Advances in treatment of coking wastewater – a state of art review

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ABSTRACT

Coking wastewater poses a serious threat to the environment due to the presence of a wide spectrum of refractory substances such as phenolic compounds, polycyclic aromatic hydrocarbons and heterocyclic nitrogenous compounds. These toxic substances are difficult to treat using conventional treatment methods alone. In recent years much attention has been given to the effective treatment of coking wastewater. Thus, this review seeks to provide a brief overview of recent developments that have taken place in the treatment of coking wastewater. In addition, this article addresses the complexity and the problems associated with treatment followed by a discussion on biological methods with special focus on bioaugmentation. As coking wastewater is refractory in nature, some of the studies have been related to improving the biodegradability of wastewater. The final section focuses on the integrated treatment methods that have emerged as the best solution for tackling the highly unmanageable coking wastewater. Attention has also been given to emerging microwave technology which has tremendous potential for treatment of coking wastewater.

Key words: coking wastewater, integrated, refractory, toxic, treatment

HIGHLIGHTS

- Cost-effective treatment of coking wastewater is a challenge.
- A brief overview of recent developments that have taken place in the treatment of coking wastewater.
- Discussion on integrated treatment methods.
- The conventional biological treatment has been replaced with more efficient and practical solutions.
- Technical and economic viability studies related to treatment methods.

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GRAPHICAL ABSTRACT

ABBREVIATIONS

PAHs  Polycyclic Aromatic Hydrocarbons
BOD  Biochemical Oxygen Demand
COD  Chemical Oxygen Demand
DO  Dissolved Oxygen
NH₄-N  Ammonium Nitrogen
TOC  Total Organic Carbon
TN  Total Nitrogen
TDS  Total Dissolved Solids
SS  Suspended Solids
NTU  Nephelometric Turbidity Units
ANAMMOX  Anaerobic Ammonium Oxidation
HRT  Hydraulic Retention Time
MBR  Membrane Bioreactor
MBBR  Moving Bed Bioreactor
UASB  Upflow Anaerobic Sludge Blanket Reactor
RSM  Response Surface Methodology
INTRODUCTION

In recent years, there has been a rise in the number of steel products owing to the growth in the economy. This has directly led to the establishment of coke oven operations which are an integral part of the steel manufacturing process. Coking wastewater originates during the heat recovery process of coke quenching, coke oven gas purification and recovery of coal derivatives (Duan et al. 2015; Liu et al. 2018b). The complex and toxic nature of wastewater is evident with the existence of poor biodegradability refractory constituents, such as polycyclic aromatic hydrocarbons (PAHs), phenolic compounds, heterocyclic compounds, thiocyanate, ammonia, and cyanide (Wang et al. 2015b; Zheng et al. 2019b). The organic pollutants in coking wastewater are known to be carcinogenic in nature and can cause conjunctivitis, respiratory problems, immunotoxicity, reproductive toxicity and genotoxicity (Farris 2014). Furthermore, the complex nature of these pollutants raises a question on the environmental fate and bioaccumulation in the ecosystem. Therefore, coking wastewater must be treated properly to reduce its adverse impacts on the environment and human health. But even after meeting the discharge standards, coking wastewater still shows some potential risk to the environment in the form of genotoxicity (Na et al. 2017).

The complications in removing the refractory organics and the inhibitory effects of compounds in the treatment process are stumbling blocks for effective treatment of coking wastewater. For instance, the presence of phenol in coking wastewater suppresses the bacterial degradation of thiocyanate (Liu et al. 2016). The nitrification process in biological treatment is significantly affected when the concentration of phenol exceeds 200 mg/l (Liu et al. 2018b). Similarly, thiocyanate also shows noxious behavior similar to phenol under certain conditions (Kim et al. 2008). Cyanide removal has shown limitations pertaining to its toxic and inhibitory nature (Pueyo et al. 2016). Free cyanide concentrations above 0.2 mg/L cause serious inhibition of nitrification (Sharma et al. 2012). The inhibitory nature of compounds present in coking wastewater is justified by many research works (Pereira et al. 2014; Guo et al. 2017; Oshiki et al. 2018). In order to minimize the toxicity of coking wastewater, conventional treatment techniques like air stripping, solvent extraction, and activated sludge treatment are used. But the presence of wide range of recalcitrant compounds makes the conventional techniques inefficient (Wang et al. 2015b; Zheng et al. 2019b). More refined and efficient biological techniques using a combination of anaerobic, anoxic and aerobic processes have succeeded in treating the wastewater to an extent (Yu et al. 2015; Liu et al. 2018c). The anaerobic process as the first step in biological process reduces the toxicity and improves biodegradability for further aerobic oxidation (Liu et al. 2018b). But in order to achieve better removal efficiency, the modified biological treatment should be followed by advanced treatment. The coking wastewater can be successfully treated and brought to reuse only if the treatment methods are integrated and sequenced properly (Pal & Kumar 2014; Zhu et al. 2018).

For the last decade, most of the research works have been oriented towards effective treatment of coking wastewater with proper integration of biological treatment with advanced treatment techniques (Liu et al. 2016; Kumar et al. 2017; Xue et al. 2017; Wang et al. 2019). Investigation towards enhancing the biodegradability of coking wastewater has also been in the limelight of research (Duan et al. 2015; Yu et al. 2015). Microbial degradation studies (Wang et al., 2015b; Singh et al. 2018) have been carried out for the identification of bacterial strains and their performance in removal of various contaminants. The addition of a metabolic substrate has helped in enhancing the degradation by amplifying settlement performance and biomass formation (Zhou et al. 2017). To further improve microbial degradation, algal biomass has also been used to treat cokes wastewater using electrochemical techniques (Ryu et al. 2018). Bioaugmentation is slowly emerging as an efficient strategy for enhancing the ability of the microbial community to biodegrade contaminants (Nzila et al. 2016; Raper et al. 2018). Studies (Jing et al. 2009; Bai et al. 2011; Zhou et al. 2013; Gu et al. 2014; Rodríguez et al. 2014) have also shown that fixed attached biological processes offer better process stability with improvement in sludge settling, biomass growth and longer sludge retention time. However, all methods suffer from some limitations and one single method alone does not stand out that is capable of treating coking wastewater without any shortcomings. Furthermore, rigorous operating conditions and instability usually hinder the practical applications of treatment systems (Shi et al. 2018). Even though coking wastewater is considered to be one of the most toxic industrial wastewater, very few reviews have been reported on coking wastewater addressing the problems and the treatment advances that have taken place in current years. This review therefore intends to highlight the recent developments that have taken place in the field of coking wastewater treatment addressing some of the major issues and ways the coking wastewater can be treated more efficiently.
HIGHLIGHTS FROM PREVIOUS REVIEWS ON WASTEWATER TREATMENT

Abdel et al. (2019) described beautifully the various wastewater treatment methods addressing the positive and negative aspects of each method. Figure S1 (Supporting Information) illustrates a brief overview of various treatment technologies that have been employed for the treatment of wastewater. Cost-effective and environmental friendly methods such as adsorption and biological treatment stand out among others as a result of their ease in operation and profitability on the commercial scale. Adsorption of pollutants using cheap waste materials, modified adsorbents and natural polymers has become a commonly used method for treatment of wastewater. In search of highly efficient adsorbents, use of nanoadsorbents and hydrogels has received attention in recent times. The major drawback in adsorption is found to be the problem associated with regeneration of the adsorbent used. Furthermore, emerging processes like advanced oxidation processes (AOPs) capable of degradation of large numbers of pollutants have proved to be one of the highly efficient treatment methods. Production of secondary sludge, the high cost involved, high energy consumption, and formation of harmful byproducts are the major limitations of these processes. The operating conditions for some oxidation processes with their limitations are presented in Table S1 (Supporting Information).

