

Comprehensive review on wastewater discharged from the coal-related industries – characteristics and treatment strategies

Deblina Maiti, Iqbal Ansari, Mohd Ashraf Rather and Arukula Deepa

ABSTRACT

Wastewaters discharged from various coal-related activities deteriorate fresh water quality and inflict possibilities of groundwater contamination. Their characteristics mostly depend on the parent coal properties, though some of the pollutants are cyanide, thiocyanate, ammonia, phenol, heavy metals and suspended solids. This paper has reviewed the treatment techniques along with the characteristics of all such kinds of wastewater and also identified the challenges and future perspectives. Primarily, demineralization of coal can attenuate and control release of pollutants in wastewaters if implemented successfully. Mine water from non-lignite mines can be purified using simple techniques, for its reutilization. Acidic mine water and leachates can be treated using passive bioreactors with microbial activity, different organic substrates and limestone drains. Additionally bio-electrochemical systems, membranes, macrocapsules, zeolite filters, ores, physical barriers, and aquatic plants can also be used at various stages. Coal washery wastewater can be treated using natural coagulants obtained from plant extracts along with conventional coagulants. Nitrification and denitrification bacteria fixed in reactors along with activated carbon and zero-valent iron can treat coke oven wastewater. Some other sophisticated techniques are vacuum distillation, super critical oxidation, nanofiltration and reverse osmosis. Practical use of these methods, wisely in an integrated way, can reduce freshwater consumption.

Key words | acid mine drainage, coal washery, coke plant, leachate, microbes, mine water

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ABBREVIATIONS

A1-A2-O	Anaerobic–anoxic–aerobic biofilm system	WW	Wastewater
ALD	Anoxic limestone drain	ZVI	Zerovalent iron
AMD	Acid mine drainage		
BDD	Boron-doped diamond anode system		
COD	Chemical oxygen demand		
FA	Fly ash		
MBR	Membrane bioreactor		
MW	Mine water		
NF	Nanofiltration		
RO	Reverse osmosis		
SRB	Sulphate reducing bacteria		
TCN	Total cyanide		
TN	Total nitrogen		
TOC	Total organic carbon		
TPP	Thermal power plant		

INTRODUCTION

Rapid increase in the usage of coal has resulted in release of coal-related toxic pollutants through wastewaters (WWs) originating from coal mining sites, washeries, coke plants, and thermal power plants (TPPs). The quality of these WWs depends on the coal properties and the process involved in coal processing. Discharge of these WWs gravely impacts the quality of nearby water bodies, aquatic life and the food chain along with the groundwater (Pal & Kumar 2014).

The above coal-related industries mainly release four kinds of WW. The first of is mine water (MW), which is generated in ample amounts during non-lignite coal mining. It is mostly affected by hardness, suspended solids and bacterial contaminants (Soni & Wolkersdorfer 2016). Comparatively, one other kind of MW also called acid mine drainage (AMD) originating from lignite mines is highly contaminated with metals. Oxidation of pyrite present in the mine overburden dumps in presence of moisture is the actual cause of AMD generation, which also dissolves many toxic metals such as zinc, copper, iron, manganese, cadmium, nickel, chromium, lead, cobalt, and mercury during its passage from the rocks (Equeenuddin *et al.* 2010). Seeping of the AMD into water systems leads to 2–1,200 times more metal concentration in them compared to the world average metal concentration in water bodies. The metals are non-biodegradable and some of them such as lead, cadmium, and mercury, are highly toxic to living organisms. Metals bioaccumulate when passed from a lower trophic level to the next higher trophic level and seriously deteriorate metabolic functions of organisms. Natural reduction in acidity and subsequent metal concentration in AMD is only possible if there are sufficient carbonates in the coal rocks (Miranda *et al.* 2010).

The second kind of WW is that which is generated from coal washeries during coal washing. The mined-out coal is sent to coal washeries through a conveyor belt, wherein its impurities are removed through specific gravity-based processes, to increase its heating value (Ghose 2001). The process generates a huge amount of coal slurry having large amounts of solids, chemical oxygen demand (COD) and metals. This effluent is often disposed of in surface waters which affects aquatic life by having a blanketing effect. Optionally it is also injected into abandoned underground mines (Yan *et al.* 2012). In the event, the process also results in loss of coal fines, usually 6–10 tonnes per day from a single washery plant (Tiwary & Dhar 1994; Ghose 2001).

The third and fourth kind of WW is generated from steel plants and TPPs respectively, which are responsible for steel production and electricity generation from the washed coal. The former makes metallurgical coke from coal through carbonization for further use in blast furnaces, which ultimately generates coke oven WW having highly hazardous nitrogenous, phenolic and other organic pollutants. The contaminants have the property to migrate to long distances from disposal sites and are genotoxic as well as carcinogenic to the living organisms (WHO 1997; Pal *et al.* 2011; Sharma & Philip 2016). Presence of these pollutants in surface

water largely increases the oxygen demand and rapidly destroys the aquatic life. Eutrophication of the water bodies are observed due to rapid phenol degradation (Yu *et al.* 2016). TPPs generate fly ash (FA) through coal combustion. FA contains many heavy metals attached to its alumino-silicate matrix, which are often 4–10 times higher than the parent coal and are readily leachable in aqueous medium (Maiti & Prasad 2016). These metals find their way through wet and dry disposal sites of FA. Wet or slurry form disposal is done in ponds, from which the leachate left after FA settling often overflows to surface water bodies during the monsoon or leaches into groundwater through unlined ash ponds; also the ash itself can spill out through breached earthen walls (Yao *et al.* 2015). Dry disposal of FA creates huge unstable dumps which are the primary cause of slurry erosion during the monsoon. The dumps also lead to blowing of fine particles of FA which can also deposit on water bodies causing turbidity and metal pollution (Stefaniak *et al.* 2015).

