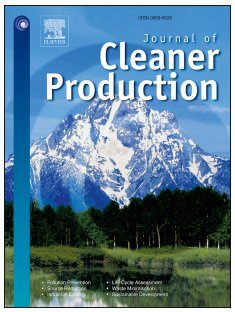

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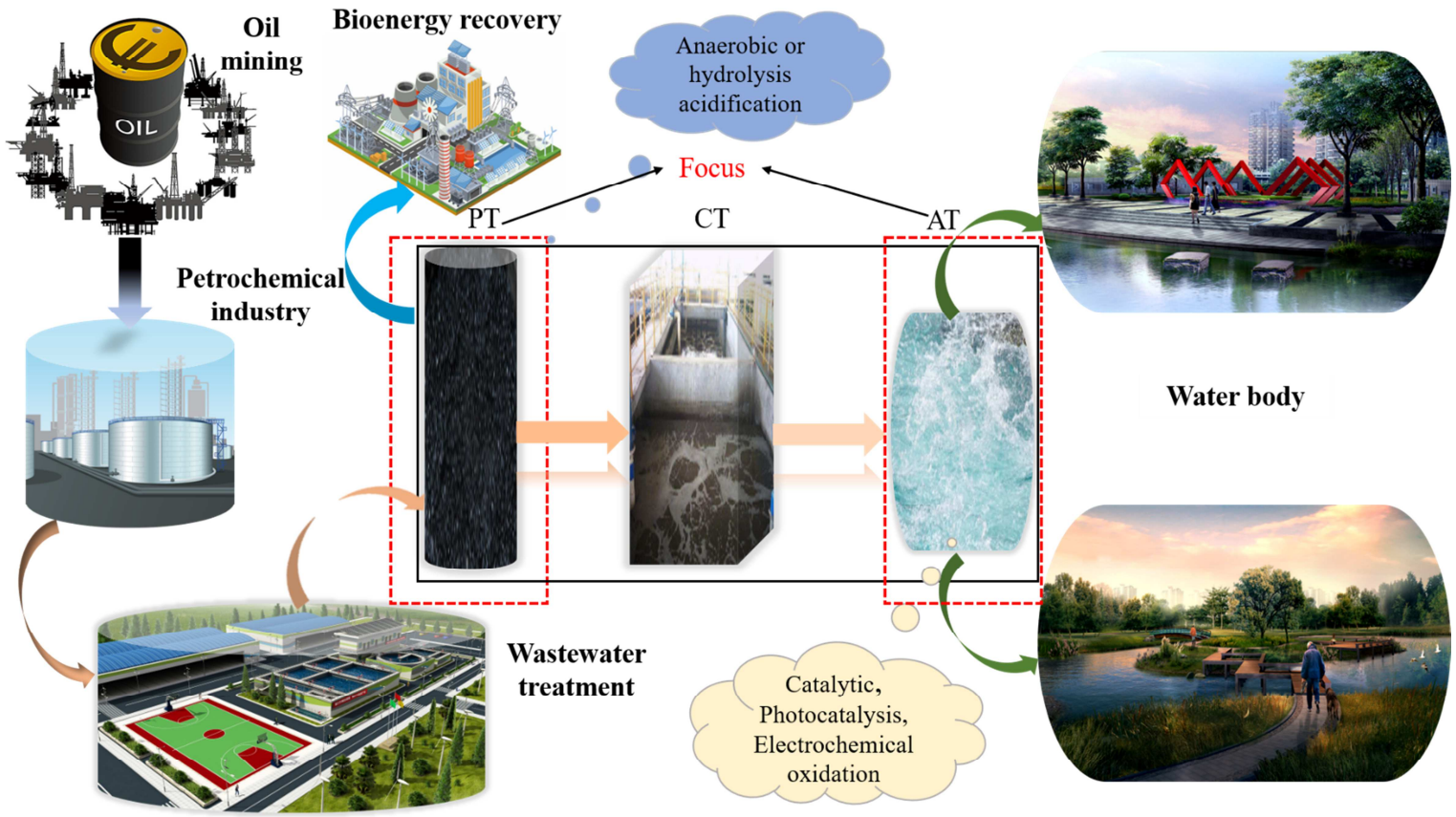
Graphical abstract



A comprehensive review on toxic petrochemical wastewater pretreatment and advanced treatment

Xiangmiao Tian [a, b], *Yudong Song* [a], *Zhiqiang Shen* [a, *], *Yuexi Zhou* [a, *], *Kaijun Wang* [b, *], *Xiaoguang Jin* [a, b], *Zhenfeng Han* [a, c], *Tao Liu* [a, d]

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Abstract: Petrochemical wastewater is a major industrial source of pollution that produces a variety of refractory and toxic organic pollutants that are detrimental to animals and plants in natural water bodies; it is especially harmful to biological treatment systems. Regardless of these threats, studies on these specific organic pollutants are limited at present. Consequently, it is extremely essential to promote relevant problem solving efficiently. Currently, only limited processes are available for pretreatment of high-concentration effluent, and advanced treatment methods as compared to the conventional treatment are introduced by current studies. Therefore, in this review, we have systematically generalized the characteristics of petrochemical wastewater; we have particularly summarized and compared different methods (recent developments, influencing factors, etc.) which are applied in pretreatment and advancement of current methods. Additionally, the interaction mechanisms of microbes under a wide range of concentrations of specific organic pollutants and associated degradation pathways have been described comprehensively. Moreover, we have analyzed bioenergy recovery from the degradation/removal of specific organic pollutants with environmental-friendly and economically methods because it can help realize the goal of circular resource utilization during the process of detoxification and minimization.

Key words: Petrochemical wastewater; specific organic pollutants; pretreatment;

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advanced treatment; bioenergy recovery.

1 Introduction

The petrochemical industry is a fundamental industry that plays an essential role in any country's national economy and provides support to many other sectors, such as agriculture, energy, transportation, etc. (Fig. 1) (Clews, 2016c; Jafarinejad 2017). Petrochemical wastewater is generated via several routes from this industry, including effluent from raw materials, factory rainwater, cooling water, and domestic sewage. It has been reported that global petroleum production has reached 4.40 billion tons based on related industrial statistics (https://www.sohu.com/a/119515720_122917). Moreover, relevant studies have revealed that 3.00-3.50 m^3 of petrochemical wastewater is generated per ton of petroleum refinery process (Zhang and Fan, 2016; Siddique et al., 2017). For instance, China's annual industrial wastewater discharge is 2.10×10^{10} t, of which the proportion of petrochemical wastewater was 3.00-5.00% in 2016. A report derived from China's oil and chemical industry has shown that 2.00 billion tons of petrochemical wastewater were produced only in 2016, and the total emission increased up to 4.00 billion during the 13th 5-year plan period (CMEP, 2015). Moreover, massive petrochemical wastewater was produced all over the world during the same period that needed to be treated properly and urgently.

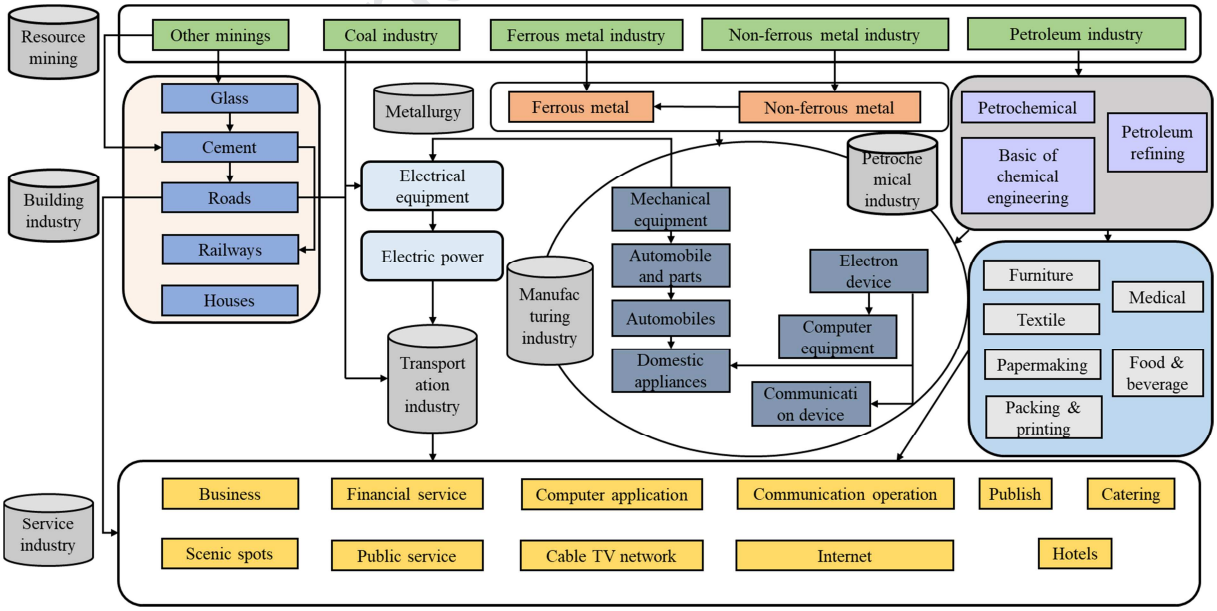

Figure 1 The relationships of petrochemical industry and other industries.

Owing to the use of complicated raw materials, complex processes, and complicated side reactions characteristic of the petrochemical industry, its wastewater generally contains many poisonous substances that are classified as inorganic and organic pollutants. Inorganic pollutants are referred to as heavy metals and other

ingredients (sulfides, fluorides, etc.), and only a few studies have paid attention to their removal and recycling so far (Cechinel et al., 2016; Wei et al., 2016). Organic pollutants are composed of specific organic pollutants, such as benzenes, aldehydes, phenols, etc. (Capello et al., 2009; Kumar et al., 2013), which are considered to be the main problem in petrochemical wastewater because of their high toxicity. Therefore, China implemented a new discharge standard for the petrochemical industry that outlined 60 specific organic compounds (CMEP, 2015). Rapid industrial development often causes petrochemical wastewater to enter natural water bodies; the specific organic pollutants that are released with it in case of the lack of a proper treatment are also carried by this wastewater and prove to be highly poisonous to water bodies, microorganisms, human beings, and ecosystems without proper treatment (Oliveira et al., 2004; Abdullah et al., 2012). For instance, Sponza & Oztekin (2010) discovered acute toxicity of *Daphnia magna* by polycyclic aromatic hydrocarbons (PAHs) released from a petrochemical industry at concentrations as low as 42.6 ng/mL (Yuan et al., 2019) toxicity evaluation methods revealed 2,4-dichlorophenol, formaldehyde, and pyridine originating from petrochemical wastewater to be toxicants. Moreover, some studies have suggested that specific organic pollutants from petrochemical wastewater also pose threats to human health, for example, Iran's workers' exposure to volatile organic compounds from the petrochemical industry made them suffer from cancer (Hajizadeh et al., 2018). Similar studies in China have also reported workers in petrochemical industries to suffer from occupational exposure to PAHs, which put them at a higher possibility of acquiring cancer than a common person (Wang et al., 2015; Wei et al., 2015). Consequently, it is necessary to treat petrochemical wastewater properly and efficiently to meet discharge quality standards before it is allowed to enter the environment.

Immense attention has been paid to conventional treatment of petrochemical wastewater in the last few decades with physical, chemical, and biological technologies being developed for effluent of lower organic loading rate (OLR) and lower toxicity. For example, coagulation−flocculation (Verma et al., 2010; Teh et al., 2016), catalytic ozonation (Zhang et al., 2018b; Huang et al., 2019), submerged membrane bioreactor (sMBR) (Qin et al., 2007), anaerobic expanded granular sludge bed (EGSB) (Liang et al., 2019), microaerobic hydrolysis-anoxic/oxic processes (Yang et al., 2015), microaerobic hydrolysis–acidification–anoxic–oxic processes (MHA–A/O) (Yang et al., 2015) etc. have been extensively applied in conventional

treatment methods. Admirable performance has been achieved in reduction of contaminants and improvement in water quality by conventional treatments, however, some limitations are still present in the optimization of the overall performance. These include the high costs of maintaining equipment's stable operation, strict safety management demands, and sludge disposal problems in physical and chemical methods, while biological wastewater treatment plants may suffer from toxicity. To allow the effective treatment of highly concentrated and toxic petrochemical wastewater so that it can meet stringent discharge quality standards, substantial attempts have been conducted to improve pretreatment and advanced treatment in the recent years. The purpose of advanced treatment is to achieve the partial removal of specific organic pollutants, toxicity reduction, and biodegradability improvement at high concentrations and toxicity levels in petrochemical wastewater before it is subjected to the conventional treatment (Yi et al., 2016; Zhang et al., 2018a; Zheng et al., 2018). Advanced treatment aims to deeply treat wastewater by subjecting it to pretreatment, followed by conventional treatment to satisfy highly stringent quality control demands, for example, soluble microbial products (SMPs), microbial flocs, specific organic pollutants, etc. (Ponce-Robles et al., 2018; Wang et al., 2018). Only a few studies have focused on both pretreatment and advanced treatment in the past because industrial wastewater that is released into municipal wastewater treatment plants is subjected directly to conventional treatment. In recent decades, more studies have explored the possibilities of pretreatment and advanced treatment to meet environmental-friendly and cleaner production requirements. For example, a full-scale bioreactor was adopted by Dao et al. in 2014 to treat highly toxic styrene and propylene oxide (SPO); their results showed that significant detoxicating effect could be achieved by this process. Electrochemical oxidation (ECO) was employed by dos Santos et al. (2014) on petrochemical wastewater to achieve removal of organic matter and COD (92.7%) efficiently. Iron-nickel foam has been used as a catalyst in catalytic oxidization (Huang et al., 2019), which made is possible to remove two-thirds of specific organic pollutants and 96.0% COD in 120 min. Considering the economy, operational safety, and engineering applications, pretreatment and advanced treatment are more capable of employing biological and physicochemical methods of pollutant removal. Nonetheless, the availability of economic and effective processes of pretreatment and advanced treatment are limited and no studies have been conducted to systematically analyze these strategies. Moreover, removal mechanisms

for specific organic pollutants has not been comprehensively reviewed.

Recently, methods of energy-recovery attracted immense attention with regard to recovery of biohydrogen and biomethane from petrochemical wastewater by anaerobic digestion treatments (Luque et al., 2008; Brentner et al., 2010; Rahman et al.,2018). For example, Elreedy and Tawfik (2015) and Elreedy et al. (2018) proposed biohydrogen recovery from petrochemical wastewater to be a feasible, green, and profitable strategy. Siddique et al. (2014, 2015a, 2016) studied biomethane recovery from petrochemical wastewater and achieved the desired biomethane recovery potential. Moreover, they reported that ultrasonic and microwave pretreatment can enhance anaerobic co-digestion to obtain biomethane yields of around 53.0% and 25.0%, respectively (Siddique et al., 2017). For the purpose of pollution reduction and renewable energy generation to alleviate the fossil fuels crisis, bioenergy recovery is becoming a hot research topic.