Due to more stringent conditions for wastewater discharge and growing concern on emerging pollutants which are considered to be harmful even if present in trace amounts, alternative wastewater treatment technologies have achieved widespread attention. AOPs are the treatment techniques that have yielded very promising results in removal of various refractory pollutants. The classical Fenton’s process alone has high efficiency in mineralization of various pollutants in wastewater. However, shortcomings like narrow pH range, significant iron sludge production, high costs involved in handling of reagents make it practically unfeasible for application (Zhang et al. 2019). To counteract this, and make it beneficial, it is often used in conjunction with other AOPs. The coupling of Fenton’s reagent with other methods in the form of photo-Fenton, electro-Fenton, sono-Fenton, and heterogeneous Fenton processes overcomes the limitations and improves its feasibility. Electrochemical oxidation is one of those technologies which have proved to be an emerging and promising alternative for wastewater treatment in recent decades (Sasidharan Pillai & Gupta 2016; He et al. 2013; Wang et al. 2015a). They are considered more advantageous than other oxidation processes because of several factors such as smaller footprint, absence of secondary waste generation, mild operating conditions, free of chemicals, and versatility to combine with other treatment techniques. Another major positivity is the ease with which the process can be scaled up for larger applications.

Nanotechnology is another such promising technologies which has wide scope in the field of wastewater treatment. Nanomaterials can be successfully molded and incorporated for specific treatment purposes due to their high surface area and catalytic activities. Their application in the form of nanoadsorbents, nanocomposites and functionalization with active components has yielded positive results in eliminating a wide range of pollutants from wastewater. However, there is a growing concern that nanomaterials may pose a risk to the environment. Some of the drawbacks of nanotechnology have been addressed in previous reviews on wastewater treatment. (Ghasemzadeh et al. 2014). The energy intensive process during synthesis of nanomaterials and detailed characterization is a challenge as more nanomaterials are produced. There is a lack of proper risk assessment of nanomaterials on human health and the environment. Finally, nanotechnology alters the chemical structure of the material generating questions on its toxicity and the threshold needed for measuring nanotoxicity.

BIBLIOMETRIC ANALYSIS

Bibliometric analysis is a convenient statistical tool to evaluate the published documents in a particular research area to understand the trend, metrics and influence of the publication in the related research field (Mao et al. 2021; Santiago et al. 2021). The tool helps in identifying of patterns between relatedness and performance of authors, countries, journals and institutions (Ismail et al. 2021). In view of this, to the best of our knowledge, we hardly found any review on bibliometric analysis of coking wastewater treatment. A bibliometric analysis was thus carried out by analyzing the related published literatures in the Web of Science database from years 2000–2021. The search in the database was done using four sets of terms contained in the title and abstract which included coking wastewater, coking wastewater treatment, coke oven wastewater treatment or coke effluent treatment and steel industry wastewater treatment. The publication search was confined to only articles and review papers. The publications data were then extracted which included author information, journals, citations and institutional affiliation, which was then later used for data analysis. The yearly trend of publications was analyzed for all the four sets of terms used and is depicted in Figure 1(a)–1(c). In the first set of terms coking wastewater, the total number of publications were 825.
The first decade covered less publications on coking wastewater treatment but a considerable growth trend was observed especially in the second decade after 2013, when there was sudden rise in the number of publications. A similar trend was noticed for the other terms used, in which coking wastewater treatment search yielded 626 publications with the highest number of publications in the year 2019. The term coke oven wastewater treatment or coke oven effluent treatment was also used, which resulted in fewer publications (282) with only 74 publications in the first decade (2000–2010) compared with 208 in the second decade (2011–2021). As the coke oven plant is an integral part of the iron and steel industry, a search was also conducted using the term steel industry wastewater treatment. However most of the documents were unrelated to coking wastewater and included results either pertaining to wastewater from some other processes in the steel industry or terms related to steel, industry and wastewater. Table 1 shows the contribution of the top 11 countries in terms of quantity of publications along with the percent contribution, total citations, average citations per publication and h-index.

The extracted data were also used to analyse the contribution of countries in the research field of coking wastewater treatment. It can be clearly seen that China is the leading country with highest number of publications, accounting for 65.88% of the total number of publications having a high h-index (46). The other three countries that had a slight contribution to the publications were India (9.27%), Poland (4.55%), and South Korea (3.61%). However, in terms of the average citations per paper South Korea ranked first, followed by Iran (rank 2nd), Singapore (rank 3rd), and China (rank 4th). The high number of publications from China on coking wastewater can be linked to it being the top steel producing country in the world. Moreover, this rapid industrialization has led the government to add amendments to various laws and regulations in order to control the environmental pollution. This laws have directly influenced the academic research output in industrial

![Figure 1](image-url)
wastewater treatment. Figure 2 represents the top performing journals in the research field of coking wastewater treatment for the past two decades (2000–2021). It can be interpreted from the graph that the top five journals with significant publications contribution are Desalination and Water Treatment (42 publications), Bioresource Technology (35 publications), Journal of Hazardous Materials (35 publications), Water Science and Technology (34 publications), and Chemical Engineering Journal (22 publications). It can also be noted that when it comes to comparisons between the decades, there is a major differentiation in the number of publications. Most of the journals had greater impact in the second decade, having a total count of 203 publications in comparison with less than 50 publications. Out of the overall 253 publications, 80% of the publication contribution fell in the second decade, which shows the emerging nature of the research area.

The extracted data were further analyzed using VOSviewer software (version 1.6.15), to generate bibliometric networks to understand the relatedness of the items in the documents and may be author relationship, occurrence or citation analysis. For these data, the co-occurrence analysis was done and provides visual information of the relatedness of items based on the number of documents in which they occur together. Full counting method was selected to give equal weightage and the number of occurrences to be included in the network was restricted to three. The analysis was done for the keywords ‘coking wastewater’, ‘coking wastewater treatment’, and ‘coke oven wastewater treatment or coke effluent treatment’. The co-occurrence analysis network is displayed in Figure 3, in which the selected keywords are represented by circles and labels. The size of the circle and label highlights the occurrences of the keywords where the bigger size denotes the higher

**Table 1 | Publications contribution of the top 11 countries in the research field of coking wastewater treatment**

| Countries   | Total publications | % Contribution | Total citations | Avg. citations per document | h-index |
|-------------|--------------------|----------------|----------------|-----------------------------|---------|
| China       | 419                | 65.88          | 8,267          | 19.73                       | 46      |
| India       | 59                 | 9.27           | 714            | 12.1                        | 14      |
| Poland      | 29                 | 4.55           | 260            | 8.97                        | 9       |
| South Korea | 23                 | 3.61           | 825            | 35.87                       | 13      |
| Spain       | 22                 | 3.45           | 506            | 23                          | 11      |
| USA         | 22                 | 3.45           | 378            | 17.18                       | 12      |
| Japan       | 15                 | 2.35           | 281            | 18.73                       | 10      |
| England     | 14                 | 2.20           | 171            | 12.21                       | 8       |
| Australia   | 12                 | 1.88           | 264            | 22                          | 6       |
| Singapore   | 11                 | 1.72           | 327            | 29.73                       | 9       |
| Iran        | 10                 | 1.57           | 344            | 34.4                        | 9       |

**Figure 2 | Top 10 journals in terms of publications in the field of coking wastewater treatment.**
Figure 3 | Bibliometric networks for (a) coking wastewater in general; (b) coking wastewater treatment; (c) coke oven wastewater treatment or coke effluent treatment.
frequency of occurrence of that particular keyword. The relatedness of the keyword can be visually understood by the closeness of the terms with each other, which are grouped under clusters having different color scales. The co-occurrences of the connected keywords is defined by the link strength and frequency of occurrences in which the thickness of the linkage is associated with the strength of the link.

The bibliometric networks for each of the keywords used is illustrated in Figure 3. For the first search coking wastewater [Figure 3(a)], five distinctive clusters were identified in which the term ‘coking wastewater’ itself had the largest occurrence having wider linkage and connections. The important keywords in each cluster for all the searched terms are explained below along with the occurrence and total link strength values given in brackets.

* The frequent keywords appearing in the cluster one (red) were ‘phenol’ [9,25], ‘degradation’ [8,39], cyanide [5,17], ‘Fenton’ [4,13], ‘electrochemical oxidation’ [3, 11]. As phenol and cyanide are the major toxic pollutants in coking wastewater, their removal by degradation and AOPs like the Fenton process justify the relatedness of the co-occurrences. For the cluster two (green), common keywords included ‘coking wastewater’ [32,97], ‘removal’ [22,81], ‘oxidation’ [8,32], ‘activated carbon’ [4,15]. For cluster three (blue), ‘PAHs’ [6,27], and ‘biodegradation’ [6,20] were the important keywords. For the adsorption cluster (purple), ‘adsorption’ [11,54], ‘reactor’ [6,22], and COD [3,16] were the common keywords. ‘organic pollutants’ was the major occurring keyword for cluster 5 (black).

