce its band gap energy.

Metals or nonmetals ions incorporation in the metal oxide catalyst is a promising method as it favors the electron–hole separation and enhances the light absorption property. According to Ramandi et al. (2017), the band gap energy of TiO_2 was successfully reduced from 3.2 to 2.66 eV TiO_2 after doping with nonmetal dopants such as carbon (C), nitrogen (N) and sulfur (S). Degradation of diclofenac under solar irradiation without and with the presence of S–N–C–TiO_2 were 9.44% and 76.48%, respectively. The synergistic effect of each dopant resulted an increasing surface acidity, band gap narrowing, reduction of charge recombination, red shift in adsorption edge and also by modification of its textural properties. Lin et al. (2017) investigated the TiO_2-reduced graphene oxide (TiO_2-rGO) nanocomposites to degrade carbamazepine, ibuprofen and sulfamethoxazole under visible light irradiation. They found that appropriate amount of rGO in catalysts capable to absorb wider light region for both UV and visible light, as well as present faster photocatalytic kinetics. Additionally, rGO can work as an electron acceptor/transporter for TiO_2 particles to enhance the lifetime of electron–hole pairs.

Besides, Hassani et al. (2017) stated that sonocatalytic degradation efficiency of ciprofloxacin employing bare TiO_2 and TiO_2/montmorillonite were 45% and 65%, respectively. Dispersion of TiO_2 particles into the layered montmorillonite structure capable to stabilize TiO_2 particles and extended surface area, increase electron–hole separation and improve the catalytic performance. In addition, Tabasideh et al. (2017) applied 0.4 g/L of bare TiO_2 and iron-doped TiO_2 in sonophotodegradation of diazinon. Based on their findings, iron-doped TiO_2 had better catalytic activity as compared to bare TiO_2 and greatly enhanced the degradation efficiency of sonocatalysis (35% to 50%), photocatalysis (45% to 75%) and sonophotocatalysis (65% to 85%). Small amount of iron ions in the mixed TiO_2 able to prolong the lifetime of electrons and holes by trapping generated electron–hole pairs.

Moreover, Bilgin Simsek (2017) had investigated boron (B)- TiO_2 for the photocatalytic degradation of several types of ECs. Based on their experimental results, the degradation efficiency of TiO_2 and B-TiO_2 for 2,4-dichlorophenol, ibuprofen and flurbiprofen had improved from 75.7, 89.7, and 93% to 89.7, 96, and 98%, respectively, after 4 h of UV-A light irradiation. This proved that boron doping could increase photocatalytic activity of TiO_2 toward ECs. The electron deficient and oxytropic characteristics of B atoms enabled them to function as electron traps to induce the splitting of electron–hole pairs. Besides, boron doping improved crystallinity of anatase TiO_2, increased the amount of superficial hydroxyl groups and decrement in the forbidden band gap energy. However, excess amount of dopant at the surface of metal oxide could notably inhibit the metal oxide from the light illumination and also the interfacial electron and hole to transfer, which would result in a lower catalytic activity.

In addition, Durán-Álvarez et al. (2016) had studied photolysis and photocatalysis degradation of ciprofloxacin using monometallic (Au, Ag and Cu) and bimetallic (Au–Ag and Au–Cu) nanoparticles supported on TiO_2. Photocatalysis achieved a complete mineralization of ciprofloxacin after 90-180 min using either UV-C light (254 nm) or simulated sunlight (290–800 nm). Bimetallic-modified photocatalysts showed the highest mineralization percentage, attributed to the carbonaceous species were efficiently adsorbed onto Au nanoparticles in bimetallic alloys, while the second metal was able to convert dissolved O_2 into reactive oxygen substances. Even though low toxicity was reported after 360 min when using simulated sunlight irradiation, the by-products of ciprofloxacin still expressed antibiotic resistance.

Mosleh and Rahimi (2017) investigated the sonophotocatalytic degradation of abamectin pesticide in the presence of synthesized $Cu_2(OH)PO_4$-HKUST-1 metal–organic frameworks (MOF). The modification of composite and hybrid of copper (Cu)-based materials with MOFs and PO_4^{3-} anions enables the fabrication of an efficient, visible light active, and stable catalyst. The addition of PO_4^{3-} anions enhances the stability of Cu, while MOF improves the crystalline structures with larger surface area. By applying the modified-catalyst into the sonophotocatalysis process, the degradation of abamectin increased from 4.38 to 99.93% after 20 min. The synergism evaluation on the combination of sonocatalysis and photocatalysis led to a synergistic index of 2.19.