The fundamental aim of this article is to comprehensively review and compare state-of-the-art pretreatment and advanced treatment technologies of petrochemical wastewater. Moreover, the advantages and disadvantages of different technologies have been summarized and compared systematically. The removal mechanism of specific organic pollutants in petrochemical wastewater by pretreatment and advanced treatment disposal systems has been stated systematically and comprehensively. Furthermore, we have also summarized the process of bioenergy recovery from petrochemical wastewater to aid the realization of a dual purpose of pollutant-reduction methods by enabling bioenergy recovery. In addition, we have discussed feasible future trends of petrochemical wastewater treatment and given some promising viewpoints.

2 Method and analysis section

The recent developments in petrochemical wastewater pretreatment (high-strength) and advanced treatment (low-strength) that have been considered in this review are shown in Fig. 2. In this review, we focused on economical, efficient, and feasible technologies (anaerobic digestion and hydrolysis acidification) for pretreatment. Moreover, important parameters that effect the equipment performance were investigated; these included temperature, pH, HRT, OLR, DO, etc. Moreover, we have systematically reviewed and summarized functional microbes and some removal mechanisms of typical specific organic pollutants in these biological treatment systems. Furthermore, we have reviewed potential methods of high efficiency for

advanced treatment, which mainly included fenton, ozone, catalytic, photocatalysis, and electrochemical oxidation. We have also drawn conclusions about critical factors involved in these processes, such as catalyst types, specific organic pollutant species, catalyst dosage etc. Furthermore, we have explored typical pathways of specific organic pollutant degradation. Additionally, we have presented insight into recovery of bioenergy (biohydrogen or biomethane) from petrochemical wastewater directly, while achieving reduction in pollution and resource recovery. We have summarized other factors related to bioenergy recovery, including time, OLR, pH, and co-digestion of substrates etc. The significance of this work is to provide essential guidance for petrochemical wastewater remediation and to support future work on circular society construction.

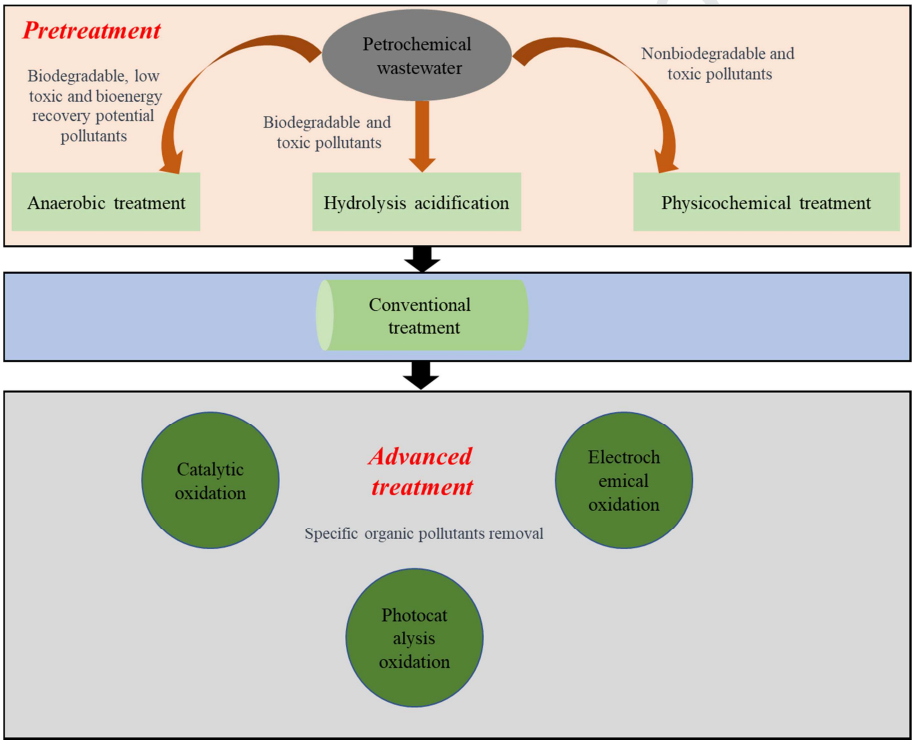

Figure 2 the roadmap of method and analysis section.

3 Petrochemical wastewater pretreatment and advanced treatment

3.1 Pretreatment

3.1.1 Anaerobic treatment

Anaerobic digestion is capable of treating high organic loads and of tolerating high toxicity levels; it can also give low sludge yields from recalcitrant wastewater; therefore, it has been extensively used for industrial wastewater treatment. An anaerobic baffled reactor (ABR) was used by Ji et al. (2009) and Zhang et al. (2011) to treat nutrient-deficient (COD:TN:TP, 1200:15:1) heavy oil-containing wastewater

and acetone–butanol–ethanol wastewater (ABE), respectively. Both studies indicated ABR to have great shock resistance capability. The inhibitory effect increased progressively as the OLR increased during anaerobic digestion (Almendariz et al., 2005), however, the anaerobic digestion still displayed stable and effective performance. The up-flow anaerobic fixed bed (UAFB), up-flow anaerobic sludge blanket (UASB), and anaerobic hybrid reactor (AHR), anaerobic migrating blanket reactor (AnMBR), as well as the ABR, were applied by Ma et al. (2015), Ramakrishnan and Surampalli (2012), and Kuscu and Sponza (2009a, b) to pretreat high-strength petrochemical wastewater with high toxicity levels of specific organic pollutants; results are achieved because of the activity of key enzymes being enhanced at higher temperatures. However, mesophilic (35.0 or 37.0°C) and thermophilic conditions (55.0 ± 3.00°C) can significantly increase anaerobic digestion OLR treatment and reduction in specific organic pollutants (Sreekanth et al., 2009; Majone et al., 2010; Li et al., 2014b); this happens in conjunction with improving microbial activity as compared to EGSB that only degraded 0.290 kg COD/kg in VS/d 2-propanol-contaminted wastewater at 25.0°C (Chang et al., 2005). Furthermore, UASB was used by Liu et al. (2013) and Chen et al. (2017) to treat petrochemical wastewater and to demonstrate that the supported materials have a positive effect on resistance to pH change and acceleration to the treatment process. Moreover, scrap zero-valent iron (SZVI) generated more Fe^{2+}-stimulating protein in the extracellular polymeric substance (EPS); this, in turn, promoted cell aggregation and enhanced methanogenesis using the hydrolysis acidification products for performance enhancement of anaerobic digestion systems (Wang et al., 2017b). Other assisted additives, such as turf soil, were also investigated to enhance UASB performance in COD. Specific organic pollutants removal was reported to be feasible by Chen et al. (2018). Therefore, it is helpful to explore suitable assisted additives for promotion of pretreatment of specific organic pollutants during anaerobic treatment.

3.1.2 Bioenergy recovery from petrochemical wastewater

Nowadays, some researchers pay great attention to bioenergy recovery from high concentration and biodegradable petrochemical wastewater. As compared to anaerobic pretreatment, bioenergy recovery can help ensure dual goals, i.e., pollution reduction and energy recovery, where the bioenergy produced helps offset treatment costs to a certain extent. Currently, bioenergy recovery mainly focuses on biohydrogen and biomethane recovery.

3.1.2.1 Biohydrogen recovery

Recently, bioenergy recovery (biohydrogen and biomethane) from petrochemical wastewater has become a popular trend. Hydrogen is an important intermediate product that is produced during anaerobic digestion (Eqa. 1-8) (Giovannini et al., 2016) with a balance between H_2-producing and H_2-utilizing activities. More hydrogen can be achieved as biohydrogen when parameters controlled at benefiting H_2-producing microorganisms (Khan et al., 2018; Lin et al., 2018). Considering hydrogen is cheap, green, and of a high heating value, some investigations explored hydrogen-producing recovery from petrochemical wastewater treatment (Tab. 1). Parameters involved in biohydrogen recovery are time, OLR, pH and the co-digestion of substrates (Prabakar et al., 2018). Elreedy and Tawfik (2015) investigated ABR from biohydrogen recovery of specific organic pollutants and found that biohydrogen yield increased from 45.5 to 377 mL H_2 /g COD removed as HRT decreased from 70.0 to 18.0 h, Zhu et al. (2010) and Elreedy et al. (2016) also confirmed maximal yields under HRT 6.00 and 9.00 h when PTA and petrochemical wastewater were treated using CSTR and AnPBBR, respectively. This was because methanogens turning H_2 into CH_4 or transforming H_2 and CH_4 into CH_3COOH via homoacetogenesis by homoacetogenic bacteria gradually accumulated at prolonged HRT. OLR also affects biohydrogen production, Elreedy et al. (2018) discovered that biohydrogen yield was 438 ± 43.0 mL/L/d at an OLR of 4.00 g COD/L/d ,and that yield increased from 13 ± 10.8 to 189.1 ± 22.4 mL/g mono-ethylene glycol $(MEG)_{initial}$ when OLR was increased from 1.00 to 4.00 g COD/L/d with AnPBBR. Furthermore, the maximum biohydrogen content attained at this OLR was 47.4 ± 3.60%. However, Elreedy et al. (2015) found that the optimal yield achieved was 359 ± 33.5 $mLH_2/g\ COD_{removed}$ when stepped anaerobic baffled reactor treated petrochemical wastewater was used. Thus, a suitable OLR is necessary to biohydrogen recovery because higher OLR increases hydrolysis acidification, producing more hydrogen; the reduction in hydrogen consumption results from the toxicity of increased specific organic pollutants and accumulated VFAs inhibition of methanogens (Sreethawong et al., 2010). Another important factor, pH, affects extracellular enzyme activity and fermentation pathways, because the optimal pH of hydrolytic bacteria (< 6.00) is lower than that for methanogens (6.80–7.50) (Zhu et al., 2010); clearly, acidic conditions improve biohydrogen recovery. For example, Zhu et al. (2010) confirmed an optimum pH was 4.20-4.40 treated PTA wastewater, Elreedy

et al. (2015) showed optimal pH of 5.23, and Elreedy et al. (2018) depicted the best pH of 6.00 for treating mono-ethylene glycol (MEG) wastewater. Equally important, the effect of the co-digestion substrate should be investigated deeply based on currently limited exploration; a clear example is how Ho et al. (2010) co-degraded phenol and cresol-containing wastewater with *Clostridium sp. R1* with cellobiose providing biohydrogen; they obtained a maximum biohydrogen yield of 3.50 mol H_2 /mol cellobiose at pH 6.00 and 30.0 ˚C.

Determining the microbial community in biohydrogen production is critical. The dominant hydrogen-producing bacteria are related to *Clostridiaceae* (Elreedy et al., 2015), *Clostridium sp. R1*, *Clostridium butyricum* (Ho et al., 2010), *Bacillus*, *Clostridium*, *Desulfovibrionales*, *Ethanoligenens*, *Enterobacter*, *Rhodobacter*, *Thermoanaerobacterium*, and *Thermotogales spp.* (Elreedy et al., 2016), *Clostridium butyricum*, *Lactobacillus casei* (Park et al., 2018). However, the transformation pathway by these microorganisms is still unclear. Based on the economical-technological superiority, treating petrochemical wastewater for pollutant reduction and biohydrogen recovery is a promising technology. Some economic analysis and biohydrogen recovery potential revealed the net profits changed from 14,000.00 to 6.70×10^4 dollars (Elreedy et al., 2018; Elreedy et al., 2016).

$$2CO_2 + 4H_2 \rightarrow CH_3COO^- + H^+ + 2H_2O \ \Delta G=-138.2\ kJ/mol \quad (1)$$

$$2CH_3CHOHCOO^- + H^+ \rightarrow CH_3(CH_2)_2COO^- + 2H_2 + 2CO_2 \ \Delta G=-71.8\ kJ/mol \quad (2)$$

$$2CH_3CHOHCOO^- + CH_3COO^- + H^+ \rightarrow CH_3(CH_2)_2COO^- + CO_2 + H_2 \ \Delta G=-21.3 kJ/mol \quad (1)$$

$$CH_3(CH_2)_2COO^- + H^+ + 2H_2O \rightarrow 2CH_3COO^- + 2H^+ + 2H_2 \ \Delta G=48.1 kJ/mol \quad (4)$$

$$C_6H_{12}O_6 + 2H_2 \rightarrow 2CH_3CH_2COO^- + 2H^+ + 2H_2O \ \Delta G=-308.5\ kJ/mol \quad (5)$$

$$C_6H_{12}O_6 + 2H_2O \rightarrow 2CH_3COO^- + 2H^+ + 4H_2 + 2CO_2 \ \Delta G=-292.3 kJ/mol \quad (6)$$

$$C_6H_{12}O_6 \rightarrow CH_3(CH_2)_2COO^- + H^+ + 2H_2 + 2CO_2 \ \Delta G=-309.4 kJ/mol \quad (7)$$

$$CH_3CH_2COO^- + 3H_2O \rightarrow CH_3COO^- + HCO_3^- + H^+ + 3H_2 \ \Delta G=76.1 kJ/mol \quad (8)$$

3.1.2.2 Biomethane recovery

Biomethane produces from methanogenesis in anaerobic digestion which is described in (Eqa. 9-10). Acetoclastic methanogens utilize CH_3COOH generates CH_4 and H_2 utilizing methanogens use hydrogen and CO_2 is biosynthesized into CH_4.

$$CH_3COOH \rightarrow CH_4 + CO_2 \quad (9)$$

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O \quad (10)$$

Bioconversion processes (mainly anaerobic digestion) provide an excellent possibility to convert containing-rich organic wastes into CH_4, for example from food

waste (Li et al., 2018b) and slaughterhouse waste (Ning et al., 2018). Lately, several studies were carried out to recover biomethane from municipal wastewater and even industrial wastewater, and biomethane was recovered from petrochemical wastewater (Tab. 2). Generally, the methanogens are more sensitive to surroundings in bioconversion system and important factors are initial concentration, temperature, HRT, OLR, pH, and co-substrate.