* When the search was made for the term ‘coking wastewater treatment’, the number of clusters was still five but most of the keywords were related to the treatment. For the red cluster, keywords such as ‘biodegradation’ [9,40], ‘membrane bioreactor’ [3,9] ‘aromatic hydrocarbons’ [3,15] and ‘COD’ [5,25] occur together signifying their relatedness in coking wastewater treatment. Similarly for the green cluster, keywords like ‘removal’ [27,110], and ‘advanced treatment’ [6, 31], represent the study of advanced treatment for removal of pollutants from coking wastewater. Performance analysis of treatment methods like electrochemical oxidation in degrading pollutants is justified by the occurrence of keywords ‘degradation’ [11,56], ‘performance’ [4,14], ‘electrochemical oxidation’ [5,20]. Likewise, cluster 4 (blue), shows that commonly used treatment methods such as coagulation and adsorption have also been used for the treatment of coking wastewater. The most common keywords for cluster 5 (purple), were ‘oxidation’ [12, 52], ‘phenol’ [11, 49], ‘biological treatment’ [3,11] and ‘thiocyanate’ [5,25].

* The network for the ‘coke oven wastewater treatment’ term was well differentiated and had four distinct clusters in its network. Cluster one (red) included keywords related to removal and treatment such as ‘removal’ [16,65], ‘ultrafiltration’ [7,37], ‘nanofiltration’, ‘nitrogen removal’ [4,21] and ‘effluent’ [4,22]. The second cluster (green) was mostly associated to adsorption which included keywords ‘adsorption’ [11,60], ‘aqueous solution’ [7,38], and ‘activated carbon’ [6,29]. The blue cluster had keywords involving major pollutants studied in coke oven wastewater like ‘phenol’ [16,82], cyanide [14,69], ‘ammonia’ [3,17], and ‘thiocyanate’ [6,30]. For cluster 4 (purple), the keywords were related to biological treatment terms like ‘biodegradation’ [13,65], ‘bioremediation’ [4,22], and ‘bacteria’ [4,16].

All the three search terms related to coking wastewater treatment were combined, which resulted in a total of 854 documents. Out of the total documents, the author contribution towards the scientific publications revealed that Wei had the highest publications (67) followed by Wu (30 publications) and Zhang (20 publications). The documents were classified into subject areas as: Environmental Sciences (42.87%), Chemical Engineering (28.68%), Environmental Engineering (27.98%), Water Resources (17.68%), Biotechnology Applied Microbiology (12.41%). Regarding affiliations, most of the publications originated from Chinese research institutes.

**COKING WASTEWATER GENERATION**

Coal is converted into coke by destructive distillation in the absence of oxygen. The process takes place in coke oven batteries which are heated to remove the volatile substances from coal resulting in the release of coke oven gases. Typically, a coking plant comprises of processes of coal preparation, coking, and byproduct recovery (Figure 4). Coking wastewater loaded with toxic organic and inorganic contaminants is formed during the production of coke in coke oven plants.

The wastewater is formed during the process of coke oven gas cooling, coal derivatives separation and byproduct recovery. Further operations like coke quenching and washing of ammonia stills consume large quantities of water, resulting in wastewater generation laden with toxic compounds like phenol, cyanides, thiocyanate, ammonia, etc. (Pathak et al. 2019). On the basis of literature review, the typical characterization of coking wastewater is presented in Table 2, highlighting the maximum and minimum concentrations generally found in the wastewater. The quantity as well as quality of wastewater generated...
depends on the type of coal used and the operating conditions in the coking process. Even after removal of basic substances like oils, tars and ammonia the coking wastewater is highly contaminated with toxic substances like phenolic compounds, PAHs, cyanide, thiocyanate etc. Most important contaminant which is a prime matter of concern are the phenolic compounds because of their high solubility in water (Pathak et al. 2019).

Complexity and toxicity of coking wastewater

Phenols, which are highly corrosive and toxic in nature, account for about 80% of the total COD in coking wastewater (Chen et al. 2012; Khatri et al. 2018). The inhibitory nature of phenol on microbial activity is manifested in both individual and combined effects (Guo et al. 2017). Studies have shown that phenolics response to degradation varies with conditions in which some compounds exhibit more resilient nature in comparison to others (Sharma et al. 2012; Sharma & Philip 2014). Both phenol and thiocyanate have been shown to deteriorate the nitrogen removal process in an ANAMMOX reactor (Pereira et al. 2014; Osliki et al. 2018). The coexistence of phenol and cresol have been shown to hamper the removal of other contaminants and metabolic intermediates (Oberoi & Philip 2017). The most problematic treatment process deals with the removal of cyanide which is toxic to microorganisms and leads to the inefficient performance of biological treatments (Sharma et al. 2012; Pueyo et al. 2016; Yu et al. 2016). Another major problem related to coking wastewater is associated with the accumulation of toxic hydrophobic PAHs in the sludge. However a recent study (Kong et al. 2018), showed that phenols play a very important role in solubilization of PAHs from sludge phase to aqueous phase. The wide range of compounds present in coking wastewater exhibits different toxicity, inhibition and interactions with each other, thereby making it difficult to treat it effectively.

The typical concentration of these compounds both phenolics and PAHs are presented in Tables S2 and S3 (Supporting Information).

The phytotoxicity of coking wastewater treated with biological means showed higher germination inhibition in comparison to the sample treated with reverse osmosis (Smol et al. 2018). The study highlighted the necessity of monitoring the toxicity degree of coking wastewater. In another study (Na et al. 2017), the raw coking wastewater exhibited acute toxicity to aquatic ecosystems accompanied by genotoxicity. The anaerobic-anoxic-oxic biological treatment removed the fish and embryotoxicity but was unsuccessful in removing the genotoxicity. However, the genotoxicity was reduced using ozonation and Fenton oxidation as post-treatment. Similarly, a study done on toxicity on embryonic development of maize concluded that organic compounds are the main materials in coking wastewater to cause toxicity (Wei et al. 2012). Interestingly, it was observed that...
**Table 2 | Characterization of coking wastewater**