In view of the above problems various laws have been formulated, which instruct the related industries to treat the WW up to a standard quality before discharge (EPR 1986; WHO 1997; BIS 2012). In this regard an extensive literature survey has been done to advocate possible treatment strategies for the WWs based on their individual characteristics, recent developments, and eco-friendly nature, while also addressing their benefits and challenges.

MINE WATER AND TREATMENT

Opencast mining encounters a permanent flow of water into mine pits along with subsequent loss of water from the nearby aquifers; as a result the communities living near the mine sites suffer from water scarcity. Annually only 13% of MW is reutilized for sprinkling and dust suppression in sites and a majority amount of the water is disposed of (obtained from an unpublished report of Mines of India). The amount of water depends on the size of mine. For example, different coal mine subsidiaries in India release 0.07–0.41 Mm³ of MW every day and the total amount of water released per day is 1.24 Mm³ (Debnath 2013). The mine water is characterized by dissolved solids, hardness, alkalinity, calcium, magnesium, iron and manganese (Table 1), at concentrations which are comparatively higher than the standard drinking water limits (BIS 2012). However, these contaminants can be easily removed by simple cost-effective techniques to achieve the purity level of drinking water (Figure 1).

Table 1 | Characteristics of mine water, acid mine water, coal washery effluents, coke plant wastewater, and fly ash leachates

Parameters	Mine water ^a	Acid mine water	Coal washery effluent ^b	Coke plant wastewater ^c	Fly ash leachate ^d	Discharge limits ^e
pH	5.02–8.7	2.9–5.8	2.5–8.2	6.5–11.5	4.4–12.9	4–12
Colour, Hazen unit	<2.0	–	–	–	–	<5–15
Turbidity, NTU	–	–	5,387–23,360	84–528	–	1–5
Odour	Agreeable	–	–	–	–	Agreeable
Total suspended solids	–	–	110–30,000	2–712	12–140	15–50
Total dissolved solids	174–1,510	–	291–729	1,920	142–1,743	1,000–2,000
Alkalinity	132–488	–	1	224–17,200	41–681	200–600
Chloride	4–92	–	184	2,730	3–95	350–700
Sulphide	–	–	–	1.4–49	–	0.05–0.2
Sulphate	5–749	489–14,920	66–564	–	49–916	300–500
Nitrate	0.02–3	–	0.15–0.37	–	–	10–20
Ammonium nitrogen	–	–	–	49–520	0.04–0.64	5–10
Total Kjeldahl nitrogen	–	–	–	50–1,625	–	50
Biological oxygen demand	–	–	1,001	60–5,450	–	15
Chemical oxygen demand	–	–	192–6,468	81–16,000	–	100
Total organic carbon	–	–	–	100–4,390	–	50
Phosphorus	–	–	–	–	0.01–0.08	3–30
Fluoride	0.03–2.05	–	–	–	–	50
Hardness	390–712	–	358–720	–	105–985	200–600
Calcium	30–484	136–309	118–133	–	32–17,250	150–200
Magnesium	24–400	67–115	16	–	0.4–352	150–200
Sodium	–	–	30	–	17–601	200
Iron	0.061–0.371	50–4,537	0.13–212	–	0.02–6.7	1–20
Aluminium	–	7–800	–	–	1.4–373	5
Nickel	–	–	0.06	–	0.03–2.1	0.02–0.5
Manganese	0.018–12.93	15–155	0.07	–	0.01–36.4	1–5
Lead	0.018–0.021	–	–	–	0.01–2.8	0.1–2
Cadmium	–	–	0.01	–	0.001–0.8	0.003–0.2
Zinc	0.012–1.533	0.01–4	–	–	0.01–37	1–10
Copper	0.008–0.023	73	–	–	0.01–1.7	0.2–10
Arsenic	<0.01	–	–	–	0.28	0.1–1
Selenium	<0.01	–	–	–	0.002–2.4	0.05–0.5
Chromium	<0.01	–	–	–	0.004–3.3	0.05–0.5
Barium	–	–	–	–	0.1–1.32	0.7–4
Beryllium	–	–	–	–	0.01–0.2	0.3
Mercury	–	–	–	–	0.038	0.001–0.1
Boron	–	–	–	–	0.3–109	0.5–2
Molybdenum	–	–	–	–	0.06–3.92	0.05–1
Lithium	–	–	–	–	0.68–26.3	10
Vanadium	–	–	–	–	0.3–1	1
Phenols	0.002	–	–	50–1,650	–	0.1–1.2
Oil and grease	–	–	1.7–4.6	4.7–1,250	–	5–15
Cyanide	–	–	–	0.1–210	0.01	0.05–2

All parameters are in mg L⁻¹ except pH, colour, turbidity.