Initial petrochemical wastewater concentration displays a significant impact on biomethanation as specific organic pollutants have different biotoxication to methanogens. Rahman et al. (2018) found optimal benzene initial concentration was 200 mg/L for biomethanogenesis and that maximal biomethane yield was achieved with anaerobic bioreactor treated benzene-laden wastewater, while inhibition caused when the concentration higher than 300 mg/L. A similar phenomenon was also confirmed by Elreedy et al. (2016), who treated petrochemical wastewater with anaerobic packed bed baffled reactor; Yen et al.(2016) who disposed of PTA with UASB-MBR; and Siddique et al. (2014) who handled petrochemical wastewater with CSTR. Therefore, an appropriate initial concentration is needed to seriously determine different specific organic pollutants and technologies considering the inhibition effect on methanogens. Some investigations were carried out at 15.0 to 55.0 $^{\circ}$C for petrochemical wastewater biomethanogenesis, which showed that temperature has an essential role for methanogens. Many studies conducted at mesophilic condition (20.0 to 40.0 $^{\circ}$C), for instance, Elreedy et al. (2015) recovered CH_4 from MEG at 21.0 $^{\circ}$C, benzene at 35.0 $^{\circ}$C (Elreedy et al., 2015; Rahman et al. 2018), PTA (Yen et al., 2016) at 35.0 $^{\circ}$C and (Zhang et al., 2011) petrochemical wastewater at 40.0 $^{\circ}$C. Their studies firmly affirmed that excellent recovering CH_4 performance achieved at mesophilic conditions. Moreover, some work explored the capability of biomethanogenesis from petrochemical wastewater under thermophilic conditions; Patel and Madamwar (2002) investigated the effect of temperature on CH_4 recovery from petrochemical wastewater in thermophilic condition (45.0, 55.0 $^{\circ}$C), with a maximal CH_4 yield of 0.670 m^3/kg COD/d at 55.0 $^{\circ}$C with OLR of 6.00 kg COD/m^3/d; this was better than at mesophilic condition where more CO_2 produced affected reactor alkalinity at thermophilic condition and seriously affect the stable operation of bioreactor. Therefore, mesophilic conditions are recommended for biomethanogenesis. HRT is another important parameter in biomethanogenesis because it has a significant influence on the microbial community. A number of investigations explored

recovering CH_4 from petrochemical wastewater at HRT (9.00 h to 60.0 d) (Elreedy et al., 2016; Patel and Madamwar, 2002; Zhang et al. 2011) these clearly indicated that prolonging HRT had a positive influence on CH_4 recovery resulted from the slow growth of methanogens; thus, a longer HRT is good for methanogens growth efficiently. Owing to dramatic changes in the different specific organic pollutant toxicity, OLR is also a critical factor for biomethanogenesis. For example, the optimal OLR was 1.67 g COD/L/d (Elreedy et al., 2015), 20.0 g COD/L/d (Yen et al., 2016) and 5.40 kg COD/m^3/d (Zhang et al., 2011) for biomethane recovery from MEG, PTA and petrochemical wastewater, respectively. pH is an important factor in biomethanogenesis as it affects methanogen growth; for example, Siddique et al. (2014) conducted biomethane recovery from petrochemical wastewater at pH 5.45-7.55. Other studies focused on pH 6.12 (Siddique et al., 2016), pH 7.60 (Patel and Madamwar, 2002), benzene at pH 6.50-7.00 (Rahman et al., 2018), petrochemical wastewater at pH 6.50-7.50 (Siddique et al. 2015a), petrochemical wastewater at pH 7.00-7.50 (Siddique et al., 2017). These studies indicated a pH near neutral is optimal for methanogen survival compared to an acidic or alkaline condition. Co-substrate is current research niche for biomethane recovery from petrochemical wastewater (Siddique and Wahid, 2018), such as, glucose (Rahman et al., 2018), dairy and beef cattle manure (Siddique et al., 2014), thickened manure activated sludge (Siddique et al., 2015a) etc. demonstrated co-substrate-adding can significantly ameliorate the petrochemical wastewater performance and improve the biogas production, while the further mechanism should be explored systematically and thoroughly.

The function of methanogens in biomethane from petrochemical wastewater in anaerobic digestion is an important topic to explore. Elreedy et al. (2015) discovered predominant methanogen was *Methanobacterium* (hydrogenotrophic methanogens) at stepped anaerobic baffled (SAB) bioreactor and dominant microbes were *Methylosarcina fibrate*, *Methylophilusmethylotrophus*, *Methylobacteriumisbiliense*, *methylocaldumtepidum*, *Methylocaldumszegendiense*, *Methylocystis spp.* and *uncultured methylobacterium strains* in AnPBBR reactor (Elreedy et al., 2016), (Cheng et al., 2014) *Methanosaeta* (aceticlastic methanogen), *Methanoculleus* and *methanthermobacter* (hydrogenotrophic methanogen), *Methanoculleus*, *Methanocorpusculum*, *Methanobrevibacter*, *Methanobacterium*, and *Methanosarcina* in ABR (Zhang et al., 2011), *Methanosaeta*, *Methanobacterium*, *Methanolinea* and M*ethanogenic archaea* in UASB (Chen et al., 2017, 2018). Some explorations should

be further conducted to search the effect of methanogens in petrochemical wastewater biomethanogenesis considering different technologies and specific organic pollutants.

The abovementioned studies show that high concentration and biodegradable petrochemical wastewater can be pretreated by anaerobic treatment. Furthermore, these specific organic pollutants are used to recover biohydrogen and biomethane for reducing treatment cost through anaerobic treatment. The parameters initial concentration, temperature, HRT, OLR, pH, and co-substrate, especially the type of specific organic pollutants displaying important impact on anaerobic treatment—particularly specific organic pollutants transformation in the bioenergy recovery, the effect of microbes are needed to be explored and the engineering application of bioenergy recovery—still to be improved.

Table 1 Recent studies on biohydrogen recovery from petrochemical wastewater's specific organic pollutants.

Compounds	Initial conc. (COD mg /L)	Processes	Temperature (◦C)	HRT (h)	pH	OLR	Optimal parameters	Hydrogen yield L CH_4 g^{-1} VS	Ref.
Petrochemical wastewater	1.00-6.00×10^3	AnPBBR	15.0-30.0	9.00	-	0.67- 4.00 g COD/L/d	OLR 4.00 g COD/L/d, HRT 9.00 h	438.1 ± 43.0 mL/L/d	(Elreedy et al., 2016)
Petrochemical wastewater	1.50-6.00×10^3	ASBR	15.0-25.0	-	5.50	1.00- 4.00 g COD/L/d	OLR 4 .00g COD/L/d, C/N ratio 28.5, salinity 5.00 g NaCl/L	586 ± 69.3 mL/L/d	(Elreedy et al., 2018)
Ethylene glycol	1.00×10^3	ABR	23.0-27.0	18.0-70.0	5.23± 0.19	333- 1.33×10^3 g COD/L/d	HRT 18.0 h	377 mL H_2/g COD removed	(Elreedy and Tawfik, 2015)
PTA	4.00×10^3	CSTR	35.0	6.00	4.20 - 4.40	16.0 kg COD/m^3 /d	6.30 gVSS/ L, OLR 16.0 kg COD/m^3 /d, HRT 6.00 h temperature 35.0±1.0 ˚C, pH 4.20 - 4.40, alkalinity 280-350mg $CaCO_3$/L and ORP -220 ~ -250. mV (effluent)	0.070 L/g MLVSS/d	(Zhu et al., 2010)
MEG	-	SAB	21.0 ± 6.0	72.0	-	0.330-1.67 g COD L /d	1.67 gCOD/L/d	3.59×10^3 ± 33.5 mL H_2 g COD^{-1}removed	(Elreedy et al., 2015)
Phenol	2.00×10^3	-	30.0		6.00	-	pH 6.00 and 30.0 ˚C	3.50 mol H_2 mol^{-1} cellobiose	(Ho et al., 2010)
Glycerol	1.28 g O_2/g waste	UFCB	35.0 ± 0.500	24.0-48.0	5.30-6.10	8.70 ± 0.50 - 29.7 ± 0.50 0g COD/L /d	-	107 ± 0.700 L/kg waste glycerol	(Dounavis et al., 2015)

1 Table 2 Recent studies on biomethane recovery from specific organic pollutants.

Compounds	Initial conc. (mg/L)	Processes	Temperature (°C)	HRT (h)	pH	OLR	Optimal parameters	Co-substrate	CH_4 yield L CH_4 g^{-1} VS	Ref.
Benzene	200 mg/L	Anaerobic bioreactor	35.0 ± 1.00	-	6.50-7.00	-	-	Glucose	1.80 mM	(Rahman et al., 2018)
Petrochemical wastewater	COD 1.00 - 6.00×10^3	AnPBBR	15.0 – 30.0	9.00	-	0.67-4.00 g COD/L/d	OLR 4.00 g COD/L/d, HRT 9 h	-	238± 21.7 mL/L/d	(Elreedy et al., 2016)
MEG	-	SAB	21.0 ± 6.00	72.0	-	0.330-1.67 g COD L/d	1.67 g COD L/d	-	159 ± 14.7 mL H_2/ gCOD removed	(Elreedy et al., 2015)
Petrochemical wastewater	-	Anaerobic upflow fixed-film	25.0-55.0	1.44×10^3	7.60	3.60-21.7 kg COD/m^3/d	55.0 °C, 6.00 kg COD/m^3/d, pH 7.00, Total alkalinity 4.90-5.19	-	0.670 m^3/kg COD/d	(Patel and Madamwar,2002)
PTA	COD 1.02×10^3 -1.04×10^3	UASB-MBR	35.0	12.0-24.0	6.50–8.50	20.0 g/L/d	35.0 °C, 20.00 g/L/d, HRT 12.0 h	-	66.0 L/ L/ d, CH_4 content 62.0-80.0%	(Yen et al., 2016)
Petrochemical wastewater	1.50×10^4 ± 30.0	CSTR	Mesophilic	36.0-240	5.45-7.55	6.31-25.2 kg COD/m^3/d	6.31 kg COD/m^3/d, HRT 10.0 d, pH 7.55, F/M (g COD/g VSS/ d) 0.290, Total alkalinity (as $CaCO_3$ mg/L) 540 ± 70.0 mg/L	Dairy and beef cattle manure	50.0–60.0%/kg COD	(Siddique et al., 2014)
			Thermophilic						50.0–65.0%/kg COD	
Petrochemical wastewater	15.0 ± 0.080	CSTR	37.0	360	6.50-7.50	-	Flow rate 370mL/d, HRT 9.00 d, COD: N: P 250:5:1, pH 6.80	Thickened manure activated sludge	13.1 ± 0.700 m^3/NB/d	(Siddique et al., 2015a)
Petrochemical wastewater	15.0 ± 0.300	CSTR	37.0	72.0-288	6.12 ± 0.20	1.25 – 5.00 g/L/d	HRT 9 d, pH 6.89 ± 0.090	Activated manure	419 ± 15.0 mL/ $COD_{removed}$	(Siddique et al., 2016)
Petrochemical wastewater	-	Batch	33.0 55.0	-	-	-	-	-	3.70 ± 0.300 and 2.80 ± 0.300 g oil	(Cheng et al., 2014)
Petrochemical wastewater		ABR	40.0 ± 1.00	40.0	-	0.960-5.40 kg COD/m^3/d	OLR 5.40 kg COD m^3/d, COD: N: P 200–300:5:1	Pig manure and rice straw	0.250 L /g $COD_{removed}$	(Zhang et al., 2011)
Petrochemical wastewater	sCOD 8.54 ± 0.030	Batch	37.0	744	7.00 - 7.50	-	HRT 32.0 d	Waste activated slugde	0.220 L CH_4/g VS_{added}	(Siddique et al., 2017)

3.1.3 Hydrolysis acidification treatment and its factors of dependence

Hydrolysis acidification has attracted immense attention for improving wastewater biodegradability and reducing toxicity to microbes in biological processes with metabolic activities enhanced by exoenzymes are secreted from facultative hydrolytic and acidogenic bacteria (Gu et al. 2018, Xie et al. 2016). Wu et al. (2016) explored hydrolysis acidification-anoxic-oxic (HA-A/O) to pretreat petrochemical wastewater, COD removal rate was 88.0% and 87.0% for bench-scale and actual wastewater treatment plants, besides, the toxicity significantly reduced because the major 5 specific organic pollutants were biodegraded essentially, and the wastewater biodegradability improved by 0.13. Moreover, hydrolysis acidification has the possibility to other compounds removal, MHA-A/O treated actual petrochemical wastewater achieved 72.0-79.0% COD (MHA accounted for 33.0-42.0%) and ammonium removal (>94.0%) at HRT 24 h (Yang et al., 2015). Limited-aeration hydrolysis acidification (Wu et al., 2015) pretreated petrochemical wastewater revealed that sulfate eliminated by sulfate-reducing bacteria (SRB) with DO 0.200-0.300 mg/L. Two-stage system hydrolysis acidification coupling with algal microcosms employed by Huo et al., 2018b pretreating acrylonitrile butadiene styrene (ABS) resin manufacturing wastewater, which showed excellent removal of NH_3–N (100.00%) and phosphorus (89.0%) were discovered under pH 6.47–7.45, ORP -110 ± 25.0 mV and temperature 35.0 °C. Thus, hydrolysis acidification is a promising technology for enhancing petrochemical wastewater's biodegradability and removing specific organic pollutants and other pollutants in the future.

3.1.3.1 Temperature

Microbiota survived in wastewater treatment plants needed a suitable temperature to sustain normal growth and metabolic activity, improving temperature properly is highly good for pollutants removal to a certain extent. Li et al., 2014b compared the COD and terephthalate removal in HAR at 33, 37, 43 and 52 °C, their removal rates were 77.4%, 91.9%, 87.4%, 66.10% and 77.6%, 94.0%, 89.1%, 60.8%, respectively, which clearly indicated that enhanced temperature properly facilitated the COD and specific organic pollutants removal was attributed to the key microbial communities improved. Moreover, appropriate increase in temperature can improve key temperature-sensitive enzymes activities, for example, Vermorel et al., 2017 discovered 2-propanol degrading and acetate-utilizing methanogenesis inhibited when temperature under the psychrophilic conditions and aromatic compounds degrading

enzymes showed higher activity at 37.0 ˚C than 35.0 ˚C (Ma et al., 2015). Especially, more studies treated petrochemical wastewater under mesophilic condition (25.0-40.0 ˚C) to guarantee outstanding performance (Huo et al., 2018b; Li et al., 2017a), thus, the mesophilic condition is recommended as reaction temperature in biological treatment petrochemical wastewater.

3.1.3.2 pH

pH (usual 6.00-8.00) plays very essential in wastewater treatment plants steady operation in the biological treatment system, peracidity or peralkaline are detrimental to wastewater treatment plants continuous operation. Chen et al., 2017 discovered peralkaline caused dramatic effect on UASB stable operation when pH adjusted from 7.30 to 9.00, the COD removal declined about 20.0%, particularly important, it took a long time (31.0 d) to recover and stabilize performance. The FT wastewater pH was 3.00 caused a detrimental effect on wastewater treatment plants stable operation, it was extremely necessary to adjust pH to 6.00 for avoiding adverse influence (Wang et al., 2017b). The influent pH was held at 6.50-8.00 to maintain wastewater treatment plants steadily operating during a steady-state period, the pH was adjusted to acidity (2.20) and alkalinity (9.45) aimed at investigating the impact of acid/alkaline shock on wastewater treatment plants, their researches indicated peracidity had no influence on combined system and shorter recovery time at alkalinity condition compared to without turf soil system, therefore, turf soil combined with UASB had better capability to resist pH shock (Chen et al., 2018). Consequently, some efficient measures should be taken to keep the influent pH is in neutral condition for biological treatment.