| Sl. no | Parameter       | Minimum | Maximum | Tolerance limit | References                                                                 |
|-------|----------------|---------|---------|----------------|---------------------------------------------------------------------------|
| 1     | pH             | 7.2     | 10.3    | 6.5–8.5        | Ghose (2002), Vazquez et al. (2007), Zhang et al. (2009), Zhao et al. (2009), Lai et al. (2009), Chu et al. (2012), Miuczarek et al. (2014), Duan et al. (2015), Zhou et al. (2015), Sasidharan Pillai & Gupta (2016), Sharma & Philip (2016), Pueyo et al. (2016), Ryu et al. (2017), Zhou et al. (2017), Liu et al. (2016b), Raper et al. (2018), Smol et al. (2018), Maneesh et al. (2018). |
| 2     | COD            | 880     | 16,800  | 250           | Ghose (2002), Vazquez et al. (2007), Staib & Lant (2007), Zhao et al. (2009), Zhang et al. (2010), Bai et al. (2011), Wei et al. (2012), Duan et al. (2015), Sasidharan Pillai & Gupta (2016), Sharma & Philip (2016), Huang et al. (2016), Ryu et al. (2017), Liu et al. (2018b), Zhu et al. (2018), Maneesh et al. (2018), Wang et al. (2019). |
| 3     | DO             |         |         | 4             | Ghose (2002)                                                               |
| 4     | BOD            | 80.6    | 5,450   | 30            | Lai et al. (2007)*, Lai et al. (2009), Zhang et al. (2009), Zhu et al. (2011)*, Zhou et al. (2013), Zhou et al. (2014), Zhang et al. (2014a)*, Zhu et al. (2015), Huang et al. (2016), Sasidharan Pillai & Gupta (2016), Liu et al. (2018b), Zhu et al. (2018). |
| 5     | TOC            | 77      | 4,390   |               | Lai et al. (2007)*, Lai et al. (2009), Zhu et al. (2011)*, Bai et al. (2011), Zhang et al. (2014a, 2014b)*, Zhou et al. (2014), Sharma & Philip (2016), Pueyo et al. (2016), Liu et al. (2018b), Zhu et al. (2018). |
| 6     | NH₄-N         | 90      | 708     | 50            | Bai et al. (2011), Chu et al. (2012), Gu et al. (2014), Miuczarek et al. (2014), Duan et al. (2015), Zhu et al. (2015), Pueyo et al. (2016), Huang et al. (2016), Maneesh et al. (2018), Wang et al. (2019). |
| 7     | Phenols        | 25.6    | 1,650   | 1             | Staib & Lant (2007), Zhao et al. (2010), Wei et al. (2012), Zhou et al. (2013), Gu et al. (2014), Miuczarek et al. (2014), Zhou et al. (2015), Ryu et al. (2017), Liu et al. (2018b), Zhu et al. (2018), Raper et al. (2018). |
| 8     | Total cyanides | 4       | 36.4    | 1             | Ghose (2002), Vazquez et al. (2007), Wei et al. (2012), Sharma & Philip (2016), Pueyo et al. (2016), Zhou et al. (2017), Liu et al. (2018b), Wang et al. (2019). |
| 9     | Free cyanide   | 0.1     | 11.42   | 0.2           | Zhang et al. (2010), Zhou et al. (2013), Miuczarek et al. (2014), Zhou et al. (2014), Sharma & Philip (2016), Bargiel & Zabochnicka (2018), |
| 10    | Thiocyanate    | 30      | 560     |               | Vazquez et al. (2007), Staib & Lant (2007), Gu et al. (2014); Bargiel & Zabochnicka (2018), Liu et al. (2018b), Raper et al. (2018), Wang et al. (2019). |
| 11    | Oil and grease | 5.3     | 80      | 10            | Ghose (2002), Vazquez et al. (2007), Zhang et al. (2010), Wang et al. (2019). |
| 12    | TN             | 370     | 1,820   | 100           | Vazquez et al. (2007), Zhao et al. (2009), Zhao et al. (2010), Duan et al. (2015), Zhou et al. (2015), Zhou et al. (2017) |
| 13    | SS             | 10      | 1,460   | 100           | Ghose (2002), Vazquez et al. (2007), Staib & Lant (2007), Zhang et al. (2009), Zhang et al. (2010), Sharma & Philip (2016), Pueyo et al. (2016), Sharma & Philip (2016), Huang et al. (2016), Ryu et al. (2017). |
| 14    | TDS            | _       | 1,521.35|               | Sharma & Philip (2016), Sasidharan Pillai & Gupta (2016) |
| 15    | Chlorides      | 130     | 2,730   | 500           | Sharma & Philip (2016), Sasidharan Pillai & Gupta (2016) |
| 16    | Alkalinity     | 250     | 560     | 200           | Vazquez et al. (2007), Zhao et al. (2009), Pueyo et al. (2016), Miuczarek et al. (2014), |
| 17    | Turbidity      | 71.2    | 691     |               | Zhao et al. (2009), Zhang et al. (2010), Pueyo et al. (2016), Sasidharan Pillai & Gupta (2016) |
| 18    | Hardness       | _       | 462.76  | 300           | Ghose (2002) |
| 19    | Color          | _       | 2.5 × 10⁴| 550           | Zhang et al. (2010) |

Note: all values are in mg/l except for turbidity (NTU), pH and color.

(*) References are the characterization of biologically treated wastewater rather than raw coking wastewater.

(**) General Standards for Discharge of Environmental Pollutants, Environment Protection Rules, 1989.
photodegradation as an advance treatment increases the toxicity of wastewater on maize development. We can deduce from the studies that coking wastewater is one the most toxic industrial wastewaters, showing toxicity in all forms such as bio-toxicity, ecotoxicity, acute toxicity and genotoxicity (Zhou et al. 2015; Na et al. 2017; Smol et al. 2018). Therefore, integrated advanced treatment using the physicochemical–biochemical combined process is necessary for reducing the toxicity of coking wastewater (Liu et al. 2017b).

TREATMENT METHODS

Biological treatment

The basic principle of biological treatment is the conversion of dissolved and colloidal organics into stable end-products by means of microorganisms (bacteria, fungi, algae). Biological systems can be aerobic/anaerobic processes or the combination of both depending upon the requirement. Biological methods are the most commonly used treatment techniques as they are eco-friendly, cost effective and efficient (Zhou et al. 2017; Zheng et al. 2019a, 2019b). But when it comes to coking wastewater, the presence of refractory organic compounds makes the biological treatment process difficult (Liu et al. 2018b). Ammonia and phenols are among the toxic pollutants of coke wastewater which restrict the biological treatment process (Zheng et al. 2019b). The conventional biological treatment units such as the activated sludge process have limitations such as longer hydraulic retention time (Zhu et al. 2016), instability during fluctuation (Yu et al. 2015), longer startup period (Wang et al. 2011), and high energy consumption (Liu et al. 2018b). To tackle this problem the combination of anaerobic-anoxic-oxic processes along with other hybrid biological systems have now proven to be a better biological treatment method for coking wastewater (Zhou et al. 2014; Liu et al. 2017a). Placing anaerobic process in the first step improves biodegradability and helps in the removal of some inhibitory compounds, ensuring better performance in the subsequent stages (Wang et al. 2011; Zhou et al. 2013; Huang et al. 2016). In one study, the use of a hybrid anoxic reactor with completely mixed aeration tanks utilizing granular biomass under fluidized conditions resulted in the removal of 88% of ammoniacal nitrogen, 90% COD, and almost 99 and 100% for cyanide and phenol (Maneesh et al. 2018). Similarly, a hybrid anaerobic reactor showed more tolerance towards cyanide, providing knowledge on the development of efficient systems (Gupta et al. 2018). In recent times, attached biofilm reactors have been successful in treating high-strength wastewater due to high microbial biomass, long MCRT and their stability against organic and hydraulic shock loads (Zhou et al. 2014; Oberoi & Philip 2017). The biological treatment of coking wastewater is possible using various combinations such as anoxic-oxic (A/O), anaerobic-anoxic-oxic (A/A/O), anoxic-oxic-oxic (A/O/O), anaerobic-anoxic-oxic-oxic (A/A/O/O), anoxic-oxic-oxic-oxic (A/O/O/O), and oxic-hydrolytic-oxic (O/H/O) processes (Wei et al. 2020). Selection of optimum biological treatment for real-scale treatment is a challenging multi-criteria decision problem that is governed by so many variables. Wei et al. (2020) developed a multi-criteria decision model based on an analytic hierarchy process (AHP) using indicators for helping the decision makers in selecting the optimum biological treatment (Wei et al. 2020). The ranking of the criteria layers were technical indicators > economic indicators > environmental indicators > administrative indicators'. The results stated that the anaerobic-anoxic-oxic (A/A/O) combination was best suited for low COD, whereas the oxic-hydrolytic-oxic (O/H/O) process was prioritized for the high COD load.

Nitrogen removal

Complete removal of nitrogen from the wastewater is necessary to prevent eutrophication in water bodies. Nitrification–denitrification is the conventional process used for eliminating nitrogen from wastewater systems. The conventional nitrification–denitrification processes have the drawbacks of higher energy and external carbon source requirements (McCarty 2018). To overcome this problem, researchers have come up with other cost-effective alternatives like anaerobic ammonium oxidation (ANAMMOX), shortcut nitrogen removal processes, modified biological methods etc. (Zhou et al. 2014; Ramos et al. 2016). The ANAMMOX process is a cost-effective and environment-friendly nitrogen removal process as it requires less energy, the addition of organic matter to support the denitrification activity, and produces less sludge (Zhang et al. 2017; Oshiki et al. 2018). The process is a novel and groundbreaking finding which has now been implanted in many full-scale treatment plants because of its smaller footprint. The process in general follows a shortcut pathway of direct conversion of ammonium to nitrogen gas bypassing the intermediate steps, as shown in Figure 5. The ANAMMOX process uses nitrite as an electron
acceptor for ammonium oxidation which is carried out by ANAMMOX bacteria as depicted in Equation (1) (Huy et al. 2015):

\[
\begin{align*}
\text{NH}_4^+ + 1.32 \text{NO}_2^- + 0.066 \text{HCO}_3^- + 0.13 \text{H}^+ & \rightarrow 1.02 \\
\text{N}_2 + 0.26 \text{NO}_3^- + 0.0066 \text{CH}_2\text{O}_{0.5}\text{N}_{0.15} + 2.03 \text{H}_2\text{O}
\end{align*}
\] (1)

For the ANAMMOX process to take place, half of the ammonia present in wastewater is converted to nitrites by ammonia oxidizing bacteria in a process called partial nitritation (PN). The typical temperature in which the ANAMMOX bacteria operate usually ranges between 20 °C and 43 °C (Huy et al. 2015). Some of the challenges of this process involve sensitivity to higher C/N ratio, sensitivity to low temperature, slow bacterial growth and longer startup period. Microbial studies show that the ANAMMOX bacteria grow preferentially in granular sludge, forming a network with other heterotrophic microorganisms (Yang et al. 2013; Huy et al. 2015).