^aTERI (2013) and ECL (2014) (non-lignite mines).

^bGhose (2001), Das et al. (2006), Tiwary & Dhar (1994), Menkiti & Onukwuli (2011a, 2011b), Menkiti et al. (2011) and Nnaji et al. (2014).

^cZhang et al. (2009), Zhao et al. (2009), Zhang et al. (2010), Wei et al. (2012), Pal & Kumar (2014), Ozyonar & Karagozlu (2015), Yu et al. (2015), Sharma & Philip (2016), Yu et al. (2016) and Zhao et al. (2017).

^dChu et al. (1978), Eisenberg et al. (1986), Nathan et al. (1999), Baba & Kaya (2004), Sarode et al. (2010), and Silva et al. (2010).

^eBIS (2012), Ministry of Environment and Water Resources (1986) and RGC (2009).

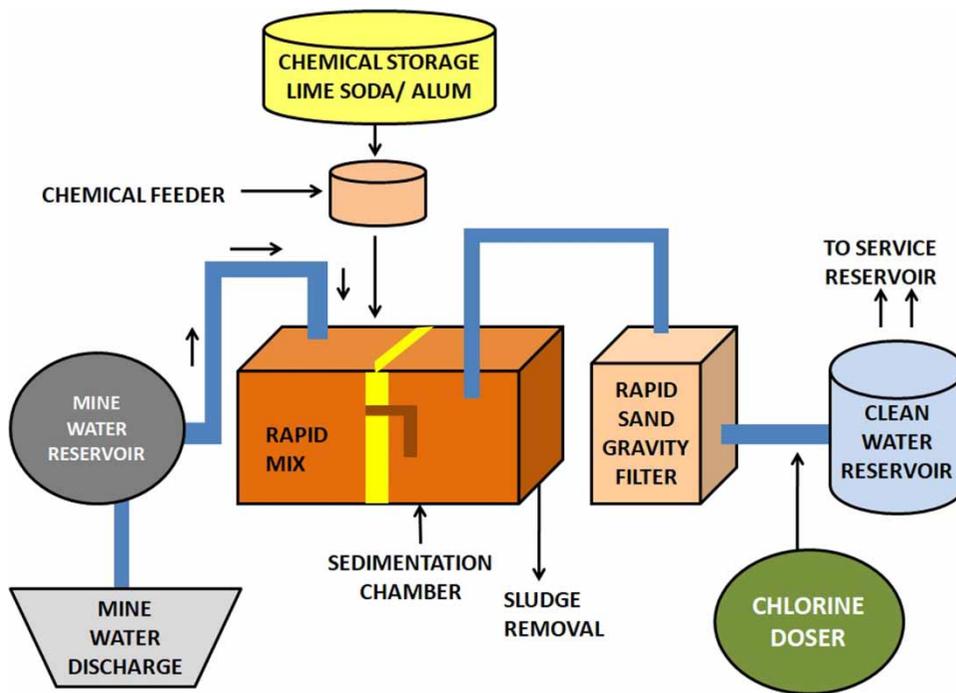


Figure 1 | Low-cost treatment for mine water.

The technique includes removal of floating impurities, followed by storage, and sedimentation of coarser materials in horizontal flow tanks. The dimensions of the tanks would depend on the capacity of the water treatment plant. The settled particles can be removed periodically after formation of a sludge zone of >0.8 m, followed by aeration to remove odours and precipitation of iron as well as manganese. Hardness can be removed by rapid dissolution of lime-soda. Coagulants such as alum conjugated with FeSO_4 or lime can be added to remove turbidity. Final disinfection can be ensured through chlorination or ozone treatment. A few highly sophisticated techniques such as nanofiltration (NF), reverse osmosis (RO) and ion exchange are practised in some mines across the globe, which remove salt ions, total dissolved solids, hardness, alkalinity, ammonia and metals (USEPA 2014). However, they extend the budget of the mining industry and are unfeasible for the developing countries.

On the other hand, pollutants present in AMD (Table 1) are remediated through biological procedures (Table 2) such as passive remediation using sulphate reducing bacteria (SRB) in bioreactors and wetlands. Metabolically the bacteria utilize the carbon and nitrogen source of a substrate and produce hydrogen sulphide in anaerobic conditions, which in turn reduces dissolved metals to sulphides. The later precipitates are ultimately absorbed in sedimentary

deposits. Industrial utilization of this process can treat AMD and induce alkalinity in it. Although the process requires minimal energy, it is less effective at high flow volumes and metal concentrations (Wieder 1989; Greben et al. 2009). This technique can be supported through installing anoxic limestone drains (ALDs), which increase the metal removal efficiency by 30%, add bicarbonate alkalinity and also have a lifetime of >20 years (Hedin et al. 1994; Ziemkiewicz et al. 2003). These ALDs are also affected in presence of highly acidic AMD with a coating of reddish iron oxyhydroxide within 48 h of contact. This can be reduced by using CO_2 (Hammarstrom et al. 2003). Coupling of ALD with SRB (or zinc-tolerant SRB) is known as successive alkalinity-producing systems, which can also be installed as vertical passage systems to reduce area requirement (Kepler & McCleary 1994). This integrated technique has been effective in reducing high sulphate and zinc in AMD (Castillo et al. 2012). Macrocapsules having a pH-sensitive polymer and a phosphate buffer are better than limestone for AMD treatment (Aelion et al. 2009).