3.1.3.3 Hydraulic detention time

HRT is one of the parameters significantly affecting wastewater treatment plants performance in biological processes. Minimum HRT was required 4.00 d for phenol effective removal in the three-stage system (Zhao et al., 2009), while optimum HRT was 24.0 h for phenolics pretreated by HUASBs (Sreekanth et al., 2009), which significantly indicated HRT greatly depends on processes and specific organic pollutants types. Usually, the performance reduced as HRT decreased, Ji et al., 2009) confirmed HRT had an essential relationship with oil removal, the oil removal efficiencies both reduced as HRT decreased at different COD loading rates. Phenolics (Ramakrishnan and Surampalli, 2012) and NB (Kuscu and Sponza, 2009b) removal also agreed with the removal decreased with reducing HRT (1.50 to 0.330 d and 10.4

to 2.50 d). From Tab. 3, the HRT ranges from 12.0 h to 8.0 d is ascribed to different specific organic pollutants pretreated via different processes, hence, we can conclude applicable HRT determination should consider specific organic pollutants kinds and technology types seriously.

3.1.3.4 Organic loading rate

OLR refers to the organic pollutants content entering wastewater treatment plants under per volume per time, which expresses the wastewater treatment plants treatment capability measuring generally with COD or BOD and organic pollutants loading. Suitable OLR is beneficial to effectively and economically treat wastewater because lower OLR limits the wastewater treatment plants performance while higher OLR affects its stable running. Almendariz et al., 2005 studies displayed the COD removal decreased from 96.0% to 90.0% as the OLR increased from 1.50 to 1.60 g COD/L/d, 2,4 dichlorophenol (DCP) (Sponza and Uluköy, 2006; Sponza and Cigal, 2008) and phenols (Guo et al., 2015) removed by UASB also confirmed this phenomenon. The reason was the microorganism activity started to be inhibited as OLR increased, and the microbe's tolerance enhanced as the extended adaption for keeping stable performance (Majone et al., 2010). Additionally, the OLR is closely related to process and specific organic pollutant's types, Ramakrishnan and Surampalli, 2012 revealed AHR withstood 4.00 times shock loading better than UASB (2.50 times shock loading). Joung et al., 2009 compared the 4 specific organic pollutants OLR shocks on AHR, which displayed acetic acid and benzoic acid were more critical to AHR stable running than PTA and pTOL because they had higher inhibition effects. To avoid loading shock, sufficient considerations should be taken for specific organic pollutants kinds, removal performance and technology types to determine suitable OLR.

3.1.3.5 Dissolved oxygen

DO has an essential role in biological treatment, proper DO concentration is needed to specific organic pollutants and other substances removal and construction and operation cost reduction. A little DO (0.20-0.30 mg/L) added into hydrolysis acidification is due to enhance hydrolytic and acidogenic bacteria in the hydrolysis acidification (Yang et al., 2015), however, 2.00-3.00 mg/L are needed to realized overall performance by HA-A/O in the full-scale wastewater treatment plants (Wu et al., 2016). Importantly, DO increase inhibits sulfate reduction (Eqa. 11) by enhancing SRB diversity and richness (Wu et al., 2015; Yang et al. 2015. Gu et al. 2018 discovered DO affects NO_2-N, NH_4-N removal under different DO concentration,

NO_2-N accumulated in anoxic and aerobic phase at DO which demonstrated nitrobacteria (NOB) inhibited at low DO concentration (1.00 and 2.00 mg/L), whereas no NO_2-N could be detected at DO 3.00 and 7.00 mg/L, moreover, NH_4-N removal enhanced as the DO concentration increased. Therefore, although adding or improving DO for satisfying efficiently some specific organic pollutants removal, however, it also furtherly considers other inorganic substances effective removal and toxic gas generation avoidance simultaneously.

$$SO_4^{2-} + ATP + 8e^- + 10H^+ \rightarrow H_2S + 5H_2O + 2Pi + AMP \quad (21)$$

3.1.3.6 Key microbes in pretreatment

Microbes contained sludge aggregates displayed a crucial role with degrading specific organic pollutants into low toxicity or micromolecule substances and utilized by microorganisms through metabolism and anabolism. specific organic pollutants removal has a positive relationship with microbial species; therefore, focuses have been paid on hydrolysis acidification and anaerobic digestion microbes resulted from previous studies. The predominant genera were *Anaerolineaceae* and *Sulfuritalea* in MHA (Yang et al., 2015), while *Acidobacteria* > *Proteobacteria* > *Bacteriodetes* (Wu et al., 2016). However, *Proteobacteria*, *Chloroflexi*, *Firmicutes*, *Bacteroidetes*, *Planctomycetes*, *Acidobacteria*, *Deferribacteres,* and *Actinobacterium* existed (Wu et al., 2015) under the phylum level, *Chloroflexi*, *Proteobacteria* and *Bacteroidetes* in the phylum level and *Anaerolineaceae* uncultured and *Desulfobacte*, *Blastocatella* and *Anaerolineaceae uncultured*, *Saprospiraceae uncultured* and *Nitrosomonadaceae uncultured* were dominant genera in hydrolysis acidification (Ding et al., 2016) differed from limited aerated hydrolysis acidification. In the phylum level, predominant microbes discovered in anaerobic digestion were *Proteobacteria* (anaerobic biofilm) (Li et al., 2017a), *Clostridia* (ABR) (Ji et al., 2009), *Proteobacteria*, *Chloroflexi* and *Bacteroidetes* (UAFB) (Ma et al., 2015), *Bacillales* and *Rhodobacterales* (UASB reactor) (Liu et al., 2013), *Acetothermia*, *Proteobacteria* and *Firmicutes* (UASB) (Chen et al., 2018), *Proteobacteria*, *Chlorobi*, *Bacteroidetes*, and *Firmicutes* (Anaerobic biofilm reactor , AnBR) (Dong et al., 2016), *Proteobacteria* (ABR) (Lin et al. 2012), *Anaerolineaceae* (Anaerobic SBR) (Rosenkranz et al., 2013) and *Proteobacteria* and *Firmicutes* (EGSB) (Lim et al., 2014). These studies suggested microbiotas are highly related to process and specific organic pollutants types and environmental conditions.

Nowadays, some reports explored the function of microbes in hydrolysis

acidification (Tab. 4), *Firmicutes* and *Actinobacteria* (Ma et al., 2019), *Chloroflexi* (Li et al., 2017a) facilitated hydrolysis acidification, *Desulfobacter*, *Desulfofustis* and *Desulfomicrobium* were SRB in hydrolysis acidification utilizing specific organic pollutants as electron donor for reducing sulfate to H_2S, *Chlorella sp.* was benzene-degrading bacteria in hydrolysis acidification for benzene degradation (Huo et al., 2018b), degrading or mineralizing organic matters (*Proteobacteria*), facilitating hydrolysis and acidogenesis by secreting extracellular enzymes such as lipases, proteases etc. (*Firmicutes*), degrading macromolecular organics (*Chloroflexi*) (Mielczarek et al., 2012), degrading protein and carbohydrate for producing acetic and propionic acids (*Bacteroidetes* and *Actinobacteria*) (Ammar et al. 2013, Zeppilli et al., 2015), degrading aromatic compounds, PAHs, chlorinated hydrocarbons and other toxic substances *Syntrophorhabdus* (Ma et al., 2015), *Bacillales* and *Rhodobacterales* (Liu et al., 2013), *Acinetobacter* (Dong et al., 2016), *Pseudomonas* (Liao et al., 2015), *Bacillus* (Anwar et al., 2009), *Klebsiella* (Cui et al., 2014), *Xanthobacter* (Ding et al,. 2016), *Novosphingobium* (Segura et al., 2017), removing sulfite and thiosulfate *Desulfomicrobium* (Thevenieau et al., 2007), *Sulfurovum* (Huang et al., 2015), *Sulfurovum riftiae* (Giovannelli et al., 2016), degrading nitrogen-containing substances *Nitrosomonas* (Ding et al., 2016), *Nitrospira* (Yu et al., 2018), *Nitrosomonadaceae* (Shi et al., 2018) uncultured. From abovementioned researches, we can conclude the process, specific organic pollutants and external conditions displayed significant microbial communities and specific organic pollutants removal depends on the combined function of different microbes (Janbandhu and Fulekar, 2011; Gu et al.,2018; Palma et al., 2018).

At present, it's more meaningful to figure out specific organic pollutant's degradation pathways for future research. nonylphenol (NP) degradation pathway displayed in Fig .3 (Duan et al., 2018), phenolic hydroxyl removed after alkyl chain firstly was oxidized and then intermediates mineralized into CO_2 with *Proteiniphilum acetatigenes* and *Propionibacterium acidipropionici*. Tetrachlorobisphenol-A (TCBPA) biodegradation pathway (Fig. 4) Yuan et al., 2011 discovered TCBPA dechlorinated by SRB significantly. Yen et al., 2016 revealed PTA can be transformed into intermediates (i.e. CH_3COOH, C_6H_5COOH, $C_8H_7O_2$, and $HOOCC_6H_4CHO$) firstly and then transformed into methane under anaerobic condition. Chen et al., 2018 studies indicated phenolic hydroxyl firstly removed into benzoate following benzene ring removal for generating hydrogen and acetate, and intermediates were

transformed into methane via methanogenesis finally (Fig. 5). However, it should be noted that more efforts are taken into exploring specific organic pollutants degradation pathway which considering the petrochemical wastewater's composition and structure complexity.

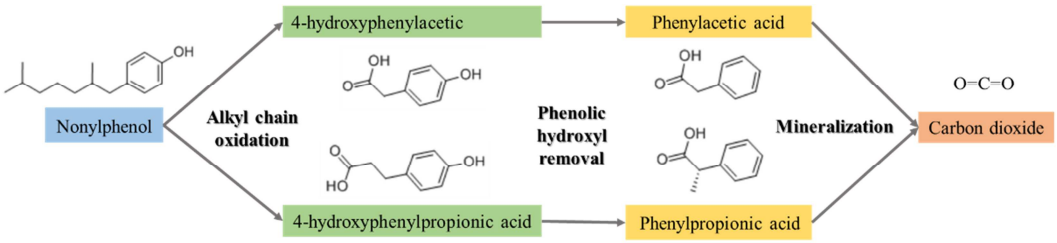

Figure 3 The proposed degradation pathway of NP by anaerobic fermentation.

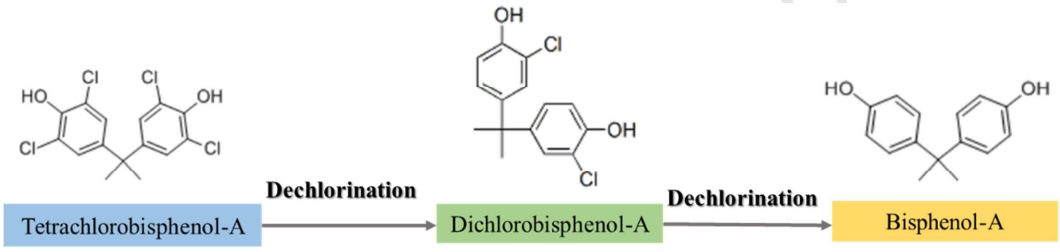

Figure 4 The proposed degradation pathway of TCBPA by anaerobic fermentation.

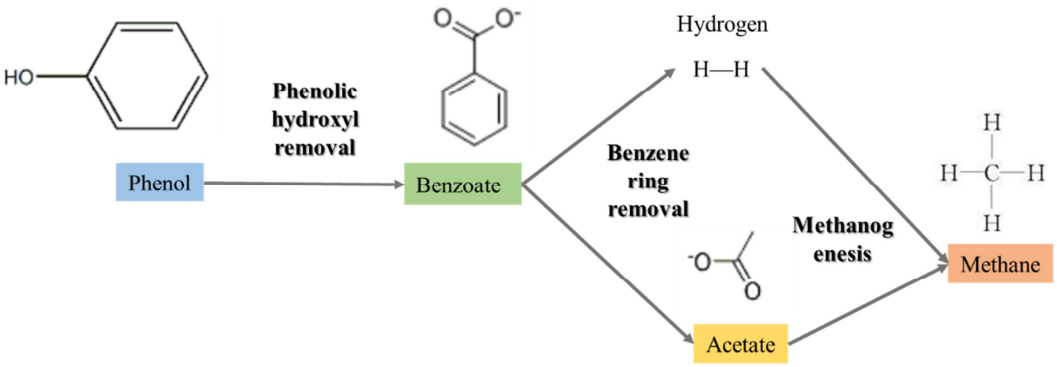

Figure 5 The proposed degradation pathway of phenol by anaerobic fermentation.

1 Table 3 Petrochemical wastewater's pretreatment by different biological methods.

Methods	Key Parameters	specific organic pollutants (mg/L)	$BOD_5/COD_{(Cr)}$	Inf. Con. (COD mg/L)	Eff. Con. (COD mg/L)	Other pollus. (Removal rate)	References
EGSB	HRT 62.8 h	PO/MTBE	-	4.65×10^3 ± 236	669 ± 167	-	(Liang et al. 2019)
ABR	HRT 60.0-144 h, loading rate 0.070-212 kg COD/m^3/d	Oil	-	700-2.12×10^5	7.42×10^4	Oil (88.0%)	(Ji et al., 2009)
UAFB	VLR 2.10 kg COD/m^3/d, temperature 37.0 ˚C and HRT 18.9 h	Terephthalate, benzoic acid	-	1.25×10^3-2.25×10^3	74.5-134	-	(Ma et al., 2015)
UASB	HRT 0.330-0.750 d, SRT 32.0-58.0 d, temperature 27-35 ˚C	Phenolics	-	2.24×10^3	200-385	-	(Ramakrishnan and Surampalli,2012)
AHR	HRT 0.330-0.750 d, SRT 42.0-68.0 d, temperature 27-35 ˚C	Phenolics	-	2.24×10^3	202-358	-	
UASB	HRT 12.0 h	Alkanes	-	130–1.25×10^33	46.7-446	NH_3-N (94.0%) SS (98.0%)	(Liu et al., 2013)
UASB	HRT 10.0-20.00 d, temperature 36.0 ± 2.00 ˚C, OLR 0-11.0 kg COD/$m^{3/}$d	-	-	500 -5.00×10^3	35.0-350	-	(Chen et al., 2017)
ABR	HRT 1.00-10.4 d, OLR 0-11.0 kg COD/m^3 d, NB 30.0-700 mg/L	NB	-	3.00×10^3	240-630	NB (100.0%)	(Kuscu and Sponza, 2009b)
AMBR	HRT 6.00-9.00 d, OLR 3.33-66.8 g NB/m^3, NB 20.0-40.0 mg/L	NB	-	3.00×10^3	270-450	NB (100.0%)	(Kuscu and Sponza, 2009a)
HAR	HRT 32.0 h, VLR 1.60-4.50 kg COD/(m^3·d), temperature 33.0, 37.0, 43.0, 52.0 ˚C	PTA	-	1.50×10^3-4.00×10^3	122.-1.36×10^3	Terephthalate (60.8-94.0%)	(Li et al., 2014b)
ABR	Acetone 0.700–0.800 g/L, ethanol 0.560–0.630 g/L, butanol 4.90–5.60 g/L, OLR 5.40 kg COD m^3/d, COD: N: P ratio of 200–300:5:1	ABE	-	45.0-63.0 kg/L	5.31-7.50 kg/L	-	(Zhang et al., 2011)
CFPBR	HRT 1.40 d, OLR 3.40-20.0 g COD/L/d, temperature 35 ˚C	FT	-	2.80×10^4	1.12×10^3	-	(Majone et al., 2010)
UAFB	HRT 1.70-6.00 d, COD: N: P 200:5:1, temperature 34 ± 1 ˚C, pH 6.00	FT	-	3.29×10^4–3.85×10^4	6.57×10^3-7.69×10^3	-	(Wang et al., 2017b)
EGSB	HRT 2.00 d, OLR 0.870-1.87 g COD/L /d, temperature 30 ˚C	Phenol and cresols	-	1.83×10^3-3.39×10^3	183-339	-	(Almendariz et al., 2005)
EGSB	HRT 8.00 d, loading rate 0.290 kg COD kg-VS/ d, temperature 20.0 ˚C	Ethanol and 2–propanol	-	2.00×10^3-2.66×10^5	-	-	(Chang et al., 2005)
UASB	HRT 20.0 h, flow rate 3.00 L/d, temperature 20 ˚C	2,4 dichlorophenol	-	3.00×10^3	690	-	(Sponza and Uluköy, 2006)
UASB	HRT 48.0-72.0 h, phenol 210-840 mg/L, SO_4^{2-} 1.00-2.00E3 mg/L, salinity 1.0-3.0%, temperature 35.0 ± 1.00 ˚C	Phenol	-	1.00×10^3-3.00×10^3	300 -900	Sulfate (85.0%)	(Guo et al., 2015)
UASB	Turf soil 1.12 mg/g, OLR 1.00-2.50 kg COD/m^3/d, pH 6.50-8.00, temperature 36 ± 2 ˚C	Phenol	-	1.00×10^3-3.00×10^3	30.0-90.0	Phenol (97.0%)	(Chen et al., 2018)
EGSB	HRT 8.00 d, SLR = 0.25–0.29 kg COD/ kg VS/d, temperature 25.0 ˚C	2-propanol	-	-	-	-	(Vermorel et al., 2017)