The nitrogen removal with PN-ANAMMOX is possible for low C/N ratio wastewater treatments, and approximately 30–40% of the overall nitrogen removal cost will be saved compared to the conventional process and does not require an organic carbon source (Guo et al. 2017). The application of the ANAMMOX process for coke oven wastewater treatment has been limited due to the inhibition of the process by phenol and thiocyanate (Yang et al. 2013; Pereira et al. 2014). Denitrification without adding external carbon source can even be done utilizing the aromatic compounds in the influent in a granular biomass reactor (Ramos et al. 2016).

**Microbial degradation**

Microbiology is the backbone of biological treatment for improving treatment performance. The diversity of microbial population and species richness is dependent on different biological treatment processes, operation modes, temperature, and flow rate (Zhu et al. 2015; Liu et al. 2018c). Microbial community distribution using gene sequencing analysis has helped in understanding the correlation of species with the degradation of biorefractory compounds (Huang et al. 2016). For instance, *Pseudomonas putida* and *Pseudomonas stutzeri* have been identified as potential bacterial strain for the effective degradation of phenol and cyanide (Singh et al. 2018). Micro algal–bacterial cultures have also proven effective in degrading compounds like phenol and ammonia (Ryu et al. 2017). But the proposed process still lacks optimization of the algae/activated sludge ratio and HRT. In one study carried out (Zhou et al. 2017), the settlement performance, size of biomass and dehydrogenase activity (DHA) increased due to the addition of a cometabolic substrate. The improvement in degradation was attributed to a change in bacterial community composition.
Bioaugmentation

Bioaugmentation, which consists of adding specific pollutant biodegrading microorganisms into the microbial community, is emerging as an effective strategy for the removal of recalcitrant compounds from wastewater. The environmentally friendly aspects and less cost involved makes the process promising for wastewater treatment (Nzila et al. 2016). Quinoline, which is difficult to treat in nature, was effectively biodegraded through bioaugmentation by k4 strains isolated from the activated sludge of coking wastewater (Wang et al. 2015b). Similarly, improvement in the removal of PAHs was observed after bioaugmentation using commercially available strains (Raper et al. 2018). The added bacteria accounted for 4.4% of the activated sludge biomass after 25 h but the bacterial cells could not survive after exposure to the river ecosystem. In another study, bioaugmented MBR exhibited better removal of phenol and PAHs in comparison to conventional MBR (Zhu et al. 2015). Other studies (Bai et al. 2011; Zhang et al. 2014a) reported that bioaugmentation affects the shift in the bacterial community structure against shock loads. In another study, good removal efficiencies of over 99% for quinoline and pyridine and 65% COD were obtained (Bai et al. 2011). However, in comparison to the bioaugmented bacteria, the indigenous bacteria were more dominant in the treatment system. The findings concur with another study (Hailei et al. 2017), where bioaugmentation increased the reactor performance by influencing the microbial community structure. However, large-scale application of bioaugmentation still poses some challenges. The survival and maintenance of bioaugmented culture in wastewater is the prime concern associated with bioaugmentation (Nzila et al. 2016). Factors such as protozoan grazing, inoculum size and bacteriophage infection are responsible for low bacterial density in the bioaugmented reactor. Many biotic and abiotic stresses influence the survival of the introduced species and are caused primarily due to changes in temperature, pH, nutrients and substrate limitations, as well as the competition between the bioaugmented species and the indigenous microorganisms. Ways to improve the efficiency of bioaugmentation need to be explored. To overcome the limitations, techniques like encapsulation have been used by bioaugmentation of selected microbial cultures in a confined capsule for effective biodegradation (Kurzbaum et al. 2017). Different pollutants removed from coking wastewater using bioaugmentation are summarized in Table 3.

Biodegradability enhancement of coking wastewater

Due to the presence of refractory constituents, coking wastewater has low biodegradability (BOD5: COD < 0.3) resulting in low efficiency of the biological treatment systems. Even though phenols and amines, which account for the major fraction in coking wastewater, exhibit less toxicity to microorganisms in many scenarios, the inhibition of microbial activity depends on several factors like concentration of pollutant, type of microbial species, type of bioreactor and co-interaction with other compounds. Conversely, other refractory hydrophobic compounds such as PAHs, indole, pyridine etc. present in coking wastewater account for a minor fraction and exhibit greater toxicity to the biodegradation process (Yu et al. 2015). The biodegradability of coking wastewater is often improved by staging a pretreatment before biological treatment or by post treatment of biologically treated wastewater. In recent years, several works (Chen et al. 2012; Duan et al. 2015; Ren et al. 2019; Zang et al. 2021) have been reported on biodegradability enhancement of coking wastewater using different methods.

The biodegradation of PAHs has always been a bigger problem owing to its highly recalcitrant nature. Despite having lower concentrations in coking wastewater, its toxicity and insolubility raise a serious concern for environmental pollution. During the treatment, the PAHs tend to accumulate in the sludge because of their low solubility which causes serious disposal problems if the volume of the treatment plant is considered. Researchers, in recent times, have come up with a solution of improving the solubility of PAHs with the assistance of biosurfactants. A biosurfactant-producing strain (Pseudomonas aeruginosa S5) was isolated from the coking wastewater itself for the in situ degradation of PAHs (Sun et al. 2019b). The strain S5 could effectively remove the high-molecular-weight PAHs from the sludge of coking wastewater.

Similar results were reported in another study when the same strain (Pseudomonas aeruginosa S5) was bioaugmented in the reactor (Zang et al. 2021). The degradation of total PAHs increased by 18.97% accompanied by human toxicity reduction by 26.66% after inoculation with S5. The studies show that emerging technologies like bioaugmentation can effectively degrade refractory organics like PAHs, thereby improving the biodegradability of coking wastewater. It is now well understood that targeting the removal of refractory organics like PAHs can significantly improve biodegradability. A study (Yu et al. 2015), conducted on adsorption dealt with the selective removal of PAHs by using different modified non-π hydrophobic adsorbents. Three types of adsorbents, namely organic modified acid-vermiculites, powdered activated carbon (PAC) and XAD-16 polymer adsorbent, were selected to test the removal of PAHs. It was observed that organic modified...
acid-vermiculites containing non-π functional groups could effectively adsorb the PAHs in comparison to PAC and XAD-16. In addition, the acid-vermiculites could enhance the biodegradability because of the presence of non-π functional groups on the surface.

The oxidative treatment of raw coking wastewater using pulsed corona discharge resulted in enhancement of biodegradability of coking wastewater from 0.14 to 0.43 and degraded toxic substances like phenol and thiocyanate by fragmentation of macromolecular compounds (Liu et al. 2018b). In another study, activated carbon was used as a catalyst for catalytic oxidation of coking wastewater (Chen et al. 2012). The BOD₅:COD ratio increased from 0.23 to 0.84 and the study suggested the use of catalytic oxidation as a pretreatment method before biological treatment to improve the biodegradability. However, the full-scale implementation of the process is still limited by its high catalyst and investment cost.

High mineralization efficiency of phenol and several PAHs was observed when AOPs like non-thermal plasma technology was used for pretreating the coking wastewater (Duan et al. 2015). The BOD₅:COD ratio significantly improved from 0.14 to 0.52 within 100 min of treatment time. Similar improvement in biodegradability was reported when microbubble catalytic ozonation was used as a pretreatment to biological treatment (Liu et al. 2018a). The microbubbles improved the ozone mass transfer and enhanced the production of hydroxyl radicals, leading to better removal of organics. The bubbles also aided in oxygen transfer and the combined system had a total COD removal efficiency of 60.82%.