Substrates which have been studied in artificially constructed anaerobic reactors for efficient AMD treatment are spent mushroom compost (Dvorak et al. 1992; Christensen et al. 1996), degradation products of grasses to remove sulphate (Greben et al. 2009), mixture of cow manure, mushroom compost, sawdust, gravel, limestone and sediment

Table 2 | Treatment procedures for acid mine drainage

Material used	Scale	Results	Reference
Wetlands or with sulphate reducing bacteria	<i>In situ</i>	8–81% acidity, Fe, Al, Mn, SO ₄ reduced; pH and alkalinity increased from 3 to 7 and from 0 to 1,077 mg L ⁻¹ respectively	Wieder (1989) and McIntire & Edenborn (1990)
Anaerobic reactor + mushroom compost + sulphate reducing bacteria	Pilot	95% of Al, Cd, Fe, Mn, Ni, Zn precipitated as sulfides, hydroxides, carbonates	Dvorak et al. (1992)
Anoxic limestone drains	<i>In situ</i>	17–30% of SO ₄ , Fe, Mn, K, Na, Ca, Mg decreased	Hedin et al. (1994)
Sand bed + crushed stones + sulphate reducing bacteria	Bench	pH increased and metals decreased after 203 days	Christensen et al. (1996)
Permeable reactive wall (organic matter + sulphate reducing bacteria)	<i>In situ</i>	Fe decreased from 1,300 to 40 mg L ⁻¹ , pH and alkalinity increased from 5.8 to 7.0 and from 0 to 600 mg L ⁻¹ respectively	Benner et al. (1997)
Methanol + sulfate reducing bioreactor	Bench	88% SO ₄ reduced, Fe decreased from 100 to 2 mg L ⁻¹ , pH increased	Tsukamoto & Miller (1999)
Chitin	Bench	pH increased from 3 to 7, alkalinity increased from 0 to 235 mg L ⁻¹ , acidity decreased from 192 to 114 mg L ⁻¹ , SO ₄ decreased from 489 to 303 mg L ⁻¹ , >80% Fe, Al, Mn reduced after 9 days	Daubert & Brennan (2007)
<i>Eichhornia crassipes</i> , <i>Lemna minor</i>	Bench	Reduced metals in 21 days	Mishra et al. (2008)
Cellulose degradation products	Bench	SO ₄ reduced by >78%, iron precipitated as FeS	Greben et al. (2009)
Macrocapsules + pH-sensitive polymer + phosphate buffer	Bench + field	pH increased from 3 to 6; metals and phosphate decreased	Aelion et al. (2009)
Zn-tolerant sulphate reducing bacteria	Bench	Completely removed Zn as sulphide sphalerite, wurtzite	Castillo et al. (2012)
Nanofiltration – 2540 membrane	Bench	Removed As, Sb, Pb, Hg at moderate pressure	Sierra et al. (2013)
NaP1 zeolite from coal fly ash	Bench	Zeolite removed As, Ni, Cu, Ca, Fe, Mn at 10 g L ⁻¹	Cardoso et al. (2015)
Mussel shell + sulfate reducing bioreactor	Bench	Removed 86–90% Al, Fe, Cu, Zn; pH and alkalinity increased to 6 and 350 mg L ⁻¹ respectively	Uster et al. (2015)
Passive batch bioreactor-manure + compost + sawdust + gravel + limestone + sediment	Bench	pH and alkalinity increased, SO ₄ reduced by >70%, Fe and Zn removed by >99% after 45 days	Vasquez et al. (2016)
Bio-electrochemical system	Bench	SO ₄ reduced to <550 mg L ⁻¹ ; Al, Fe, Zn was precipitated	Pozo et al. (2017)
<i>Microspora tumidula</i>	Bench	Accumulation of sulphur and phosphorus in algae at pH 5	Oberholster et al. (2017)

(Vasquez et al. 2016), mussel shells (Uster et al. 2015) and coagulants obtained from chitinous material (Daubert & Brennan 2007). More than 95% of the activity of the substrates can be regained by adding reducing equivalents such as methanol to the substrate (Tsukamoto & Miller 1999). Macroalgae can treat AMD in hybrid systems, which in turn also increases the macroalgae chlorophyll content (Oberholster et al. 2017). Metals in AMD can be removed by aquatic plants (*Eichhornia crassipes*, *Lemna minor*) which accumulate 10 times more metals in the roots, without showing any toxicity symptoms (Mishra

et al. 2008), zeolites synthesized from coal FA (Cardoso et al. 2015), and bio-electrochemical systems (Pozo et al. 2017) without any extra chemical dosing and with reduced sludge formation. Earlier field applications in this aspect includes installation of an artificial permeable reactive wall perpendicular to the groundwater flow, which helped in treating AMD in a time period less than a year while staying effective for at least 15 years (Benner et al. 1997). Overall, AMD can be treated by combined strategies (Figure 2) such as wetlands with SRB, macroalgae, aquatic plants, different organic substrates, and ALDs, and can be