HA	DO 0.200-0.300 mg/L	-	0.230-0.430	421	316	-	(Wu et al., 2015)
HA-A/O	R 100.0%, HRT 32.0 h, SRT 17.0 d, DO 2.00–3.00 mg/L	1,3-dioxolane, 2-pentanone, ethylbenzene, 2-chloromethyl-1,3-dioxolane and indene	0.300-0.430	307–581	54.4 bench-scale 60.9 wastewater treatment plants	-	(Wu et al., 2016)
MHA-A/O	HRT 20.0 h	Alkanes, aromatic and polycyclic hydrocarbons	0.270 ± 0.180 - 0.340 ± 0.140	348-529	98.0-111	Ammonium (>94.0%)	(Yang et al., 2015)
HA-algal microcosms	pH 6.47–7.45, ORP -110±25.0 mV and temperature 35.0 ˚C	Aromatics	0.220-0.560	856 ± 11.0	146 ±1.00	NH_3–N (100.0%), Phosphorus (89.0%)	(Huo et al., 2018b)
A/O-BR	HRT 36.0-50.0 h, temperature 30.0-35.0 ˚C, DO aerobic 3.50-4.50 and anaerobic <0.130 mg/L	Oil	-	650–1.15×10^33	44.2-78.2	TN (82.8%)	(Li et al., 2017a)

Table 4 different microorganisms' functions in the removal specific organic pollutants.

Microorganisms	Key functions	Microorganisms	Key functions
Desulfomicrobium	SRB, transforming sulfate, sulfite and thiosulfate into hydrogen sulfide (Thevenieau et al., 2007)	*Azospirillum*	Fixing nitrogen (Han and New, 1998)
Thialkalivibrio thiocyanodenitrificans	Sulfur oxidizing autotrophic bacteria, electron donor with nitrate/nitrite as electron acceptor to produce sulfate and ammonia (Sahariah and Chakraborty, 2011)	*Xanthomonadales*	Heterotrophic denitrifiers, denitrification (Chon et al., 2010)
Sulfurovum	Oxidizing sulfur to sulfate (Huang et al., 2015)	*Nitrosomonadaceae uncultured*	Ammonium-oxidizing bacteria, oxidizing ammonium (Ding et al., 2016)
Sulfurovum riftiae	Sulfur-and thiosulfate-oxidizing bacteria (Giovannelli et al., 2016)	*Nitrosomonas*	Ammonium-oxidizing bacteria, oxidizing ammonium (Yu et al., 2018)
Sulfuritalea	Obligate and facultative sulfur chemolithoautotrophs, oxidize inorganic sulfur compounds (Watanabe et al., 2014)	*Nitrospira*	Nitrite-oxidizing bacteria, oxidizing nitrite (Shi et al., 2018)
Desulfomicrobium	SRB, reducing sulfate to hydrogen sulfide (Guo et al., 2015)	*Anaerolineae*	Semi-syntrophic and fatty acids-oxidizing bacteria, degrading carbohydrate (Narihiro et al., 2012)
Pseudomonas	Denitrifiers; degrading oil, alkanes, and aromatic substances (Liao et al., 2015); producing extracellular proteases and depolymerases (Liao et al., 2013)	*Novosphingobium*	Degrading aromatic compounds (Segura et al., 2017)
Rhodocyclales	degrading hydrocarbon (Shokrollahzadeh et al., 2008)	*Rhodopseudomonas*	Photosynthetic bacteria, secreting lipase and anti-salinity (Ji et al., 2009)
Bacillus	Denitrifiers; degrading toxic substances (PAHs, naphthalene) (Anwar et al., 2009)	*Syntrophorhabdus*	δ-Proteobacteria, degrading PTA (Ma et al., 2015)
Lactococcus	Lactic acid bacteria, producing lactic acid (Yang et al., 2015)	*Bacillales*	Hydrocarbon-degrading bacteria, degrading *n*-alkanes (Liu et al., 2013)
Klebsiella	Facultative anaerobic bacteria, degrading toxic substances (Cui et al., 2012, Cui et al., 2014)	*Ottowia*	Degrading phenol (Cao et al., 2014)
Saprospiraceae	Enzymolysis protein (Xia et al., 2008)	*Xanthobacter*	Degrading chlorinated hydrocarbon; degrading halogenated short-chain hydrocarbons and carboxylic acids (Ding et al., 2016)
Defluviicoccus	Glycogen accumulating organisms, using acetate and propionate as carbon source (Dai et al., 2007)		
Rhodobacterales	Hydrocarbon-degrading bacteria *n*-alkanes (Liu et al., 2013)	*Proteiniborus*,	Protein-utilizing bacteria, producing CH_3COOH and hydrogen (Niu et al., 2008)
Bacteroidetes vadin HA17	Anaerobic/facultative metabolism bacteria, degrading complex carbon organics (Baldwin et al., 2015)	*Clostridiales*	Degrading proteins, lipids and carbohydrates (Ma et al., 2019)
Caldisericum	Hydrolytic-acidogenic bacteria, hydrolyzing and degrading biorefractory pollutants (Cheng et al., 2014; Hao and Wang, 2015)	*Cetobacterium*	Digesting proteins (Hao et al., 2017)
Aminicenantes	Anaerobic bacteria, degrading hydrocarbons (Farag et al., 2014)	*Ilumatobacter*	Decomposing organic substances (Fang et al., 2015)
Longilinea sp.	Degrading alkanes (Chen et al., 2017)	*Prevotella*	Degrading polysaccharides (Nograsek et al., 2015)
Spirochaetaceae	Utilizing small organic molecules as carbon and energy sources (Chen et al., 2017)	*Thermotogaceae*	Anaerobic thermophiles, fermenting carbohydrates and peptides (Wagner and Wiegel, 2008)
Mesotoga	Utilizing small organic molecules as carbon and energy sources (Ben et al., 2013)	*P. putida F1*	Degrading aromatic hydrocarbons (da Silva and Alvarez, 2010)

3.2 Advanced treatment

3.2.1 Catalytic oxidation

Besides fenton-like oxidation, ozone oxidation, the current focus more concentrated on catalytic oxidation, photocatalysis oxidation and electrochemical oxidation. Catalytic oxidation (CO) degrading specific organic pollutants by generating free radicals which are located on the surface of various catalysts with air/O_2/O_3, such as Mn (Hou et al. 2018), Fe (Han et al. 2016) (Eqa. 12-16 (Xiao et al., 2018)), Co oxides (Sable et al., 2018). Owing to its superior specific organic pollutant's degradation capability, it has been widely introduced into advanced treating specific organic pollutants (Tab. 5).

$$O_3 + C_1 \rightarrow O_2 + O^{\bullet} \quad (13)$$

$$O_3 + O^{\bullet} \rightarrow O_2 + O_2^{\bullet} \quad (14)$$

$$O_2^{\bullet} \rightarrow O_2 + C_1 \quad (15)$$

$$SOPs + C_2 \rightarrow C_2^{SOPs} \quad (16)$$

$$C_2^{SOPs} + O^{\bullet} \rightarrow \text{Intermediate products} + CO_2 \quad (17)$$

Where C_1, C_2, $O^{\bullet}$ and $O_2^{\bullet}$ presents the active sites of ozone decomposition, specific organic pollutants and active oxygen, respectively.

3.2.1.1 Key parameters in catalytic oxidation

More studies have paid attention to essential factors are catalyst types, catalyst-loading, and supporting materials. Mn-loading MnO_x/Al_2O_3 and Fe-loading $MnO_x/Ce_{0.65}Zr_{0.35}O_2$ monolithic catalysts (Han et al., 2016; Hou et al., 2018) degraded specific organic pollutants because Mn or Fe had the stronger electron transformation ability to O_3 for enhancing the degradation. Noble metals catalysts Ru/TiO_2 had highest catalytic performance compared with Pd/TiO_2, Pt/TiO_2 and Rh/TiO_2 for chlorobenzene degraded (Liu et al., 2019a), Pt/BEA only 85.0% toluene removal compared than Pt-Ce/BEA and Ce/BEA (> 99.0%) under similar conditions (Xiao et al., 2018). The degradation increased as catalyst-loading increases firstly, further the performance decreased because over-loading catalyst reduced the exposure opportunity of catalyst and specific organic pollutants, for example, the optimal specific organic pollutants removal capacity exhibited at 10.0% Mn-loading, (Hou et al., 2018) and 16.0 wt% Fe-loading (Han et al., 2016). Different supporting materials also have different effects on specific organic pollutants removal, the supporting materials followed CeO_2 > TiO_2 > Al_2O_3 > YSZ with LaMnOx catalyst and sulfonated-ZrO_2>ZrO_2 iron oxides by CO removed 1,2-dichloropropane (1,2-DCP) (Han et al. 2016) and phenol (Sable et al., 2018). Moreover, other parameters affecting the CO include temperature and air/O_2/O_3 dosage are also considered seriously (Chen et al., 2015; Teimouri et al., 2018; Zhang et al., 2018c). However, reducing catalyst cost, modifying its structure for improving its useful life is future research hotspot.

3.2.1.2 Removal mechanism of typical specific organic pollutants in catalytic oxidation

Recently, CO has been advanced treated actual secondary petrochemical wastewater by some researchers and certain specific organic pollutants removal mechanisms been explored. The maximal COD and total organic carbon removal

obtained 75.3% and 50.3% under these conditions: 0.300 g/h O_3, 0.450 g/L catalyst and 120 min reaction time by PAC@Fe_3O_4 CO (Ahmadi et al., 2017b), the specific organic pollutants removal mechanism was mainly decomposed by hydroxyl radical (Fig .6). Fe-Ni foam catalyst petrochemical wastewater CO indicated sCOD removal was 96.0% under 110 g/L catalyst, 12.2 mg/L O_3, pH 4.00-12.0, room temperature and reaction time 120 min, the specific organic pollutants removed by •OH generated from zone reacted with oxides and hydroxides (Huang et al., 2019), moreover, TP, TN, NO_3-N, Cl^-, and some heavy metals also be removed at a certain degree. A critical role of O_3 in the CO investigated by Zhang et al., 2018b, EPS firstly dropped was ascribed to it degraded into dissolved organic matter (DOM) by O_3, while it increased because more EPS secreted by sludge to protection from adverse conditions and DOM dominated gradually reacting with O_3. The CO dibromomethane and styrene removal mechanism (Fig. 7), which revealed it greatly depended on active groups produced from CO without oxidants and catalysts (Huang et al., 2017; Mei et al., 2018).

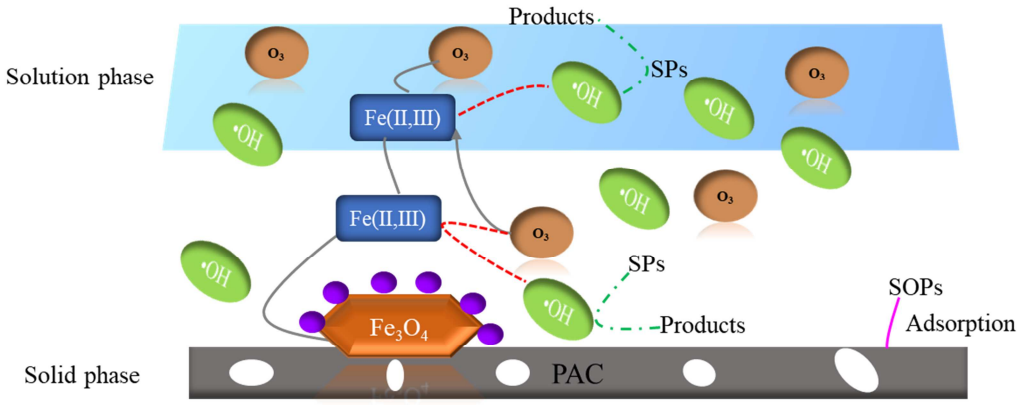

Figure 6 The proposed degradation pathway of specific organic pollutants by PAC@Fe_3O_4 CO (Ahmadi et al., 2017b).