The biologically treated coking wastewater still poses a risk to the environment due to low biodegradability. In a recent study (Zhang et al. 2020a), a sustainable approach was taken to recycle the powder coke to be used as an adsorbent for treating recalcitrant organic compounds. Modification of the functional groups introduced the hydrophobic interactions, resulting in improved removal of PAHs. Utilization of such highly efficient cost-effective adsorbents can be used for tertiary treatment of coking wastewater to improve its biodegradability. Likewise, a cost-effective method involving multi-stages flow through

### Table 3 | Application of bioaugmentation in coking wastewater

| Pollutant and Compound | Treatment Process | Bioaugmented Bacteria | Removal Efficiency | References |
|------------------------|-------------------|-----------------------|-------------------|------------|
| Pyridine and quinoline | Zeolite–biological aerated filters | *Pseudomonas sp.*, *Paracoccus sp.* | Pyridine: 90% Quinoline: 99% | Bai et al. (2011) |
| Pyridine and quinoline | SBR | Consortium of *Paracoccus sp.*, *Pseudomonas sp.*, and *Shinella zoogloea* | Pyridine: 95–99.9% Quinoline: 85% | Bai et al. (2010), Zhang et al. (2014a, 2014b) |
| Phenol, naphthalene, carbazole, DBF, DBT | Zeolite–biological aerated filters | Immobilized cells of *Arthrobacter sp.* W1 | Phenol: 98% Naphthalene: 94% Carbazole: 92.5% DBF: 94.3% DBT: 93.7% | Shi et al. (2015) |
| Mixture of phenol, quinoline, naphthalene, pyridine, and carbazole | Anaerobic-anoxic-oxic-MBR | Consortium of five strains of *Pseudomonas sp.* and *Paracoccus denitrificans* | Phenol: 99.95% Quinoline: 99.8% Naphthalene: 98.56 Pyridine: 99.67% Carbazole: 94.18% | Zhu et al. (2015) |
| PAHs and thiocyanate | Batch study | Commercially available product rich in *Bacillus sp.* and *Mycobacterium sp.* | Thiocyanate: >99% Σ6PAHs: (51–60.1%) | Raper et al. (2018) |
| Phenol, COD | SBR | *Phanerochaete chrysosporium* | COD: (95.2%–99.0%) | Hailei et al. (2017) |
| Quinoline | SBR | Immobilized strains of *Brevundimonas sp.* | Quinoline: 94.2–94.8% | Wang et al. (2015a, 2015b) |
| Phenol | Batch study | *Pseudomonas sp.* PCT01 and PTS02 | Phenol: >96% | Zhu et al. (2012) |
peroxi-coagulation (PC) was studied for improving the biodegradability of coking wastewater (Ren et al. 2019). The treatment yielded excellent removal of 71.5% COD, 72.3% phenol and 59.4% NH₃-N, thereby improving the biodegradability.

It is often found that during preaerobic treatment, there is depletion of carbon sources, resulting in poor denitrification and incomplete removal of refractory organics (Li et al. 2019). In a study conducted using corncoals as carbon source in up-flow fixed-bed bioreactor, the C/N ratio improved, which resulted in better removal of nitrate and refractory organics from biotreated coking wastewater (Sun et al. 2019a). Results showed that this pretreatment significantly improved the biodegradability prior to biological treatment. Yu et al. (2016) investigated the effect of ferrous sulfate treatment on the removal of cyanide compounds from coking wastewater. They found that the BOD₅/COD ratio of coking wastewater improved from 0.50 to 0.51, indicating improvement in biodegradability. However, it was found that the negative effect in biodegradability occurs with an excess dosage of ferrous ions and hence the determination of optimum ferrous dosage is necessary.

**Integrated treatment methods**

**Advanced oxidation processes**

As discussed in the previous section of highlights on wastewater treatment, AOPs have been gaining importance because of their high efficiency in removing difficult and emerging contaminants. This is evident from many studies related to AOPs in the field of coking wastewater treatment. The coking wastewater must be pretreated followed by biological treatment and if the biologically pretreated wastewater still contains refractory substances, advance treatment can be done to bring the wastewater under safety discharge standards. Many advanced oxidation treatment techniques (He et al. 2013; Sasidharan Pillai & Gupta 2016; Sharma & Philip 2016; Zhang et al. 2018) have been investigated to check their potential in terms of coking wastewater treatment. Wei et al. (2015) applied the ozone/Fenton process for treating coking wastewater and observed that the process was highly effective in degradation of phenol, aniline, quinoline, and ammonia in coking wastewater. Fenton oxidation can be effectively used as a pretreatment for the removal of COD from coking wastewater (Chu et al. 2012). Similarly, in another study the Fenton process was effective in advance treatment of coking wastewater and the process was optimized using RSM (Zhu et al. 2011). To prevent the recombination rate of photogenerated electrons and holes, Wei et al. (2017) came up with a new hybrid technique called photoelectrocatalytic degradation which significantly improved the degradation and mineralization efficiency of phenolic compounds. Other AOPs like electrochemical oxidation (He et al. 2013; Wang et al. 2015a), wet air oxidation (Luan et al. 2017), photocatalysis (Sharma & Philip 2014; Wlodarczyk et al. 2016) and catalytic ozonation (Zhang et al. 2018) have also been used as advanced treatment for achieving desirable effluent quality. However, high investment cost, severe conditions, sludge problems, and high catalyst limit the industrial-scale application of AOPs.

Electrochemical oxidation is one of the AOPs that has attracted the attention of researchers in recent times because of its minimal limitations among the oxidation processes. A recent study (Zhang et al. 2020b) on electrochemical treatment of coking wastewater was conducted using synthesized Ti–Sn–Ce/bamboo biochar (BC) electrodes. The system achieved the removal of 92.91% COD and 74.66% DOC within an electrolysis time of 150 min and current density of 50 mA/m². Gas chromatography-mass spectrometry analysis revealed that most of the soluble organics were either degraded or transformed and the macromolecular compounds were converted to low toxic forms. In another study (Turan et al. 2020), electrochemical oxidation using boron-doped diamond (BDD) anodes could effectively remove 84.13% thiocyanate and 94.67% phenol from coke oven wastewater at optimum conditions of pH (9), current density (43.10 Am⁻²), and electrolyte concentration (Na₂SO₄) = 2.5 gL⁻¹. An integration of electrochemical reactors consisting of three-dimensional electrochemical reactors (3DERs), three-dimensional biofilm electrode reactors (3DBERs) and three-dimensional biofilm electrode reactors for denitrification (3DBER-De) was studied for the removal of ammonia-rich coking wastewater (Wu et al. 2020). The system was able to remove 70.7% of total nitrogen and 55.8% of COD in 20 h at low energy consumption, removing the majority of the nitrogenous compounds. Combining the electrochemical treatment with biological treatment has also resulted in overall treatment of coking wastewater (He et al. 2013; Wang et al. 2015a, 2015b). For instance, when electrochemical oxidation was used as a pretreatment for biological aerated filter, over an operation of 90 days, desirable COD and nitrogen removal was achieved reaching the grade 1 discharge standards in China (Wang et al. 2015a). Similarly in a low-cost graphite electrode-based hydrogen peroxide-assisted electrochemical oxidation, high removal of cyanide, phenol, thiocyanate and aniline was achieved under optimal
Some of the advanced treatment methods that have been used for the treatment of coking wastewater are shown in Table 4.

