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Decontamination of organic pollutants from wastewater

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Abstract
Water pollution due to organic contaminants is a serious issue because of acute toxicities and carcinogenic nature of the pollutant. Persistent organic pollutants are poorly biodegradable and natural processes cause very little decomposition of these compounds to occur. So these compounds can pollute the environment for a long time. This review provides a perspective on organic pollutants and the techniques of removal of organic pollutants from wastewater.

Keywords: Organic pollutants, wastewater, advanced oxidation process, adsorption, membrane process

Introduction
The quality of our water resources is deteriorating day by day due to the continuous addition of undesirable chemicals in them [1]. The main sources of water contamination are industrialization, civilization, agricultural activities and other environmental and global changes. Few hundred organic pollutants have been found contaminating water resources. The contamination due to organic pollutants is very dangerous due to their various side effects and carcinogenic nature [2]. Industrial wastewater has a high load of different toxic metals, compounds of organic and inorganic nature, chlorinated by-products, dyes, etc. Agricultural wastewater generally consists of pesticides, weedicides, insecticides, organic pollutants etc. In Resins, lacquers and paints industry, basic materials used are higher order alcohols, aldehydes, ketones, some miscible and immiscible organic substances, many of them toxic for the environment [3, 4]. This review gives a overview on common organic pollutants and some of the methodologies for decontamination of organic pollutants from the available literature.

Common organic pollutants
Various types of organic pollutants have been found in different water bodies. These organic pollutants may be pesticides, fertilizers, hydrocarbons, phenols, plasticizers, biphenyls, detergents, oils, greases, pharmaceuticals etc [5, 6, 7]. Their side effects and toxicities were well established and reported by many workers [8]. The general characteristics of organic pollutants are; presence of one or more cyclic ring either of aromatic or aliphatic nature, lack of polar functional groups, and a variable number of halogen substitutions, usually chlorine. Due to these characteristics they are persistent in the environment [9].
Nonpolar organic substances lighter than water form a layer that blocks the flow of oxygen necessary, while polar substances are soluble in water and can be toxic to the fauna or create the effect of eutrophication causing disequilibrium in the biotope. The effluent from textile industry contains chemicals like acids, alkalis, dyes, hydrogen peroxide, starch, surfactants, dispersing agents and soaps of metals [10]. The dye in wastewater severely affect photosynthetic function in plant. They also have an impact on aquatic life due to low light penetration and oxygen consumptions. They may also be lethal to certain forms of marine life due to occurrence of component metals and chlorine. Suspended particles can choke fish gills and kill them. They also detected to hinder with certain municipal wastewater treatment operations such as ultraviolet decontamination etc [11].

Methodologies
Advanced oxidation process
The photocatalysis is one of the techniques which are so called “advanced oxidation processes (AOPs).” These processes can completely degrade the organic pollutants into harmless inorganic substances such as CO_2 and H_2O under moderate conditions.

The AOPs are characterized by the production of • OH radicals which are extraordinary reactive species and capable of mineralizing organic pollutants [12]. They are also characterized by a little selectivity of attack which is a useful attribute as an oxidant for multicompounds, for example, refinery wastewaters. The photocatalysis has been tested on many individual compounds including environmentally relevant pollutants and in many different processes.

Considerable interest has been shown in application of UV/TiO_2 process as an AOP [13]. It is due to the point that TiO_2 has proven to be an excellent photocatalyst material by which many organic substrates have been shown to be oxidatively (in some cases reductively) degraded and undergone fast mineralization under UV light exposure [14].

Javed saien *et al* 2011 investigated real petroleum refinery wastewater, containing a range of aliphatic and aromatic organic compounds, and treated using nanotitania particles, as the photocatalyst in UV/TiO_2 process. Samples were collected from the inlet point of the biological treatment unit. A conic-shape, circulating, and upward mixing reactor, without dead zone, was employed. The light source was an immersed mercury UV lamp (400 W, 200–550 nm). Optimal suspended catalyst concentration, fluid pH, and temperature were obtained at amounts of near 100 $mg\cdot L^{-1}$, 3 and 45 °C, respectively. A maximum reduction in chemical oxygen demand (COD) of more than 78% was achieved after about 120 min and, hence, 72% after only 90 min. Significant pollutant degradation was also relevant under other conditions. The identification analysis of the organic pollutants, provided by means of a GC/MS, equipped with headspace injection technique, showed that different petroleum compounds were degraded with high efficiencies [15].