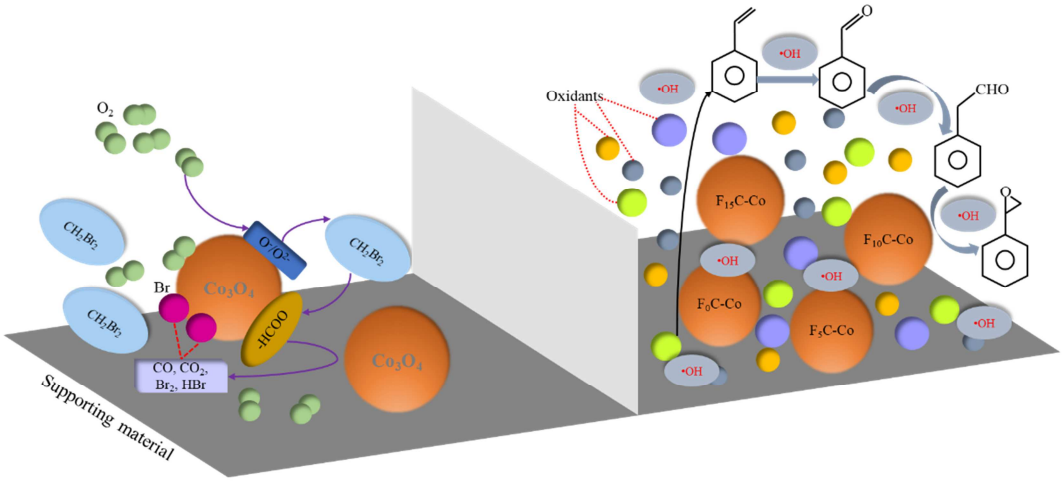

Figure 7 The proposed degradation pathway of specific organic pollutants by Co_3O_4 and Four cobalt (III) corroles CO (Huang et al., 2017; Mei et al., 2018).

1 Table 5 Summary of studies addressing for the removal of specific organic pollutants by CO.

Compounds	Reaction/hold time (min)	Initial Conc. (ppm)	Catalyst types	Synthesized methods	Assisted substances	Catalyst doses (g)	Oxygen/ozone doses	pH	Temperature (◦C)	Pressure (Bar)	% Compound	% COD	% TOC	References
1,2-DCP	60.0	1.00×10^3	$LaMnO_3/TiO_2$ $LaMnO_3/YSZ$ $LaMnO_3/Al_2O_3$ $LaMnO_3/CeO_2$	Situ citrate sol-gel	-	-	20.0% O_2	-	507 460	-	100.0	-	-	(Zhang et al., 2017)
Alkene	600	1.00 mM	F_0C-Co F_5C-Co F_{10}C-Co F_{15}C-Co	Precipitation	-	0.100×10^-3 mM	0.100 mM PhI $(OAc)_2$, TBHP, $KHSO_5$, PhIO	-	25.0	-	96.0	-	-	(Huang et al., 2017)
Toluene	-	1.00×10^3	$MnO_x/Ce_{0.65}Zr_{0.35}O_2$	Impregnation	-	15.0%	Air	-	250	-	100.0	-	-	(Hou et al., 2018)
Formaldehyde	-	200	Fe(□)/γ-Al_2O_3	Impregnation	-	16.0 wt% Fe_2O_3	500.00 mL/min air	6.00	100	-	67.0	-	-	(Han et al., 2016)
Nitrobenzene	3.00×10^3	300 mg/L	sFCCc	Calcination	-	0.250, 0.500, 0.750 and 1.00 g	0.62- 2.25 mg/min	3.00-4.00	25.0-50.0	-	-	55.6-87.2	-	(Chen et al., 2015)
Formaldehyde	200	15.0	$MnO_x/γ$□Al_2O_3	Wetness impregnation	-	10.0%	250 mL/min 150 ppm O_3, 20.0% O_2	-	200	-	92.0	-	-	(Zhu et al., 2017)
Chlorobenzene	-	50.0	Pd/TiO_2 Pt/TiO_2 Ru/TiO_2 Rh/TiO_2	Co-precipitation and calcination	-	-	20.0% O_2	-	287 337 339 340	-	90.0	-	-	(Liu et al., 2019a)
Phenol	360	0.100 g/L	Fe/ZrO_2	Calcination and impregnation	0.50 g/L H_2O_2	2.00 g/L	-	-	25.0	Atmospheric	64.0	-	-	(Sable et al., 2018)
			Fe/sulfonated-ZrO_2			0.500 g/L	1.20 g/h O_3				88.0			
specific organic pollutants	120	COD 144 mg/L	Fe-Ni foam	Precipitation	-	110 g/L	12.2 mg/L O_3	8.30	20.0–25.0	-	-	96.0	-	(Huang et al., 2019)
Dibromoethane	-	500	Co_3O_4	Evaporation and calcination	2.00% H_2O_2	80.0 mg	10% O_2	-	271	-	90.0	-	-	(Mei et al., 2018)

Benzene	-	1.50×10^3	$Co_2Mn_1O_x$	Oxalate co-precipitation	-	100 mg	20.0% O_2	-	191	Ambient	90.00	-	-	(Zhang et al., 2018c)
Toluene	2.40×10^3	22.0	Pt-Ce/BEA Pt/BEA Ce/BEA	Calcination	-	0.100 g	20.0 mL/ min 300 ppm O_3 and O_2	-	90.0	-	99.0 - -	-	-	(Xiao et al., 2018)
Dibenzothiophene	75.0	500	MoO_3 V_2O_5 MCM-41	Impregnation	57.0 µl 30.0% H_2O_2	0.100 g	-	-	70.0	-	99.1	-	-	(Teimouri et al., 2018)
specific organic pollutants	120	362± 36.0 mg/L	PAC@Fe_3O_4	Co-precipitation	-	0.150–0.750 g/L	0.050–0.300 g/h O_3	3.00–11.0	-	-	-	75.3	50.3	(Ahmadi et al., 2017b)

3.2.2 Photocatalysis oxidation

Photocatalysis oxidation (PCO) is a process depends on free radicals generating from photocatalytic reaction to advanced treat specific organic pollutants under catalyst and light existed simultaneously (Fig. 8**Error! Reference source not found.**), which has been extensively used in advanced treatment (Tab. 6) based on its high performance and low reaction time.

3.2.2.1 Key parameters in photocatalysis oxidation

The key parameters affecting performance are catalyst types, catalyst dosage, specific organic pollutants species, specific organic pollutants initial concentration and external conditions. The catalysts are usually metal oxides semiconductor (titanium dioxide, iron oxide, cerium oxide etc. (Ameta et al., 2018)). TiO_2 nanoparticles (Khaksar et al., 2017), $Bi_4O_5Br_xI_{2-x}$ (Meng et al., 2018), $PdO/Al_2O_3–Nd_2O_3$ (Barrera et al., 2014), PF/TiO_2 (Li et al., 2018a), Sn_3O_4 microballs (Balgude et al., 2019) and Fe_2O_3 (Vosoughi et al., 2017) both PCO treated phenol with 62.8-100.0% removal. La-doped Zn (O, S) nanoparticles (Abdullah et al., 2019), $B–GO–TiO_2$, $B–TiO_2$, $GO–TiO_2$ (Shokri et al., 2016), Fe_2O_3/RGO nanocomposite (Mohan et al., 2019), MoS_2/rGO, CdS-MoS_2/rGO composite (Peng et al., 2016) degraded 4-nitrophenol (4-NP) achieved 22.0-100.0% removal. These results indicated different catalysts exhibited a critical role in same specific organic pollutants and different specific organic pollutants also affected the removal. TiO_2 is considered to be an outstanding photocatalyst in PCO for specific organic pollutants based on its stable performance, high photocatalytic activity, low-cost and environmental-benign material. Recently, Co-doped and modified technologies also applied to enhance its performance further and achieved good results (Bai et al., 2019; Wang et al., 2017c).

The catalyst dosage also affects the degradation capability significantly, which the photocatalytic activity promoted as the increase of catalyst dosage because more active sites generated to produce more free radicals. Nevertheless, overdose is harmful to improve activity for attenuating the light intensity to catalyst (Morris et al., 2004). For instance, (Shokri et al., 2016) indicated modified TiO_2 dosage ($GO–B–TiO_2$) PCO degradation 25.0 mg/L 4-NP was 1g/L, (Khaksar et al., 2017) confirmed optimal TiO_2-loading was 80.0 g/m^2.

The initial specific organic pollutants concentration is one of significant parameters affecting the degradation performance resulting from limited PCO process,

Khaksar et al., 2017 found when initial phenol concentration increased from 50.0 mg/L to 700 mg/L, the phenol degradation efficiency decreased by 30.0%, similar results detected by Bai et al., 2019 for bisphenol A (BPA) removal by D35-TiO_2 nano-crystalline film. The reason is mainly resulted from overdose specific organic pollutants influencing the free radical's production as it covered on the surface of catalyst.

The external conditions (i.e., reaction time, pH, assisted materials etc.) are also exhibit different impact on PCO process at a certain degree. The reaction time is absolutely required to degrade specific organic pollutants with the action of free radicals, some of researchers found time is highly important factor than other factors, Khaksar et al., 2017 concluded the significance of different factors followed: time > initial phenol concentration > TiO_2 concentration > pH, Quispe-Arpasi et al., 2018 and Silva et al., 2015 both suggested necessary reaction time should be guaranteed for effectively or completely degrade specific organic pollutants was due to pH affected the free radicals generation , therefore, suitable pH should be selected seriously. Khaksar et al., 2017 determined the optimal pH was 9.00 for TiO_2 photocatalytic oxidized phenol was attributed to •OH are the dominant oxidant species at neutral or higher pH levels, however, Bai et al., 2019 discovered at low or high pH both had negative impact on the free radicals formation and the interaction between specific organic pollutants and catalysts, on the contrary, Hayati et al., 2018 reported acidic pH was necessary to PCO with the existence of HCO^{3-} and CO_3^{2-}, which interfered •OH formation. Assisted substances mainly are oxidants added to PCO for enhancing the free radical's formation, for instance, H_2O_2 was added by Bustillo-Lecompte et al., 2018 improved the degradation capability of BTEX efficiently and similar phenomena also confirmed by Quispe-Arpasi et al., 2018, which resulted from more •OH produced in PCO with addition of H_2O_2. PCO has been gradually implemented in industrial wastewater AT, especially in the petrochemical wastewater for the removal of specific organic pollutants in recent years. Silva et al., 2015 compared the removal of catalyst TiO_2-$ZNSiO_2$ and P25 under UV and visible radiation under determined conditions, 48.6%, 45.2% and 66.3%, 50.2% specific organic pollutants removal achieved, respectively. Spent catalyst treated (Vosoughi et al., 2017) phenol determined the COD removal reached 98.5% under optimum parameters: reaction temperature 47.1 $^{\circ}$C, 0.160 mol/L H_2O_2, 20.0 g/L catalyst and 92 min. Color and COD removal reached 93.1% and 23.4% with H_2O_2 assistance by AD coupled with PCO

(Quispe-Arpasi et al., 2018) treated petrochemical wastewater, respectively.

3.2.2.2 Removal mechanism of typical specific organic pollutants in photocatalysis oxidation

It is important to explore the specific organic pollutants removal mechanisms for overall understanding PCO reaction, some studies have depicted specific organic pollutants removal mechanism recently. Liu et al., 2019b (Fig. 9) revealed phenol removal mechanism completely, the electron-hole pairs produced from the surface of the catalyst when it exposed to light irradiation, then •OH generated by the reaction of photo-generated-holes and H_2O and oxygen anions ($\bullet O_2^-$) formed by the reaction of DO and electrons. The phenol transformation and degradation started under the action of •OH and $\bullet O_2^-$, phenol firstly oxidized into o-benzoquinone and p-benzoquinone, and then transformed into muconic acid and 2,5-dioxo-3-hexenedioic acid with free radicals, respectively. These carboxylic acids further oxidized into unknown intermediates and finally degraded into CO_2 and H_2O. Similar phenol degradation mechanisms also confirmed by Wang et al., 2017c degraded phenol by RF supported catalysts irradiated with solar-light and Balgude et al. 2019 removed phenol by Sn_3O_4 microballs photocatalysts with solar-light. Peng et al., 2016 and Abdullah et al., 2019 both clarified 4-NP PCO detoxification via MoS_2/rGO composite and La-doped Zn (O, S) photocatalyst that 4-NP could be changed into 4-AP with the effect of free radicals. Moreover, phenolic substances also degraded into CO_2 and H_2O, for example, Meng et al., 2018 confirmed $Bi_4O_5Br_{0.6}I_{1.4}$ photocatalyst decomposed fully resorcinol, o-phenylphenol, and 4-tert-butylphenol into CO_2 and H_2O. However, it should be noted that more and more specific organic pollutants removal performance and the mechanism through PCO should be explored.

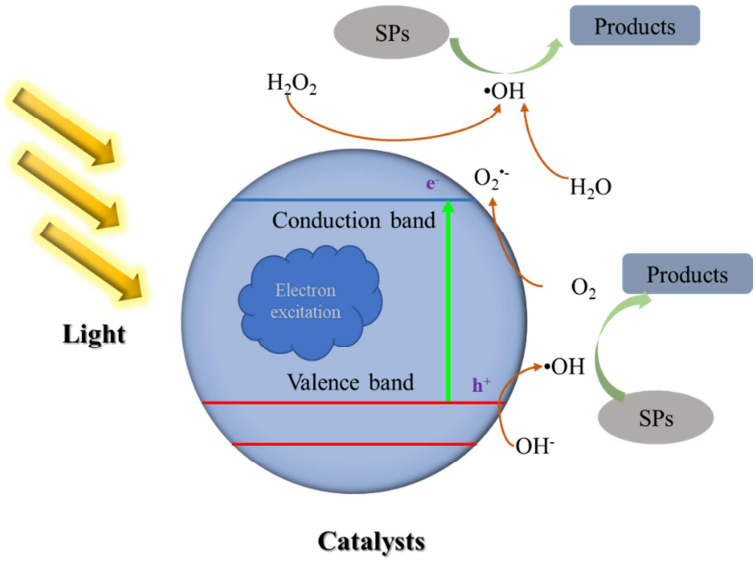

Figure 8 The general removal mechanism of specific organic pollutants by PCO.

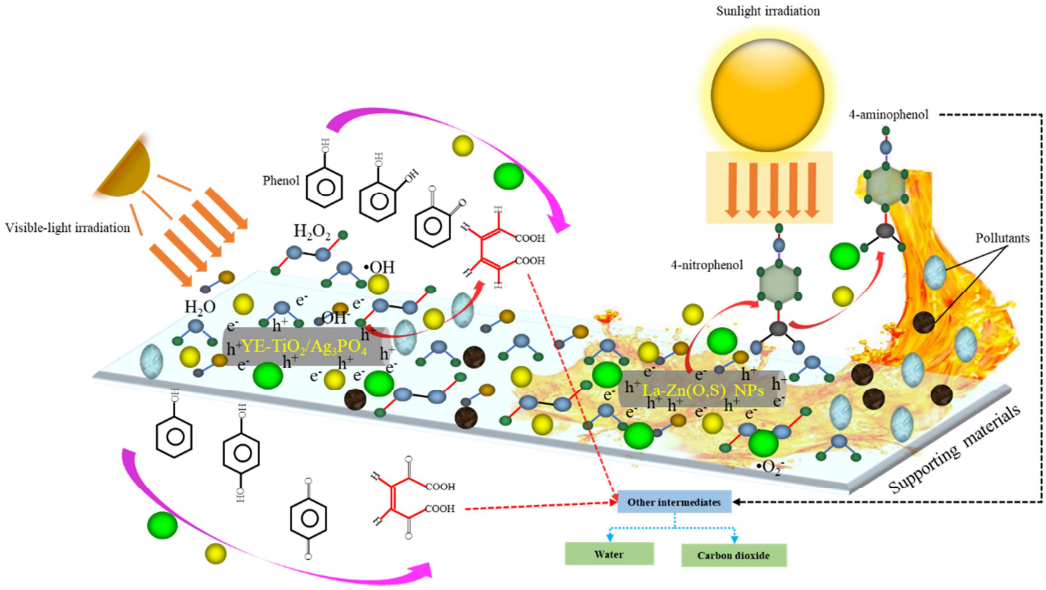

Figure 9 The proposed removal mechanism of phenol and 4-nitrophenol by PCO.