| Treatment method                  | Type of wastewater | Technical parameters | Treatment efficiency parameters | Influent characteristics | Effluent characteristics | Removal efficiency | References          |
|-----------------------------------|--------------------|----------------------|---------------------------------|--------------------------|--------------------------|---------------------|----------------------|
| Anaerobic-anoxic-oxic-oxic         | Raw                | HRT 116 h            | COD, NH$_4^+$-N                 | COD 1.230 ± 251 mg/l     | NH$_4^+$-N 1.7 mg/l      | COD: 92.3%          | Zhou et al. (2014)   |
| A$_2$/O + catalytic oxidation     | Raw                | HRT 40 h             | COD                             | COD 2.350 mg/l           | COD < 50 mg/l            | COD: 98-99%         | Zhu et al. (2018)    |
| Sand filtration + reverse osmosis  | Biologically pretreated | Filtration area 144 cm$^2$ permeate flux 4.59 $10^{-6}$ m$^3$/m$^2$s | 16 PAHs COD                 | 16 PAHs 15.04 µg/l       | COD 567.3 mg/l           | PAHs: 84.12%        | Smol et al. (2018)   |
| Catalytic ozonation               | Biologically treated |                          | COD, NH$_4^+$-N               | COD 78.1 mg/l            | NH$_4^+$-N 4.93 mg/l     | COD: 67.22%         | Na et al. (2017)     |
| Vertical tubular biological reactor | Raw                | Inflow rate 0.6l/h HRT 30 h pH 7.9 ± 0.2 | COD phenol                  | COD 1.340 ± 263 mg/l     | Phenol 605 ± 128 mg/l    | COD: 93%            | Zhou et al. (2015)   |
| Photodegradation                  | Biologically treated |                           | COD, Cyanide NH$_3$-N         | COD 346 mg/l             | Cyanide 0.54 mg/l        | COD: 9.82%          | Wei et al. (2012)    |
| Photoelectro-catalytic degradation | Raw                | Radiation time – 1.5 h Bias voltage – 1.5 V Phenol, TOC | TOC, COD                    | TOC 193 ± 5 mg/l         | COD 96.985 mg/l         | TOC: 75.3%          | Khatri et al. (2018) |

combined treatment processes

The highly toxic and recalcitrant nature of coking wastewater can be treated effectively with proper integration and management of treatment systems. Over the past years, many works have been reported based on the use of integrated treatment of coking wastewater. The microbial degradation of phenol, quinolone, and pyridine improved when the biological treatment was integrated with the electro-Fenton process (Xue et al. 2017). The integrated system showed higher degradation rates in comparison to the single magnetically immobilized cells exhibiting a synergistic effect between the processes. Many times, the suspended matter present in the wastewater, hinder the oxidation process by partially consuming the oxidizing agent (Pueyo et al. 2016). This negative effect of suspended matter can be eliminated by placing the coagulation step prior to the oxidation process. A combined technology of the oxic-anoxic-oxic process followed by coagulation and ozonation was able to bring the coking wastewater under discharge standards by significant removal of COD and PAHs (Wang et al. 2018).
It was observed that even after the prolonged HRT of 160 h in the biological treatment, the COD values were still much higher than 250 mg/L. But, with subsequent coagulation-ozonation treatment, the COD and PAHs values were reduced to less than 80 mg/L and 0.05 mg/L respectively. The combined technology removed most of the organics, bringing it under the safety discharge standards. A novel multistep three-stage treatment consisting of degreasing and air flotation (pretreatment), aerobic/hydrolysis fluidized bed process (biological) and coagulation/ozonation (post-treatment) was studied for treating the coking wastewater (Liu et al. 2016). The oils and the suspended solids were removed in the pretreatment stage followed by significant reduction of COD, TN, phenol, cyanide, and thiocyanate in the biological treatment. The combined system had removal efficiencies of COD (98.6%), NH3-N (95.4%), and PAHs (80–99%). The residual microorganisms and organics were removed in the post-treatment stage.

The advanced treatment of biologically pretreated coking wastewater incorporating a novel integrated process combining coagulation and adsorption achieved maximum removal of COD and cyanide (Li et al. 2018). The combined treatment attained the final discharge values of COD < 50 mg/L, cyanide < 0.1 mg/L and even lowered the effluent toxicity. The catalytic oxidation using AgNO3 + K2Fe4 as an advanced treatment after A2/O biological treatment was successful in reducing the COD below 50 mg/l by the degradation of refractory cyclic organics (Zhu et al. 2018). In the biological treatment stage most of the cyclic organics like phenol, quinoline, pyridine etc. were reduced, achieving 85% reduction of COD. Since the residual COD was still high (>300 mg/l), catalytic oxidation played a role in further mineralization of the organics. The silver catalyst used was found to accelerate the oxidation capacity of K2Fe4O. The photocatalytic treatment was used after the integrated biological process (anaerobic-anoxic-oxic) in treatment of coke oven wastewater (Sharma & Philip 2014). Coagulation as pre-treatment aided the biological treatment and the post-photocatalytic treatment reduced the COD from 420 mg/L to 90 mg/l in a reaction time of less than 4 h. The combined treatment had an overall treatment efficiency of 96.2%. The Fenton oxidation followed by coagulation/sedimentation could significantly remove most refractory organics and cyanides from a biotreated coking wastewater (Jiang et al. 2011). In order to attain effluent quality for recycling, the wastewater was further treated in a biological activated carbon process. The cost-effective integrated treatment produced a high-quality effluent of <50 mg/L COD and <0.5 mg/L cyanide. Coupling the Fenton oxidation with denitrification has also resulted in good removal of refractory organics and nitrogen species (Razaviarani et al. 2019). The carbonaceous organic matter including refractory organics was successfully removed in the Fenton treatment stage but total nitrogen removal still was not achieved. However, the denitrification stage was responsible for total nitrogen removal and the combined treatment had an overall TOC removal >90%. Similar reduction of COD (>90%), 100% phenol and cyanide (>90%) was achieved in a batch study when the Fenton oxidation was integrated with adsorption for the treatment of coking wastewater (Verma & Chaudhari 2020). A synchronized oxidation-adsorption post-treatment was carried out for the treatment of biologically pretreated wastewater. The coupled treatment achieved mineralization of organics by simultaneous oxidation of Fe2+ to Fe3+, hydrolysis of Fe3+, and adsorption of organic compounds under mild pH conditions. In a batch study, the two-step combined treatment of Fenton oxidation followed by biological treatment achieved a 99.3% degradation of phenol and complete removal of cyanide from synthetic coke oven wastewater. The bacterial consortium was isolated from the coke oven plant and *Pseudomonas* strain BSPS_PHE2 was found to be the most efficient with 92.45% phenol removal (Tyagi et al. 2020).

Li et al. (2016) found that membrane distillation coupled with precoagulation can be used as an effective advance treatment for biologically pretreated coking wastewater. The selection of proper coagulants like polyaluminum chloride is necessary for the prevention of membrane fouling. The integrated treatment of electrocoagulation–ozonation gave promising results when it was used for the treatment of biologically treated coking wastewater (Das et al. 2021). Under the optimum conditions of ozone generation rate (1.35 mg/s), ozonation time (40 min), current density (100 A/m²), and electrolysis time (30 min), the combined treatment resulted in removal of 99.8% cyanide and 94.7% COD. In another study (Zhang et al. 2014b), the combined process of ozonation and biological aerated filter was effective in abatement of COD, BOD₅, ammonia, and color simultaneously from biotreated coking wastewater. In a recent study (Ryu et al. 2018) bioaggregation via floc forming microorganisms with microalgae was induced using an electrochemical approach. The electrochemical approach simultaneously harvested the algal biomass and successfully degraded the soluble organics and thiocyanate. The study proposed that the algal–bacterial process could be used as efficient post-treatment for coking wastewater.

A sustainable treatment approach was proposed for a full-scale treatment by integrating the biological treatment with adsorption using the indigenous low-cost coke breeze from the steel plant itself (Das et al. 2020). The adsorption using coke breeze was very effective in removal of residual COD, phenol, cyanide and color (95%) from the biologically treated coking wastewater and the spent coke breeze was used for sinter making. The low cost involved and the absence of secondary
pollution makes the overall process sustainable and environment friendly. A novel integration of iron–carbon micro-electrolysis (ICE) was done with the biological treatment to form a micro-electrolysis biological fluidized bed (MBFB) process for the treatment of coking wastewater (Han et al. 2020). Compared to the single process, the combined treatment had high removal rates of COD (92%) and total nitrogen (95%). The active hydrogen and Fe2+ produced in the ICE process promoted the growth of denitrifying bacteria for reduction of nitrate and the oxidation/reduction reactions in the MBFB resulted in the overall total nitrogen removal.