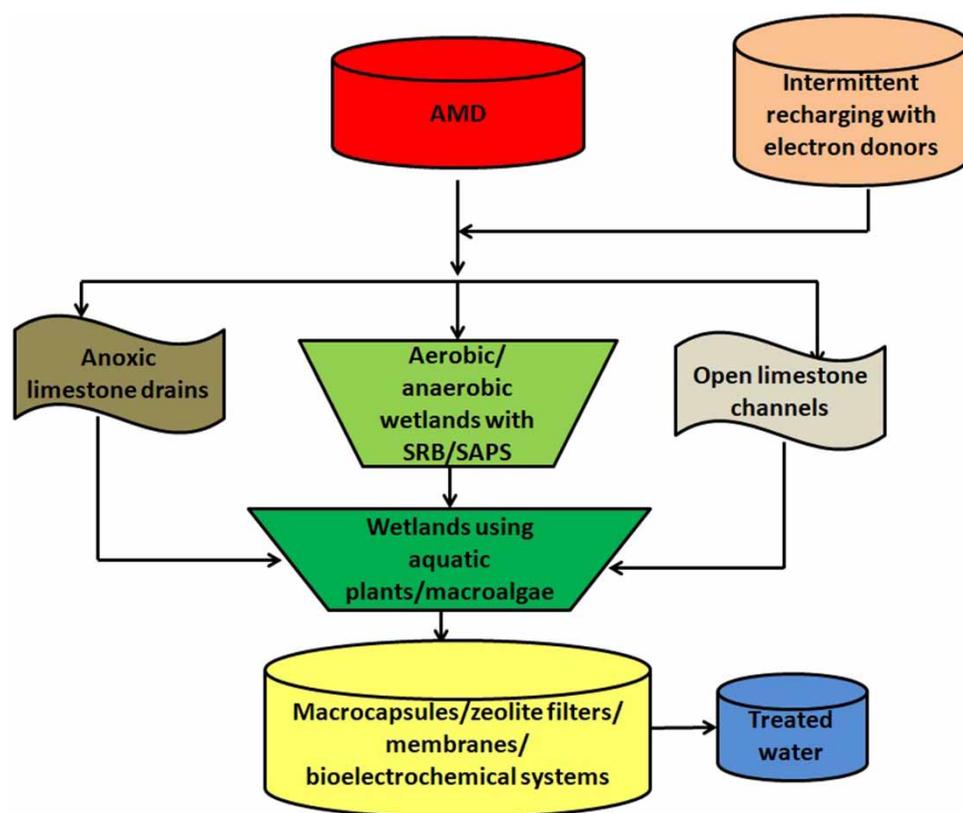


Figure 2 | Treatment procedures for acid mine drainage (AMD: acid mine drainage; SRB: sulphate reducing bacteria; SAPS: successive alkalinity producing systems).

further treated with macrocapsules, natural coagulants, membranes or zeolites.

COAL WASHERY EFFLUENTS AND TREATMENT

The huge amount of WW released by coal washeries (approximately 300–500 m³ of effluent each day per plant) can be treated for reuse in coal washing and to reduce fresh water consumption, because, as per estimates, every 100 tonnes of coal requires 6 × 10⁵ to 2 × 10⁶ gallons (2,270–7,570 m³) of water for washing (Ghose 2001). Various parameters in the effluents (Table 1) have values above standard limits (EPR 1986; WHO 1997; BIS 2012). The pollutants that are mainly coarser solids can be removed through sedimentation, and dissolved solids by coagulants (Table 4), wherein oppositely charged ions destabilize stable colloids and result in settleable flocs (Menkiti & Onukwuli 2011a, 2011b).

Poly-electrolytes such as alum, dioctyl-sodium-sulpho-succinate, N-cetyl-N,N,N-trimethyl ammonium bromide and synthetic flocculants such as Morarfloc-A or True-floc are mostly used (Folkard & Sutherland 1996). Although synthetic flocculants are more effective than alum as they

remove more solids in less time and require a much smaller dose (1 mg L⁻¹) than the latter (30–50 mg L⁻¹), they are linked with many health effects (Ghose 2001). Therefore natural products or coagulants should be considered, which not only remove pollutants, but produce less sludge, ensure potability, and are biodegradable and cost-effective. Literature has reported use of bark resins, ashes, and seaweed extracts by villagers for water treatment (Folkard & Sutherland 1996). Coagulants can be prepared from seeds of *Moringa oleifera*, *M. stenopetala*, *Hibiscus sabdariffa* and *Strychnos potatorum* (Kapse et al. 2017), or from the byproducts obtained during oil extraction from the seeds (Ghebremichel 2004). Due to presence of amino acids such as arginine, *Moringa* seed preparations can outperform Al₂SO₄ to remove 95% of the pollutants in coal washery effluent (Menkiti et al. 2011). Pilot scale studies have also proved the efficiency of coagulants from *Mucuna sloanei* and *Azalia bella* seeds for treatment of WW (Menkiti et al. 2010; Menkiti & Onukwuli 2011a, 2011b). Coagulants from natural starch and chitin also show similar results (Table 3) and can be prepared by electrolyzing a solution of cassava starch (obtained from milled cassava tubers) and calcium hypochlorite in 1:2 ratio followed by mixing