1 Table 6 Summary of studies addressing for the removal of specific organic pollutants by PCO.

Compounds	Reaction time (min)	Initial conc.	Catalysts	Catalyst dosage (g/L)	pH	Light intensity	Temperature (◦C)	% Compound	% COD	% TOC	References
Phenol	180	50.0 mg/L	TiO_2 nanoparticles	80.0 g/m^2	9.00	160 W UV radiation	Room	88.0	-	-	(Khaksar et al., 2017)
Nitrobenzene	100	20.0 mg/L	TiO_2 cake (anatase/rutile)	1.60 mg	-	500 W UV light irradiation	Room	97.0	-	-	(Li et al., 2017b)
Phenolic	80.0	-	$Bi_4O_5Br_xI_{2-x}$	1.00 mg/mL	-	500 W halogen lamp visible light	Room	92.0	-	79.0	(Meng et al., 2018)
Phenol	120	80.0 ppm	$PdO/Al_2O_3–Nd_2O_3$	1.00 mg/mL		4.40×10^-3 μW·cm^{-2} UV power supply lamp	Room	-	-	-	(Barrera et al., 2014)
Phenol	450	10.0 mg/L	PF/TiO_2	2.00 mg/mL	-	0.700 kw/m^2 sunlight	200	62.8	-	-	(Li et al., 2018a)
Phenol	360	20.0 mg/L	TiO_2@graphene nanocomposites	1.00 mg/mL	7.00	8.00 W UVC lamp	Room	-	-	-	(Shahbazi et al., 2018)
Phenol	20.0	20.0 mg/L	(Yb^{3+}, Er^{3+}) co-doped TiO_2/Ag_3PO_4	0.200 mg/mL	-	Visible-light illumination	Room	100.0	-	-	(Liu et al., 2019b)
Phenol	60.0	6.00 ppm	Sn_3O_4 microballs	1.00 mg/mL	-	Sunlight	Room	-	-	-	(Balgude et al., 2019)
Phenol	15.0	-	RGO/α-FeOOH composites	-	4.00-8.00	Solar-light-driven	Room	100.0	69.0	31.0	(Wang et al., 2017c)
Bisphenol A	300.0	10.0 mg/L	D35-TiO_2 nano-crystalline film	5.00 mg/L	7.00	116 mW/cm^2 solar power	Room	100.0	-	-	(Bai et al., 2019)
Bisphenol A	360	-	FDU-PdPcS	0.020× 10^-3 mg/mL	11.0	500 W halogen lamp	Room	100.0	-	-	(Xing et al., 2013)
4-nitrophenol	120	30.0 ppm	La-doped Zn (O, S) nanoparticles	0.500 mg/mL	-	0.088 mW·cm^{-2} UV tube lamp	Room	100.0	-	-	(Abdullah et al., 2019)
4-nitrophenol	180	25.0 mg/L	B–GO–TiO_2 B–TiO_2 GO–TiO_2	1.00 g/L	3.00	100 W tungsten lamp visible light	Room	100.0 85.0 80.0	85.0 70.0 65.0	-	(Shokri et al., 2016)
4-nitrophenol	90.0	25.0 mg/L	Bi_2O_3 TiO_2 ZnO ZrO_2	2.00 mg/mL	-	150 W halogen lamp	Room	100.0 35.0 34.0 22.0	-	-	(Muersha and Soylu, 2018)
4-nitrophenol	50.0	10.0 mg/L	Fe_2O_3/RGO nanocomposite	0.500 mg/mL	-	Visible light irradiation	Room	100.0	-	-	(Mohan et al., 2019)
4-nitrophenol	60.0	20.0 mg/L	MoS_2/rGO CdS-MoS_2/rGO composite	0.400 mg/mL	-	500 W xenon lamp visible light	Room	- 70.0	-	-	(Peng et al., 2016)
P-nitrophenol	120	1.00 mM	P-TiO_2 thin films	-		1.00×10^3 W xenon lamp	28.0 ± 1.00	-	-	-	(Méndez et al.,

			TiO_2 thin films			30.0 mW/cm^2 UV light					2015)
specific organic pollutants	60.0	20.0 mg/L	P25 (titania) TiO_2ZNSiO_2	0.700 g/L	-	125 W mercury vapor lamp UV and visible light	30.0	UV 48.6% and visible 45.2% UV 66.3% and visible 50.2%	-	-	(Silva et al., 2015)
Phenol	160	60.0 ppm	rGO/ZnO/TiO_2	0.600 g/L	4.00	150 W visible light illumination	27.0 ± 1.00	100.0	-	-	(Hayati et al., 2018)
Benzene Toluene Ethylbenzene Xylenes	360	100-300 mg/L	-	-	3.00	6.00 W UV lamp	20.0	90.0	-	-	(Bustillo-Lecompte et al., 2018)
specific organic pollutants	-	80.8 ± 0.600 mg/L COD	-	-	-	300 μmol m^{-2} s^{-1} Cool white fluorescent lamp	25.0	-	97.8	-	(Huo et al., 2018a)
specific organic pollutants	-	-	TiO_2	2.00 g/L	-	Artificial irradiation	37.0 ± 2.00	-	12.5 ± 1.60	-	(Quispe-Arpasi et al., 2018)
Phenolic	92.0	40.0–50.0 ppm	Fe_2O_3	20.0 g/L	7.00-8.00	-	47.1	98.5	-	-	(Vosoughi et al., 2017)
specific organic pollutants	240	1.05×10^3 mg/L COD	Co-TiO_2/zeolite	250 g/L	3.00	Low-pressure mercury lamp	-	-	93.4	-	(He et al., 2017)

3.2.3 Electrochemical oxidation

Electrochemical oxidation (ECO) is a promising and environment-friendly process for removal of specific organic pollutants. It works under mild operational conditions, requires less cover area, and has no need for a catalyst (Garcia-Segura et al., 2018). Organic pollutants are decomposed by allowing to react with oxidant species effectually stemming from active (i.e., iron, aluminum) or non-active anodes (i.e., graphite, platinum (Pt)) (Brillas and Martínez-Huitle, 2015; Martínez-Huitle and Panizza, 2018). Direct anodic oxidation occurs between the anode surface and the specific organic pollutants; direct electron transfer or indirect oxidation occurs in the solution, depending on electrochemically generated oxidants to avoid electron fouling (Martínez-Huitle and Panizza, 2018) (Fig. 10). Although active anodes have the potential to decompose specific organic pollutants into low-molecular weight species with electrocoagulation (Eqa. 17-27) (Nematollahi et al., 2017)), complete mineralization capability is still limited. Therefore, non-active anodes are predominantly employed in ECO (Cavalcanti et al., 2013; Scialdone et al., 2011). Reactive species originate from non-active anodes, once they have been initiated as represented in Eqa. 28-29; hydroxylated derivatives (M(•OH)) and chemisorbed active species (MO) are then devoted to mineralization of specific organic pollutants. Accordingly, specific organic pollutants are mineralized into CO_2 and H_2O (Eq. (30). However, side reactions occur as well because of free radicals (Eqa. 31-32) (Brillas and Martínez-Huitle, 2015; Martínez-Huitle and Panizza, 2018).

$$Fe \rightarrow Fe^{2+} + 2e^- \quad (18)$$

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \quad (19)$$

$$Fe^{2+} + 2OH^- \rightarrow Fe(OH)_2 \quad (110)$$

$$4Fe^{2+} + 10H_2O + O_2 \rightarrow Fe(OH)_3 + 8H^+ \quad (11)$$

$$8H^+ + 2e^- \rightarrow 8H_2O \quad (12)$$

$$4Fe + 10H_2O + O_2 \rightarrow Fe(OH)_3 + 4H_2 \quad (13)$$

$$Al \rightarrow Al^{3+} + 3e^- \quad (14)$$

$$3H_2O + 3e^- \rightarrow 3OH^- + \frac{3}{2}H_2 \quad (15)$$

$$Al + 3H_2O \rightarrow 3OH^- + \frac{3}{2}H_2 \quad (16)$$

$$R\text{-}H + (HO)OFe \rightarrow M\text{-}OFe + H_2O \quad (17)$$

$$R\text{-}H + (HO)OAl \rightarrow M\text{-}OAl + H_2O \quad (18)$$

$$M + H_2O \rightarrow M(^{\bullet}OH) + H^+ + e^- \quad (19)$$

$$M(^{\bullet}OH) \rightarrow MO + H^{+} + e^{-} \quad (20)$$

$$R + M(^{\bullet}OH) \rightarrow M + CO_2 + H_2O \quad (21)$$

$$M(^{\bullet}OH) + H_2O \rightarrow M + O_2 + 3H^{+} + 3e^{-} \quad (22)$$

$$2M(^{\bullet}OH) \rightarrow 2M + O_2 + 2H^{+} + 2e^{-} \quad (23)$$

Where, M, M(•OH), R, or RH express anode, adsorbed •OH, and specific organic pollutants, respectively.

3.2.3.1 Factors of dependence in electrochemical oxidation

Electrochemical oxidation has been applied for removal of specific organic pollutants in petrochemical wastewater over the recent years; these include phenol (Lizhang et al., 2016), nitrotoluene (Chen and Huang, 2014), benzene (Li et al., 2014a), etc. (Tab. 7). Some papers have discussed the pivotal factors in removal of specific organic pollutants, such as anode species, current density, pH, electrolyte, and initial concentration (Cavalcanti et al., 2013; Davarnejad et al., 2014; Garcia-Segura et al., 2018; Khatri et al., 2018; Li et al., 2014a). Active anodes (Al and Fe) have been considered in ECO as auxiliary reagents (Fe^{2+} or H_2O_2). Previous studies (Gümüş and Akbal, 2016; Khatri et al., 2018) employed iron anode with H_2O_2 solution to treat phenol. Here the well-known mechanism of pollutant removal was based on •OH via the Fenton reaction as follows (Eqa. 33-37):

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^{-} \quad (24)$$

$$Fe^{3+} + H_2O_2 \rightarrow HO_2^{\bullet} + H^{+} \quad (25)$$

$$R\text{-}H + {}^{\bullet}OH \rightarrow R^{\bullet} + H_2O \quad (26)$$

$$R^{\bullet} + Fe^{3+} \rightarrow R^{+} + Fe^{2+} \quad (27)$$

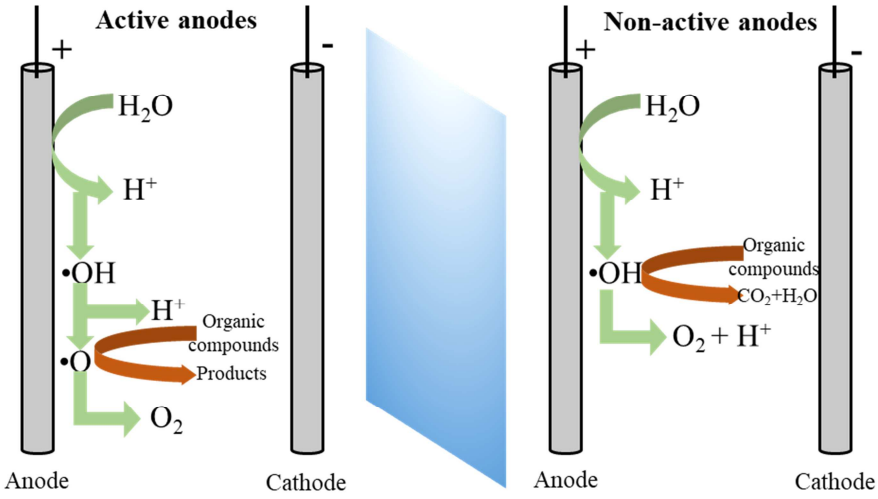

Figure 10 The proposed removal mechanism of refractory organic compounds by ECO.

$$R^{+} + {}^{\bullet}OH \rightarrow R\text{-}OH \quad (28)$$

however, substantial competitive reactions co-existed causing negative effect on

specific organic pollutants decomposition (Eqa. 38-41).

$$Fe^{2+} + {}^{\bullet}OH \rightarrow Fe^{3+} + OH^{-} \quad (29)$$

$$H_2O_2 + {}^{\bullet}OH \rightarrow HO_2^{\bullet} + H_2O \quad (30)$$

$$HO_2^{\bullet} + {}^{\bullet}OH \rightarrow O_2 + H_2O \quad (31)$$

$${}^{\bullet}OH + {}^{\bullet}OH \rightarrow H_2O \quad (32)$$

It should be noted that more and more studies are devoted to non-active anodes in ECO. For example, Pt, boron-doped diamond (BDD), and PbO_2/Ti. Current density is considered the most important factor that affects ECO, phenol removal (Khatri et al., 2018); TOC and COD removal is reported to have increased by 25.0% when current density changed from 0.100 to 2.00 mA/cm^2. A report by Cavalcanti et al. (2013) and Chen and Huang (2014) showed consistent results because •OH was enhanced greatly at higher current densities (Babuponnusami and Muthukumar, 2012b). Similarly, pH is an important prerequisite in ECO. Gümüş and Akbal (2016) discovered phenol degradation and COD removal to have been reduced by about 25.0%, when pH increased from 3.00 to 7.00. Similar results by Ren et al. (2018) and Zazou et al. (2019) indicated that pH must be kept at acidic conditions (2.80-3.00) to enable free radicals to have maximal catalytic activity. Furthermore, $Fe(OH)_3$ precipitation occurs and H_2O_2 decomposes at pH higher than 3.00 (Ma et al., 2009); consequently, pH lower than 3.00 is recommended in ECO. The electrolyte is generally added to improve degradation efficiency by intensifying the rate of electron transfer in low a conductivity effluent (Khatri et al., 2018) as compared to the effect of NaCl, Na_2SO_4, and KCl for phenol removal. It was observed that NaCl shows much better performance by producing chlorohydroxyl radicals (Eqa. 42-43). Sos Santos et al. (2014) exhibited acceleration for specific organic pollutants abatement because oxidizing agent peroxodisulfate generated. It is expected that degradation of specific organic pollutants has a negative relationship with increased initial concentration (Gümüş and Akbal, 2016). Up to only 30.0% COD removal could be achieved, when phenol concentration increased from 50.0 to 500 mg/L, which was likely due to limited •OH availability in the presence of excessive phenol (Babuponnusami and Muthukumar, 2012a).