Possible reaction pathway and kinetic study for photo-, sono-, and sonophotochemical degradation

Numerous intermediate products are produced during the photo-, sono- and sonophotodegradation process of ECs. Based on the experimental results obtained by Papadopoulos et al. (2016), the sonochemical treatment of ethyl paraben was driven by the reaction with ·OH and forming at least two main intermediate products before complete mineralization. The degradation process was initiated by dealkylation, in which ·OH attacked the ethyl chain of ethyl paraben, forming methyl paraben. Successive dealkylation would transform methyl paraben into 4-hydrobenzoic acid. Two dihydrobenzoic acids with isomers structure (2,4-dihydrobenzoic acid and 3,4-dihydrobenzoic acid) were formed during the hydroxylation of 4-hydrobenzoic acid. Continuous attack of ·OH could further degrade the intermediate products via breaking the benzene ring structure into smaller compounds such as CO_2, H_2O and inorganic molecules.

Jallouli et al. (2016) found that photolysis with or without catalyst can greatly influence the degradation period of naproxen. The degradation process started with decarboxylation of carboxylic group and naproxen was reacted with $\cdot O_2^-$ to form 1-(6-methoxynaphtalen-2-yl) ethyl-hydroperoxide. This intermediate compound further reacted via two routes: reaction with radical of $\cdot CH_3$ or ·OH and transformed into 2-ethyl-6-methoxynaphtalene and 1-(6-methoxynaphtalen-2-yl) ethanol, respectively. Subsequently, 1-(6-methoxynaphtalen-2-yl) ethanol could be dehydrated to form 1-(6-methoxynaphtalen-2-yl) ethanone. All these intermediate products were further oxidized through the opening of ring and reacted with radical species to form maleic acid which could be completely degraded into H_2O, CO_2, and a small portion of non-oxidized products. It is noteworthy to mention that, naproxen and intermediates achieved a complete degradation after 180 min of UV irradiation with the presence of TiO_2. However, naproxen and intermediates were persisted for a longer period (240 min) when subjected to photolysis.

Wu et al. (2016) investigated and reported the sonophotolytic, photolytic and sonolytic degradation of benzotriazole were 100%, 88%, and < 5%, respectively. They found that ultraviolet irradiation played a vital role as it may responsible to initiate the degradation process. Although ultrasonication solely did not perform effectively, the combination of ultrasonic and ultraviolet irradiation showed an enhancement in removal rate. This indicated that the intermediate compounds formed could be broke down thermally and thus induced a complete degradation. Besides sonophotolytic, there are also several researches studied on the sonophotocatalytic degradation and proved that sonophotocatalysis performed better than single application (Tabasideh et al. 2017).

Sunasee et al. (2017) reported that different experimental conditions could affect the generation of intermediate products during sonophotocatalytic treatment of bisphenol A. They identified that five intermediate compounds were formed during sonophotocatalysis in the presence of TiO_2-based catalyst. It was discovered that ·OH was the major contributor for the degradation of bisphenol A to initiate the aromatic rings cleavage, forming hydroxylated and ring-ruptured products. The five intermediate compounds were monohydroxylated bisphenol A, monohydroxylated 4-isopropenylphenol, 4-isopropenylphenol, dihydroxylated bisphenol A, and 4-hydroxyacetophenone. Meanwhile, four intermediate compounds included phenol, p-isopropenylphenol, p-hydroquinone, and 4-hydroxyphenol-2-propanol were identified during photocatalytic degradation in the presence of TiO_2. On the other hand, 2,3-dihydro-2-methylbenzofuran was the only intermediate compound formed during sonolytic degradation.

Rao et al. (2016) found that more than 20 intermediate compounds (epoxide, aldehyde and/or ketone, glycol) were formed during the sonolytic degradation of carbamazepine. This was due to the ·OH attack on benzene ring and olefinic double bond on heterocylic ring. Meanwhile, Durán et al. (2016) had found intermediate compounds such as epoxycarbamazepine, salicylic acid, acridine, anthranilic acid, catechol, and a few organic acids during the sonophotodegradation of carbamazepine. Therefore, it can be concluded that, intermediate products formed can be greatly influence under different experimental conditions.

On the other hand, kinetic model study on the sono-, photo- and sonophotodegradation rate of ECs is essential to achieve high process performance and scaling up the operation. Currently, many researchers conducted study on the reaction kinetic of various types of ECs such as benzotriazole (Wu et al. 2016), bisphenol A (Sunasee et al. 2017), carbamazepine(Rao et al. 2016), diquat (Shibin et al. 2015), diclofenac (Ramandi et al. 2017), and ethyl paraben (Papadopoulos et al. 2016).