Table 3 | Treatment of coal washery effluents using coagulants

Coagulant	Pollutants	Dosage	Removal efficiency	Reference
Starch + alum + FeCl ₃	Turbidity, suspended solids	30–250 mg L ⁻¹	pH = 4, efficiency =>90%	Nnaji et al. (2014)
Fly ash	Chemical oxygen demand, suspended solids	10 g L ⁻¹	pH = 9, efficiency is >90%	Yan et al. (2012)
Chitin	Turbidity, suspended solids	10 mg L ⁻¹	pH = 8, efficiency is >90%	Menkiti & Onukwuli (2011a, 2011b)
<i>Afzelia bella</i> seed		0.2–0.3 kg m ⁻³	pH = 2–6	
<i>Mucuna</i> seed		0.25 kg m ⁻³		Menkiti et al. (2010)
Periwinkle shell		0.4 kg m ⁻³		Menkiti et al. (2009)

Table 4 | Treatment procedures for coke oven wastewater

Material used	Removal efficiency	Reference
Electrochemical oxidation with PbO ₂ -Ti anode	>90% of COD and ammonia removed	Chiang et al. (1995)
Submerged biofilm + activated sludge	80–99% efficient	Junxin et al. (1996)
A ₁ -A ₂ -O + <i>Burkholderia pickettii</i>	16–59% COD removed	Jianlong et al. (2002)
Adsorption catalytic oxidation	>90% efficient	Hong et al. (2003)
Ultrasonic irradiation + catalytic oxidation + activated sludge	Activated sludge + ultrasonic irradiation or ultrasonic irradiation + FeSO ₄ removed 48–81% and 96% pollutants respectively	Ning et al. (2005)
Pilot plant sequential batch reactor	85–99% of ammonia, COD, thiocyanate, phenol removed	Maranon et al. (2008)
ZVI = 10 g L ⁻¹ active carbon + 30 g L ⁻¹ iron	44% removal of COD at pH 4	Lai et al. (2007)
Nitrifying–denitrifying biofilm	94% removal of COD + ammonia	Rong et al. (2007)
BDD anode system	Removal of TOC + NH ₃ -N	Zhu et al. (2009)
Manganese + magnesium ore	70–100% removal of phenols, COD, sulphide, ammonia, phosphate	Chen et al. (2009)
A ₁ -A ₂ -O-MBR	71–99% removal of COD, phenol, ammonia, TN	Zhao et al. (2009)
Biofilm reactor + ZVI	92% removal of COD, ammonia, TN, phenols, humic acids	Lai et al. (2009)
<i>Phanerochaete chrysosporium</i> on wood chips of Italian poplar	72–87% removal of phenol, COD in 6 days	Lu et al. (2009)
200 g L ⁻¹ activated coke	91% removal of COD, color at 40 °C	Zhang et al. (2010)
Vacuum distillation + NaOH	99% COD removed	Mao et al. (2010)
Fenton oxidation + iron + 0.3 M H ₂ O ₂	44–50% COD and 95% phenol removed at pH <6.5	Chu et al. (2012)
A ₁ -A ₂ -O + MBR + nanofiltration + reverse osmosis	45–99% removal of COD, BOD, ammonia, phenol, TCN, thiocyanate, fluoride	Jin et al. (2013)
Super critical water oxidation + H ₂ O ₂ + salt separator	99% removal efficiency at 650 °C	Du et al. (2013)
Membrane distillation + pre-coagulation	>90% removal of non-volatile organic pollutants; poly-aluminum chloride coagulated volatile organic pollutants	Li et al. (2016)

COD, chemical oxygen demand; TN, total nitrogen; TOC, total organic carbon; A₁-A₂-O, anaerobic–anoxic–aerobic biofilm system; ZVI, zero-valent iron; MBR, membrane bioreactor; BDD, boron-doped diamond; BOD, biochemical oxygen demand; TCN, total cyanide.

with FeCl₃ or mixing the extracted chitin coagulant with alum and FeCl₃ (Menkiti et al. 2008, 2009; Menkiti & Onukwuli 2011a, 2011b; Nnaji et al. 2014). FA-based

coagulants were also studied by Yan et al. (2012). The macrofungus *Pleurotus ostreatus* showed removal of manganese, zinc, nickel, copper, cobalt, chromium, iron, and lead from

50% diluted WW (Vaseem *et al.* 2017). Thus a hypothetical coal washery treatment plant can be proposed which should incorporate an adequate sedimentation time followed by a wise use of coagulants. The process can also ensure a possibility for recovery of coal fines.