$$M + Cl^{-} + H_2O \rightarrow M[{}^{\bullet}ClOH] + H^{+} + 2e^{-} \quad (33)$$

$$R + M[{}^{\bullet}ClOH] \rightarrow M + H^{+} + RO + Cl^{-} \quad (34)$$

3.2.3.2 Removal mechanism of typical specific organic pollutants in Electrochemical oxidation

Some attempts have been made to treat petrochemical wastewater in an advance manner and certain specific organic pollutants degradation pathways have also been explored. Davarnejad et al. (2014) compared the use of aluminum and iron anode for petrochemical wastewater removal to reveal that Fe anode exhibits better COD removal capability (67.3%) than the Al anode (53.9%) under optimal conditions. Similarly, dos Santos et al. (2014) investigated platinized titanium (Ti/Pt) and BDD anodes for removing actual petrochemical wastewater; the BDD system was observed to show better performance in reducing COD from 2.75×10^3 to 200 mg/L in less than five hours. The authors also evaluated the cost to be 56.2 kWh/m^3. An overall understanding of degradation pathways is beneficial for advanced petrochemical wastewater treatment of specific pollutants, such as dimethyl phthalate (DMP) and 1,2-dichlorobenzene (*o*-DCB). Ren et al. (2018) used graphite anode to treat dimethyl phthalate with CeO_2 catalyst in the presence of H_2O_2 solution; O_2 was firstly transformed into •OOH, which was then translated into H_2O_2 to produce more •OH in the CeO_2 system (Eqa. 44-45). The reason behind this was that negative charge covers the surface of CeO_2 in Na_2SO_4 solution, which transforms DO into unstable •OOH. This happens because the cleavage of the Ce-O bond, under the influence of the electric field follows the formation of •OH, which is responsible for degrading DMP

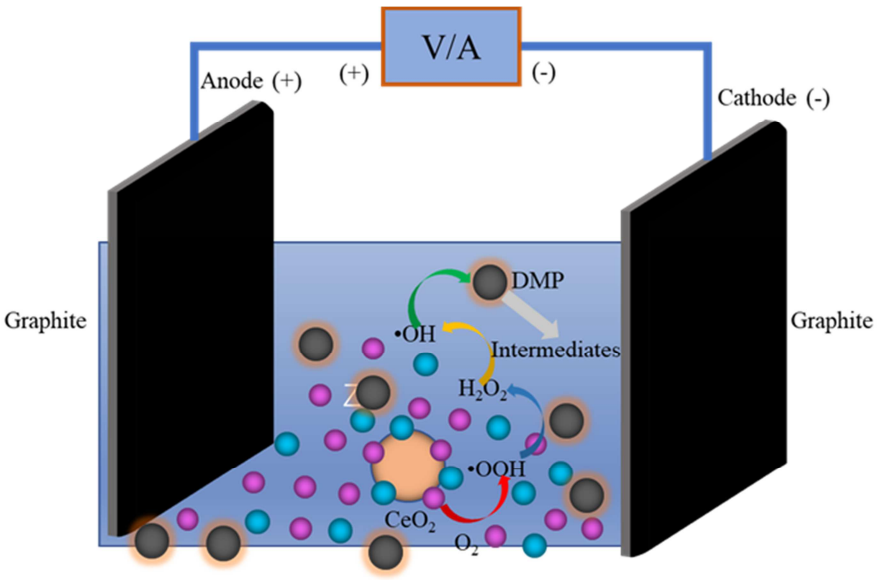

Figure 1 The proposed removal mechanism of dimethyl phthalate by ECO.

$$O_2 + H^+ + e^- \rightarrow {}^{\bullet}OOH \quad (35)$$

$$^{\bullet}OOH + H^+ + e^- \rightarrow H_2O_2 \quad (36)$$

(Fig. 11). Zazou et al. (2019) compared anode materials Pt, BDD, and Ti/RuO_2–IrO_2 (DSA) for removal of *o*-DCB; about 90.0% COD removal was achieved under optimal conditions. Moreover, the authors proposed a detailed degradation pathway for *o*-DCB in the BDD-based ECO system. The *o*-DCB firstly transformed into 2-chlorophenol by oxidative dechlorination with •OH; further dechlorination changed it to catechol; it was then transformed into o-benzoquinone and o-benzoquinone by the effect of •OH. Both compounds produced formic acid, maleic acid, malonic acid, and oxalic acid by •OH oxidation. Lastly, the four intermediate products finally completely mineralized into CO_2 and H_2O (Fig. 12).

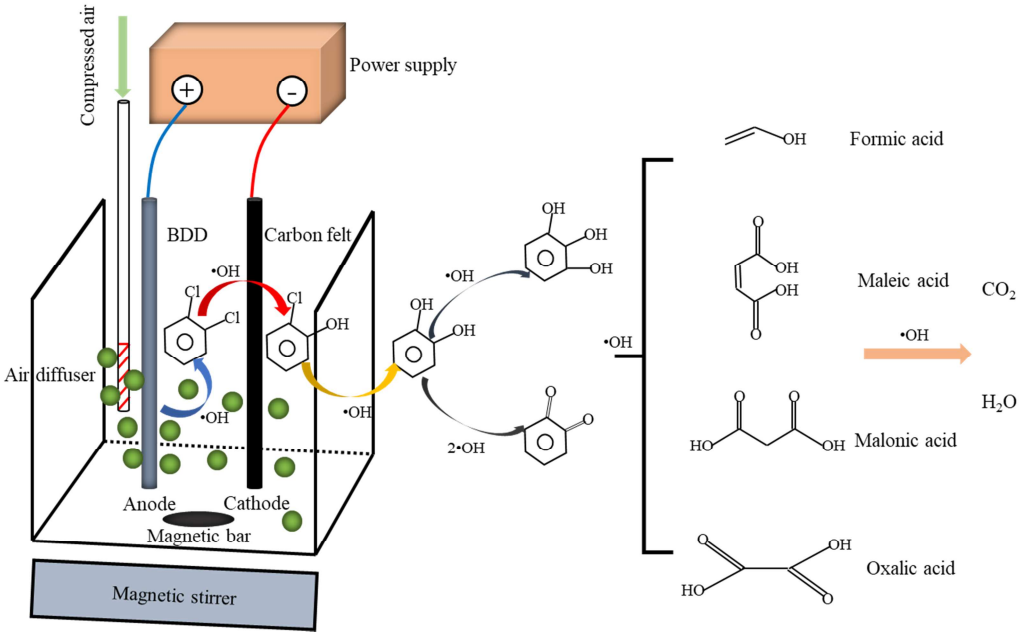

Figure 12 The proposed removal mechanism of 1,2-dichlorobenzene by ECO.

Table 7 Summary of studies addressing for the removal of specific organic pollutants by ECO.

Compounds	Reaction time (min)	Initial conc.	Anode	Electrolyte	Current density	Optimal conditions	% Compound	% COD	% TOC	Ref.
Phenol	5.00	250 mg/L	Iron	Raw effluent	1.00 mA/cm^2	pH 3.00, H_2O_2 500 mg/L,	93.3	87.5	-	(Gümüş and Akbal, 2016)
Phenol	30.0	250 mg/L	Iron	NaCl	0.800 mA/cm^2	H_2O_2 37.2 mM, pH 5.20, electrical conductivity 125 µS/cm, stirring speed 100 rpm, inter-electrode gap 4.00 cm	100.0 (5.00 min)	84.0	52.0	(Khatri et al., 2018)
Phenolic	-	10.0 mM	Platinum	0.500 M H_2SO_4	8.00 mA/cm^2	Electrode potential 6.00 V, T = 25.0 °C	-	-	-	(Nady et al., 2017)
Nitrotoluene	420	TOC 450 mg/L	Platinum	Raw effluent	-	Electrode potential 6.00 V, T = 30.0 °C, O_2 = 150 mL/min, pH = 0.100 and Fe (II) = 150 mg/L	100.0	-	-	(Chen and Huang, 2014)
BPA	30.0	1.00 mg/L	Boron-doped diamond	0.850 mg/L Na_2SO_4	-	Voltage 5.00 V, Ultrasound frequency, 24.0 kHz, Electrode distance 2.00 cm	90.0	-	-	(Dietrich et al., 2017)
1,2-dichlorobenzene	180	14.7 mg/L	DSA/Carbon felt Pt/Carbon felt BDD/Carbon felt	50.0 mM Na_2SO_4	-	Current 500 mA, BDD anode and CF cathode, 50.0 mM Na_2SO_4 and 0.100 mM Fe^{2+}, pH 3.00	-	-	90.0	(Zazou et al., 2019)
DMP	24.0	150 µM	CeO_2	0.100 M Na_2SO_4, NaH_2PO_4, $NaNO_3$ or NaCl	-	pH 3.00	93.9	-	-	(Ren et al., 2018)
Petrochemical wastewater	480	COD 2.75×10^30 mg/L	PbO_2/Ti Ti/Pt BDD	Raw effluent	40.0 mA/cm^2	Inter-electrode gap 10.0 mm, 60.0 °C	-	98.7	-	(dos Santos et al., 2014)
Petrochemical wastewater	79..	COD 1.40×10^3–1.70×10^3 mg/L	Aluminum	Fe^{2+} solution	25.0–80.0 mA/m^2	Inter-electrode gap 3.00 cm, current density 68.7 mA/m^2, pH 3.06, H_2O_2/petrochemical wastewater 2.14 mL/Land H_2O_2/Fe^{2+} 4.99	--	53.9	-	(Davarnejad et al., 2014)

73.2	Iron	Inter-electrode gap 3.00 cm, current density 59.7 mA/m^2, pH 2.76, H_2O_2/petrochemical wastewater 1.23 mL/Land H_2O_2/Fe^{2+} 3.65	67.3

4 Future perspectives

Petrochemical wastewater is typical industrial wastewater and contains numerous specific organic pollutants that are toxic to plants, animals, ecosystems, and human beings. Plenty of attention has been focused on its treatment from governments, environmental rescue institutions, researchers, and individuals, owing to the difficulty of ensuring high-strength pretreatment and low-concentration advanced treatment. In this review, we concentrated on both state-of-the-art pretreatment and advanced treatment technologies for the removal of specific organic pollutants; we also discussed the potential of these processes to provide bioenergy recovery from petrochemical wastewater. Some promising and feasible perspectives have been provided for future research with practical applications as follows:

Anaerobic digestion or hydrolysis acidification are recommended for high-strength and biodegradable petrochemical wastewater pretreatment. Anaerobic digestion and hydrolysis acidification are economical and feasible processes to ensure petrochemical wastewater pretreatment at an industrial scale; however, great efforts should be made to explore more suitable technology and optimal parameters for different and unrelated specific organic pollutants, especially, mixtures of specific organic pollutants. Particularly, dominant microbes and specific organic pollutants degradation pathways should be explored thoroughly for promoting this field of research.

Bioenergy recovery displays better prospects as compared to degradation/removal. Bioenergy recovery from specific organic pollutants is an economical and environment-friendly way of ensuring cleaner production and circular resource utilization in the society. The methods discussed here can provide high yield under steady conditions of operation with little chances of safety issues and air pollution.

Oxidation-related advanced treatment processes should be promoted. The advanced treatment methods discussed in this review can help to achieve complete specific organic pollutant removal in a very short time so that strict discharge standards can be fulfilled. However, present studies are mostly restricted to lab-scale applications, which has resulted in the increase in costs and secondary pollution. Furthermore, different removal mechanisms for specific organic pollutants should be thoroughly studied to guide industries effectively.

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Highlights

- A good toxicity reduction of petrochemical wastewater pretreatment is hydrolysis acidification.
- Specific organic pollutants removal is the key of petrochemical wastewater advanced treatment.
- The function of microbial community mainly influencing pretreatment effect.
- Bioenergy recovering from petrochemical wastewater is economic and viable measure.

Key Abbreviations

Anaerobic-anoxic-oxic	$A_1/A_2/O$
Anaerobic-anoxic-oxic membrane bioreactor	$A_1/A_2/O$-MBR
Acetone–butanol–ethanol wastewater	ABE
Anaerobic baffled reactor	ABR
Acrylonitrile–butadiene–styrene	ABS
Anaerobic hybrid reactor	AHR
Anaerobic migrating blanket reactor	AMBR
Anoxic-oxic	A/O
Anaerobic packed bed baffled reactor	AnPBBR
Anaerobic migrating blanket reactor	AnMBR
Anaerobic-anoxic-aerobic batch fed moving-bed batch model	AnOAMBR
Anaerobic-aerobic-biofilm reactor	A/O-BR
Boron-doped diamond	BDD
Bisphenol A	BPA
Continuous-flow packed-bed biofilm reactor	CFPBR
Methane	CH_4
Catalytic oxidation	CO
Chemical oxygen demand	COD
Continuous stirred tank biological reactor	CSTR
1,2-dichlorobenzene	*o*-DCB

2,4 dichlorophenol	DCP
1,2-dichloropropane	1,2-DCP
Dimethyl phthalate	DMP
Dissolved oxygen	DO
Dissolved organic matter	DOM
Electrochemical oxidation	ECO
Expanded granular sludge blanket reactor	EGSB
Ethylene oxide/ethylene glycol	EO/EG
Ethylene glycol	EG
Extracellular polymeric substances	EPS
Fischer–Tropsch	FT
Hybrid anaerobic reactor	HAR
Hydraulic detention time	HRT
Hybrid up flow anaerobic sludge blankets	HUASBs
Membrane bioreactor	MBR
Mono-ethylene glycol	MEG
Microaerobic hydrolysis-acidification-anoxic-oxic	MHA-A/O
Nitrobenzene	NB
Nitrobacteria	NOB
Nonylphenol	NP
4-nitrophenol	4-NP
Ammonia nitrogen	NH_4-N

Organic loading rate	OLR
Polycyclic aromatic hydrocarbons	PAHs
Photocatalysis oxidation	PCO
Methyl tert-butyl ether	PO/MTBE
Purified terephthalic acid	PTA
p-toluic acid	pTOL
Mixed liquor recirculation ratio	R
Stepped anaerobic baffled	SAB
Sequencing batch reactor	SBR
Submerged membrane bioreactor	sMBR
Soluble microbial products	SMPs
Styrene and propylene oxide	SPO
Sulfate-reducing bacteria	SRB
Scrap zero valent iron	SZVI
Tetrachlorobisphenol-A	TCBPA
Total nitrogen	TN
Total phosphorus	TP
Up-flow anaerobic fixed bed	UAFB
Upflow anaerobic sludge blanket	UASB
Volatile fatty acid	VFA

Declaration of interests

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

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

No conflict of interest exists in the submission of this manuscript, and manuscript is approved by all authors for publication. I would like to declare on behalf of my co-authors that the work described was original research that has not been published previously, and not under consideration for publication elsewhere, in whole or in part. All the authors listed have approved the manuscript that is enclosed.