In most of the sono- and photochemical degradation cases, heterogeneous Langmuir–Hinshelwood (L–H) kinetic and pseudo-first-order kinetics proposed by Nanzai et al. (2009) are applied to describe the behavior of ECs under various types of degradation processes (Lin et al. 2017; Ramandi et al. 2017). In most of the sonochemical degradation cases, several assumptions are made: (1) rate of adsorption (r_1) of EC at interfacial region is proportional to the concentration of EC at bulk solution, (2) r_1 is also proportional to $(1-\theta)$, where θ represents the occupied ratio of EC in the interfacial region of cavitation bubbles, (3) rate of desorption (r_{-1}) is proportional to θ. It is noted that the rate of r_1 and r_{-1} can be represented by Eqs. (34) and (35).

$$r_1 = k_1 C_0 (1-\theta) \tag{34}$$

$$r_{-1} = k_{-1}\theta \tag{35}$$

where C_0 (mg/L) is the initial concentration of EC, k_1 (min^{-1}) and k_{-1} ($M\ min^{-1}$) are the rate constants for adsorption and desorption, respectively. Meanwhile, θ can be expressed in Eq. (36) after a pseudo-steady state is achieved:

$$\theta = \frac{KC_0}{1+KC_0} \tag{36}$$

where K (M^{-1}) represents k_1/k_{-1} according to Eq. (36).

When the EC in the interfacial region is degraded, the heterogeneous L–H kinetic model is applied as in Eq. (37).

$$r = -\frac{dC}{dt} = k\theta = \frac{kKC_0}{1+KC_0} \tag{37}$$

where, r (mg/L min) represent the initial rate of degradation of EC, k (min^{-1}) and K are the rate constant and equilibrium adsorption coefficient, respectively.

When the concentration of EC in reaction system is low, assumption $KC_0 \ll 1$ is made and thus Eq. (37) can be simplified into pseudo-first-order reaction equation [Eq. (38)]. By rearranging and integration, Eqs. (39) and (40) are generated (Shibin et al. 2015).

$$r = -\frac{dC}{dt} = kKC_0 = k'C_0 \tag{38}$$

$$\ln\frac{C}{C_0} = -k't \tag{39}$$

$$\ln(1 - X) = -k't \tag{40}$$

where k' is the pseudo-first-order rate constant and X is the conversion of EC.

Furthermore, assumption of $KC_0 \gg 1$ can be made when the reaction system with high concentration of EC in which the surface of catalyst is expected to be saturated. Equation (37) is simplified to zero-order rate equation [Eq. (41)].

$$r = -\frac{dC}{dt} = k \tag{41}$$

For sonophotolytic and sonophotocatalytic degradation, synergy [Eqs. (42) and (43)] and synergy index [Eqs. (44) and (45)] are calculated to measure and evaluate the feasibility of combined oxidation processes for the intensification of ECs degradation (Expósito et al. 2017; Mosleh and Rahimi 2017; Xu et al. 2016). The synergistic effect can be calculated by considering the rate constants obtained from the individual treatment. When the value obtained from synergy index is larger than 1, it indicates that positive synergistic effect of the combined individual processes to the degradation of EC (Expósito et al. 2017; Mosleh and Rahimi 2017; Sunasee et al. 2017; Wu et al. 2016).

$$\text{Synergy for sonophotolysis } (\%) = \frac{k_{US+UV} - \left(k_{US} + k_{UV}\right)}{k_{US+UV}} \tag{42}$$

$$\text{Synergy for sonophotocatalysis } (\%) = \frac{k_{US+UV+A} - \left(k_{US+A} + k_{UV+A}\right)}{k_{US+UV+A}} \tag{43}$$

$$\text{Synergy index for sonophotolysis} = \frac{k_{US+UV}}{k_{US} + k_{UV}} \tag{44}$$

$$\text{Synergy index for sonophotocatalysis} = \frac{k_{US+UV+A}}{k_{US+A} + k_{UV+A}} \tag{45}$$

where k_{US}, k_{UV}, k_{US+UV}, k_{US+A}, k_{UV+A}, $k_{US+UV+A}$ represent the reaction rate of sonolysis, photolysis, sonophotolysis, sonocatalysis, photocatalysis, and sonophotocatalysis, respectively, whereas A denotes the type of catalyst.