COKE PLANT WASTEWATER AND TREATMENT

Every 1,000 tons of coke production requires 4,000 m³ of freshwater and generates 1,000 m³ of coke oven WW during cooling of hot coke over different coolers (Sharma & Philip 2016). This WW contains high amounts of ammonia, phenol, cyanide, thiocyanate, total nitrogen, suspended matter and COD (Table 1) at a concentration higher than the standard permissible limits (EPR 1986). Pollutants removal from the WW is either done by concentration or destructive processes (Figure 3). In the concentration process the pollutants are recovered by dissolving them out by organic solvents which have a high distribution coefficient for the particular pollutant; some of the solvents are methyl isobutyl ketone, di-isopropyl ether and butyl acetate which extracts phenol (Lu *et al.* 2009). Similarly, cyanide is often precipitated as valuable iron complexes using ferric solutions which also remove suspended matter, oil and grease. Ammonia is removed and recovered

through steam stripping, wherein alkaline ammoniacal liquor is reacted with steam. Synthetic zeolites can also remove ammonia through ion-exchange technique and advantageously can be regenerated with low-cost brine (Chu *et al.* 2012; Cardoso *et al.* 2015). The destructive treatment procedures are listed in Table 4, and mainly degrade the pollutants by various agents. For example advanced Fenton oxidation process with H₂O₂ can remove COD and phenol; activated coke can remove maximum COD and color at 40 °C (Zhang *et al.* 2010); zerovalent iron (ZVI) having carbon and iron can also remove COD through coagulation, precipitation and oxidation–reduction processes compared to ferric sulphate or activated carbon alone (Hong *et al.* 2003; Lai *et al.* 2007); electrochemical oxidation in presence of lead dioxide coated titanium anode can remove COD and ammonia (Chiang *et al.* 1995).

However, boron-doped diamond anode systems are more efficient than lead dioxide anodes as the energy consumption is only 60% (Zhu *et al.* 2009). Manganese and magnesium ores can oxidize COD, ammonia, phenols, and sulphides under acidic conditions (Chen *et al.* 2009); supercritical oxidation of water with H₂O₂ and 300% excess oxygen in a continuous-flow reactor can remove 99% of pollutants (Du *et al.* 2013); vacuum distillation with caustic soda can remove 99% of COD (Mao *et al.* 2010). Bio-augmentation is another method which is used in removing

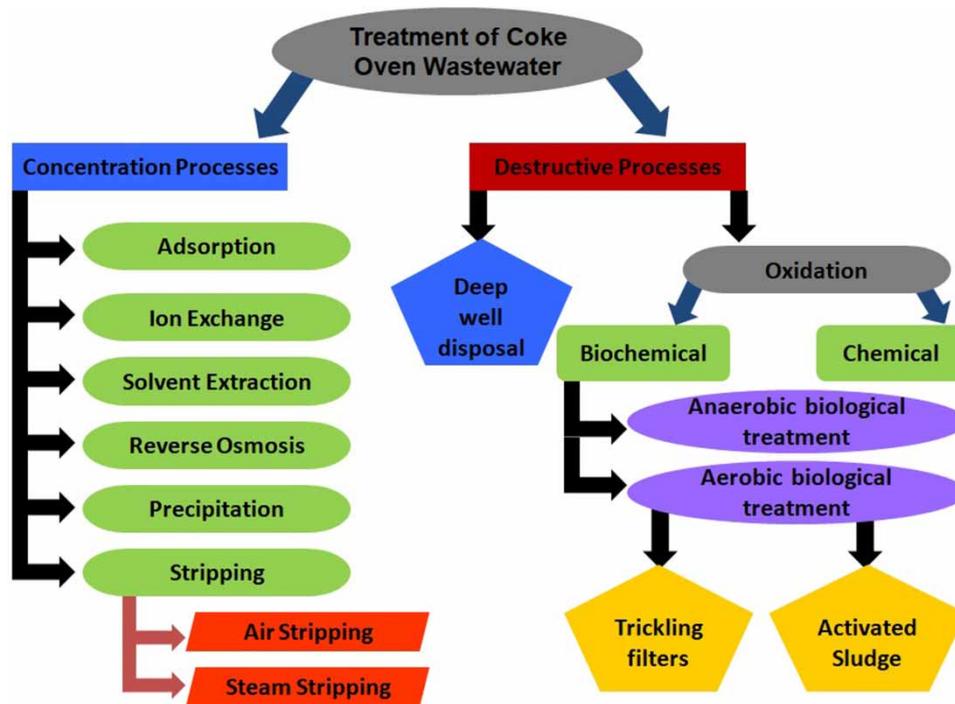


Figure 3 | Treatment processes of coke oven wastewater.