Table 5 | Integrated treatment processes

| Treatment method | Wastewater type | Pretreatment | Treatment efficiency parameters | Influent characteristics | Effluent characteristics | Removal efficiency | References |
|------------------|----------------|--------------|--------------------------------|------------------------|------------------------|--------------------|------------|
| Coagulation + Adsorption | Real coke wastewater | Biologically pretreated | COD, Cyanide | COD <30 mg/l Cyanide <0.1 mg/l | COD: 85.3% Cyanide: 99.4% | Li et al. (2018) |
| Coagulation + Ozonation | Real coke wastewater | Biologically pretreated | COD, PAHs | COD <80 mg/l PAHs 0.6316 g/l | COD: 91.5%–93.3% PAHs: 94.26% | Wang et al. (2019) |
| Biological + Coagulation + Ozonation | Real coke wastewater | Degreasing and air flotation | COD, NH$_3$-N, Cyanide, Phenols, Thiocyanate PAHs | COD 49 ± 8 mg/l NH$_3$-N 3.1 ± 1.2 mg/l Cyanide <0.1 mg/l phenols 0.01–0.16 mg/l SCN$^-$ 0.16–0.78 mg/l PAHs (8.8 ± 2.1 μg/l) | COD: 98.39% NH$_3$-N: 95.4% Cyanide: 99% Phenols: 99% SCN$^-$: 99% PAHs: 99% | Liu et al. (2016) |
| Ozone/H$_2$O$_2$ + activated carbon | Real wastewater | Biologically pretreated | COD | COD 3,690 mg/l | | 75.8%–76.79% | Kumar et al. (2017) |
| Biodegradation + e-Fenton | Synthetic | No | Phenol, pyridine, quinoline, COD | Phenol: 95.15% Pyridine: 84.8% Quinoline: 90.5% COD: 81.5% | Phenol: 95.15% Pyridine: 84.8% Quinoline: 90.5% COD: 81.5% | Xue et al. (2017) |
| Biological aerated filter (BAF) + ozonation | Real | Biologically pretreated | BOD$_5$, COD | BOD$_5$ 20.4 mg/l COD 207.6 mg/l | BOD$_5$: 49.7% COD: 58.8% NH$_3$-N: 75.8% | Zhang et al. (2014b) |
| Biological + photocatalysis | Real | Coagulation | COD | COD 2,122 ± 50 mg/l COD 94 ± 5 mg/l | COD: 96.2% | Sharma & Philip (2016) |
| BAF + electrochemical oxidation | Real | Biologically pretreated | COD NH$_3$-N | COD 180.2–255.8 mg/l NH$_3$-N 5.2–7.6 mg/l | COD 91.5 mg/l NH$_3$-N 0.62 mg/l | COD: 58.12% NH$_3$-N 90.31% | Wang et al. (2015a) |
| Fenton oxidation + biological activated carbon | Real | Biologically pretreated | COD TCN | COD (100–200) mg/l TCN 2.0–7.0 mg/l | COD <50 mg/l TCN <0.5 mg/l | | Jiang et al. (2011) |
Table 5 shows that most of the integrated treatment schemes can successfully treat the coking wastewater to bring it under safety discharge standards. Many other integrated treatment schemes such as catalytic ozonation plus activated carbon (Fang & Han 2018), MBR-catalytic ozonation (Zhu et al. 2017), catalytic ultrasound oxidation-MBR (Jia et al. 2015), MBR-PAC (Jia et al. 2014), and MBBR-photocatalysis (Xu et al. 2015) have been reported to enhance the removal of refractory compounds from coal gasification wastewater. As many of the pollutants present in coal gasification wastewater are found in coking wastewater, this type of treatment approaches can also be applied for the coking wastewater. As a summary, Figure 6 gives a schematic representation of various treatment methods that have been used for coking wastewater treatment as pretreatment, post-treatment or biological treatment.

MICROWAVE TECHNOLOGY – A POSSIBLE ALTERNATIVE FOR COKING WASTEWATER TREATMENT

In recent years, microwave (MW) application in wastewater treatment has gathered considerable attention because of its high molecular level heating and reduction in treatment time. Rapid and selective heating properties can help in the degradation of pollutants in wastewater. However, MW is often incapable of removing the refractory compounds on its own. To counteract this problem, it is often coupled with other treatment processes to improve the efficiency, as well as make the overall process cost effective (Remya & Lin 2011). Efficient treatments like AOPs have the major disadvantage of high energy consumption and operational costs which could vary from pollutant to pollutant. In addition, the use of chemicals and the formation of
byproducts is another issue with respect to environmental sustainability. Therefore in most cases the AOPs are combined with other processes to make the overall treatment economical and feasible. Hence, microwave processes when coupled with the AOPs have helped in reducing the operational cost and also improved the technical problems (Remya & Lin 2011). MW-enhanced catalytic degradation has been an emerging field of research and different MW-absorbing materials with high surface area have been developed to target the pollutant of interest (Remya & Lin 2011). The catalyst used absorbs the microwave energy, forming hot spots on the surface, thus producing selective heating, spur in the molecular rotation and decreasing the activation energy. The greater potential in different applications and ease of operation makes the microwave application in wastewater a promising alternative for processing coking wastewater. For instance, the phenol degradation efficiency reached 99.96%, corresponding to an 88.6% chemical oxygen demand (COD) removal when microwave was coupled with catalytic oxidation in a reaction time of just 4 minutes. (Liu et al. 2018d). The microwave-induced catalysts helped the hydrogen peroxide to adapt to wider pH ranges, thus making the overall process technically feasible to be operated under neutral pH conditions. Similar pollutant degradation within a short span of time was achieved in the treatment of coal gasification wastewater when microwaves were coupled with catalytic oxidation (Xu et al. 2017). Many limited works have been done with regard to the treatment of coking wastewater using microwave technology which gives a wider scope to investigate the technology for practical application in coking wastewater treatment.

DISCUSSION AND FUTURE PERSPECTIVES

As a whole, a definitive cost-effective treatment of coking wastewater is a challenge owing to its complex nature and interactions. From this brief survey of the literature, it has been established that single treatment methods are incapable of handling the coking wastewater effectively. This is evident from many recent works which have been mostly integrated treatment studies to improve the overall efficiency. The conventional biological treatment has been replaced with more efficient and practical solutions such as integration of anaerobic, anoxic and oxic processes but still the treatment time is high. With many compounds of a toxic nature, the specific pollutant degradation through bioaugmentation looks promising. However, the full-scale application still is a challenge with more in-depth studies to be conducted on a large scale to understand the fate of microorganisms. AOPs have now been regarded as an efficient method to remove refractory pollutants. However, they have challenges in terms of technical complexity, huge instrument investment, secondary pollution and harsh reaction conditions, which limit their full-scale practical application. Even though many treatment methods have been conducted for treatment of coking wastewater, the majority of the works focus on the efficiency and many studies have been conducted either at the laboratory scale or at pilot scale. The practical implementation of the treatment methods is scarce with only few methods showing promising signs when upgraded to full scale. Emerging and promising technologies like microwave irradiation and bioaugmentation should be further studied for the treatment of coking wastewater. As it is now well established that integrated methods are the only possible solutions for coking wastewater, the future studies should be directed not only in higher efficiency but the cost-effectiveness and technical/practical feasibility of the proposed treatment methods. Technical and economic viability studies related to treatment methods should be addressed in the future with prime focus directed towards sustainable and cost-effective wastewater treatment.

CONCLUSION

The review intends to highlight the progress that has been made in the field of coking wastewater treatment. Many advance techniques and processes have been successful in treating complex and refractory coking wastewater. However, industrial-scale applications of the processes are seldom owing to various factors like operating conditions, stability, etc. The biological method being the most common and preferred treatment choice has seen major improvements in recent times. Bioaugmentation has emerged as an efficient method, but the fate of bioaugmented bacteria and its impact on the system is still a matter of conflict. Large-scale application of processes like advance oxidation, reverse osmosis, and nanotechnology are costly and some processes even generate toxic byproducts. The integrated treatment scheme seems to be the only viable solution for dealing with coking wastewater and this is evident from the recent progress in works encompassing this field of research. However the balance of cost and effectiveness is necessary for the real-scale applications and needs further attention. With rise in emerging contaminants of refractory natures and recent advances in treatment processes, opportunities still exist in the field of coking wastewater treatment.
CONFLICT OF INTEREST
The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT
Data cannot be made publicly available; readers should contact the corresponding author for details.

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