Conclusion

ECs are currently not commonly regulated as reflected by no maximum discharge limits established. However, the presence of ECs in aquatic environment may cause severe water pollution even at minor concentrations (1 ng/L–100 µg/L). Continuous disposal of ECs to the environment in trace concentration may potentially cause harmful effect on environment and human beings in the long run. Public awareness on the potential risk on health for human and other living creatures has been increasing up due to the high consumption of water in most of the daily activities. Until now, there is still lack of information and data on the sources, transport pathways, behavior and effects of ECs that contributed to the limited or no control of water quality legislation toward ECs in water. In turn, the authorities face the challenges in developing protection policies to assist in the ECs management.

Most of the ECs enter into the environment through the release of effluent from WWTPs. To date, majority of WWTPs are designed for the treatment of phosphorus, nitrogen and colored compounds without catering for ECs. Behavior and complexity of the ECs mixtures are more likely to have greater negative impact than single contaminants. Therefore, further investigations are critically required to ascertain the potential harmful exposure to human, wildlife and environment by evaluating their toxicity to the environment.

Currently, there is still limited knowledge on the complex matrix of biosolid, isolation, and analysis of ECs in the form of biosolid after the common treatment method. The biosolid containing ECs may require additional treatment prior to the disposal. In addition, problem also occurs in determining and quantifying the transformation products of ECs. Some of the transformation products may have higher biological activity than the parent compounds to pose higher toxicity. In order to further study on the transformation products, appropriate analytical method is required to identify and quantify it.

AOPs appear as good alternatives to remove ECs from water, though intermediate products might persist upon long duration time. Several of the intermediate products can possess higher toxicity than their original compounds owing to their high antibiotic activity. Thus, heterogeneous sonocatalysis and photocatalysis have shown great potential to remove and mineralize target ECs and their by-products. Although they are generated from different energy sources, both of them are able to generate reactive oxygen radical species which is responsible for attacking the target ECs and subsequently transform them into harmless and less toxic final products. A quick mineralization of ECs will be the goal for this combined technology to minimize the survival time of toxic intermediate products.

However, ultrasonic and ultraviolet irradiation require high consumption of energy which lead to high operating cost and restricted the industrialization of their application. Researchers may consider the feasibility of incorporating economical light sources such as sunlight. Solar radiation shown as an alternate source instead of using lamps has interesting implications from different points of view including the use of renewable energy. Non-selective reactivity of reactive radicals on non-biodegradable and recalcitrant of ECs indicates that the sonophotocatalytic process can be designed and implemented as a pre-treatment step before subjected to biological treatment. This can improve the biodegradability index and consequently reduce the residence time for the biological treatment. Another interesting research opportunity is the search for novel 'greener' synthetic processes for nanomaterials production. The conventional chemical processes often used toxic chemicals, generate hazardous by-products and potentially release precursor chemicals to the environment. To the best of our knowledge, there are no studies available on the application of biosynthesized nanomaterials for sonophotocatalytic degradation of ECs.

Besides, ultrasonic and/or ultraviolet irradiation alone may not sufficient to achieve high removal efficiency. Thus, appropriate catalyst has been introduced in the process to accelerate and enhance the degradation efficiency. However, the system works only if the semiconductor oxide catalyst is activated by photons. Thus, it is necessary to further investigate and improve the catalytic properties used in the system. These include catalyst activation using wider light spectra, catalyst immobilization strategy to ease the solid–liquid separation, operation system at wider pH range, minimize or eliminate the addition of oxidant additives and effective design of sonophotocatalytic reactor system to reduce the operating cost.

Most studies only focus on the experimental works in artificial systems containing only one compound used as model ECs. Moreover, the volumes treated by current researchers are small and or with rather fixed concentration. The opposite real situation occurs in real contaminated aqueous bodies where the ECs vary in their chemical nature and are probably at high concentration and in high volume. There is still limited knowledge on the assessment of scaling up parameters for solar-, lamp- or ultrasound driven AOPs for industrial application. It would be necessary to perform pilot-plant scale real wastewater containing multiple ECs in different chemical natures to elucidate the mutual effects of these compounds on their removal rate. Besides, more research work related to the degradation kinetics, reaction mechanisms, reactor modeling, and dynamics of the initial treatment on primary ECs and generation of intermediate products are required to be performed.

In conclusion, the combination of heterogeneous sonocatalysis and photocatalysis is postulated to be one of the most promising pathways for developing total mineralization processes of ECs. The monetary investment of the combined process varies will usually vary greatly depending on several important industrial wastewater characteristics. Hence, a complete economic model based on contemporary industrial wastewater treatment should be carried out systematically to assess the total cost required before being implemented widely.

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Compliance with ethical standards

Conflict of interest The authors declare no conflict of interest in the preparation of this review article.

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