recalcitrant organic compounds from WW. Aerobic bacteria usually degrade pollutants at optimum pH, temperature, oxygen, nutrients, biomass concentrations and residence time (Chu *et al.* 2012). Jianlong *et al.* (2002) used a quinoline degrading bacterium, *Burkholderia pickettii*, for bioaugmenting the oxic reactor of an anaerobic–anoxic–aerobic (A1-A2-O) system. *Nitrosomonas* bacteria can oxidize ammonia to nitrite while *Nitrobacter* oxidizes nitrite to nitrate (Jianlong *et al.* 2002). They can be attached to fixed media to form fixed-film bioreactors or hybrid biofilms to remove pollutants from WW passing through them (Rong *et al.* 2007). WW passed through the immobilized fungus *Phanerochaete chrysosporium* reduced phenol and COD (Lu *et al.* 2009). Biofilm reactors with ZVI have also been used in practical applications (Lai *et al.* 2009). Ammonia can also be oxidized by activated sludge treatment process as well as oxidation ditches, which have a longer residence time than the former (Schroeder 1977; Ning *et al.* 2005), whereas lagoon systems have 5–7 days residence times and higher temperature (Kargi & Uygur 2002). Activated sludge plants generally involve three reactors which remove COD, followed by phenols, ammonia and thiocyanate, and finally nitrate. Methanol can be added as an external carbon source in such systems (Vazquez *et al.* 2006). Sequencing batch reactors (SBRs) also have three reactors which undertake simultaneous clarification along with treatment; the reactors are namely stripping tank, neutralization tank and SBR tank (Maranon *et al.* 2008). Membrane bioreactors (A1-A2-O-MBR) include an extra anoxic basin where the screened raw WW is collected, and then passed to pre-aeration as well as MBR basins to remove pollutants (Zhao *et al.* 2009). However, these reactors require frequent membrane cleaning processes to prevent membrane fouling. The propensity for membrane fouling can be reduced by coupling membrane distillation with pre-coagulation for biologically treated WWs, which can remove salts, and volatile and non-volatile organic pollutants through coagulation with poly-aluminum chloride (Li *et al.* 2016). Submerged biofilm-activated sludge hybrid system can remove maximum ammonia and COD from WW (Junxin *et al.* 1996). Additionally, ultrasonic irradiation can also be coupled with catalytic oxidation or activated sludge process, which shows more efficiency in treatment (Ning *et al.* 2005). Anammox reactors are a type of system which undertakes anaerobic ammonium oxidation using planctomycete bacteria (Aksogan *et al.* 2003). Down-flow hanging sponge reactors have a series of hanging polyurethane sponges with diverse microbial biomass upon which ammoniacal WW is trickled. The diverse microbial biomasses are for establishing ecosystems with

long food chains and reduction in sludge production during treatment (Zhang *et al.* 2008). Practically, A₁-A₂-O systems can be coupled with MBR, NF and RO systems for WW treatment (Jin *et al.* 2013). However, the disadvantage of such systems is further treatment and disposal of the concentrates from NF-RO units.

FLY ASH LEACHATES AND TREATMENT

FA leachates contain varying concentrations of metals, as shown in Table 1, which often surpass the limits of Indian drinking water standards (BIS 2012). Some management practices for these contaminated WWs include mainly prevention of their generation through covering dumps with topsoil and reducing contact of FA with rainwater. The buffering capacity of soil attenuates the metals in organic matter. Transportation of FA as high concentrated slurry during disposal can also minimize leachate generation and water consumption. Demineralization of coal before its burning is a topic of recent research, which can control release of many pollutants in all the WWs. Even if the leachates are formed, their migration to groundwater can be reduced by an adequate liner below the ash ponds, and a leachate collection system wherein the leachate is collected. Along with these the surface water flow should also be controlled (Maiti & Prasad 2016). Attenuating barriers composed of sand, iron, and bentonite can reduce chromium from 25 to 0.0025 mg L⁻¹ in alkaline leachates (Astrup *et al.* 2000). Flue gas desulphurization gypsum can precipitate 40–100% of metals in acidic leachates as sulfides (Jayaranjan & Annachatre 2012). However, metals in acidic leachates can be removed by similar procedures as mentioned for AMD, viz. use of coagulants, bio-electrochemical technology, etc. Extensive research is required for large scale treatment of leachates using latest developments in the field of water treatment.

CHALLENGES

Despite the usefulness of the above treatments, none of them if used alone can be outstanding in terms of amount of wastewater to be treated, contaminant removal potential and treatment cost. The specific challenges of the above techniques are as follows.

- (1) In the case of biological treatments, despite being cost-effective and eco-friendly, they can take a large span of

time to treat the huge amount of WW produced in real conditions.

- (2) Passive remediation techniques are expensive for long-term problems such as AMD and therefore can be combined with other technologies.
- (3) Although advanced oxidation and membrane techniques are primary processes for advanced treatment of coke oven WW, they are expensive and undergo membrane fouling. Studies should be focused on reducing treatment cost along with techniques to control membrane fouling.
- (4) Very few field studies have been found which could uphold the wide applicability and sustainability of the treatment procedures. Hence, future studies should be focused towards scaling up the pilot scale studies with the actual coal-related effluents.

CONCLUSION

A wise analysis of advantages and challenges suggest that the treatment methods can potentially purify water to the drinking level stage when used collaboratively. After further validation studies they can be practically implemented in areas where fresh water is scarce. Advantageously simple treatment techniques can remediate mildly polluted WW with less expense. There is a growing inclination towards WW treatment through safer eco-friendly techniques; hence biological approaches can be used in an integrated manner with conventional techniques to reduce use of chemicals and formation of sludges, and ensure potability, cost-effectiveness and regeneration of treatment materials.

ACKNOWLEDGEMENTS

Authors are grateful to Dr Pradeep Kumar Singh, Director, CSIR-CIMFR, Dhanbad, for continuous support and motivation. The preparation of the manuscript did not involve the contribution of funding sources.

DECLARATION OF INTEREST

All authors confirm that they have no conflict of interest.

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First received 22 March 2019; accepted in revised form 4 June 2019. Available online 11 June 2019