Suzuki *et al* 2016 studied to decompose persistent organic pollutants in wastewater from chemical factories by using Advanced Oxidation Processes (AOPs). Oxidation reactions involving ozone and •OH radicals and cleavage caused. The effects of AOPs on the decomposition properties of organic compounds with different chemical structures were confirmed in this study. It was necessary to use an O_3-UV-TiO_2 process to decompose compounds containing benzene rings as quickly as possible. However, the Total organic carbon (TOC) was removed at a similar rate when open-chain compounds were treated using the O_3-UV-TiO_2 and UV-TiO_2 processes. So UV-TiO_2 process degraded the open-chain compounds most effectively, and that the O_3-UV-TiO_2 process did not need to be used to decompose open-chain compounds. The TOC was removed more slowly when aromatic compounds were treated than when open-chain compounds were treated. Organic compounds containing carboxyl groups were more easily decomposed than compounds containing aldehyde groups, and compounds containing aldehyde groups were more easily decomposed than compounds containing methyl groups. The removal of the TOC when organic compounds were treated using the O_3-UV-TiO_2 process followed pseudo-zero-order kinetics [16].

Adsorption

Adsorption is considered as the best wastewater treatment method due to its universal nature, inexpensiveness and ease of operation. Adsorption can also remove soluble and insoluble organic pollutants. The removal capacity by this method may be up to 99.9%. Due to these facts, adsorption has been used for the removal of a variety of organic pollutants from various contaminated water sources. Basically, adsorption is the accumulation of a substance at a surface or interface. In case of water treatment, the process occurs at an interface between solid adsorbent and contaminated water. The pollutant being adsorbed is called as adsorbate and the adsorbing phase as adsorbent.

Selection and Type of the precursors

Selection of the precursor for the development of low cost adsorbents depends upon many factors. The precursor should be freely available, inexpensive and non-hazardous in nature. Moreover, for good adsorption results, high contents of carbon or oxygen in the adsorbent moiety are very necessary. Other characteristics include high abrasion resistance, high thermal stability and small pore diameters, which results in higher exposed surface area and, hence, high surface capacity for adsorption. The choice of the precursor depends upon the nature of origin, i.e. inorganic or organic. Organic precursors include plant, animal and other materials having high carbon content, like fruit waste, rice husks, bark, seaweed, algae, peat moss, hair and keratin etc. Industrial organic products include petroleum and fertilizer products. The inorganic precursors include soil, clay, mud, zeolites, ore materials, metal oxides and hydroxides.

Carbonization and activation

During carbonization, pyrolytic decomposition of precursor occurs together with the concurrent elimination of many noncarbon species (H, N, O and S). In this process, low molecular weight volatiles are first released; followed by light aromatics and finally, hydrogen gas. The resultant product obtained is in the form of a fixed carbonaceous char. The pores formed during carbonization are filled with tarry pyrolysis residues and require activation in order to develop the internal surface of the char. Activation may be accomplished via a chemical or physical treatment. In chemical activation, a catalyst is impregnated into the feedstock. The most widely used chemical activators are $ZnCl_2$, H_3PO_4, H_2SO_4, KOH, K_2S and KCNS. In this process, a near saturated solution of catalyst impregnated feedstock is dried to influence pyrolysis in such a way that tar formation and volatilization can be kept at minimum. Resulted product is then carbonized and activated in a single action although two separate temperatures are used. Chemical activation is performed at temperatures between 400 and 800^0C and used for industrially wood based carbons. The catalytically activated product undergoes a post activation treatment for the removal of residual catalyst that can be reclaimed for its reuse [17].

Vieira *et al* 2011 used the coconut shell as the biomass for the removal of textile dyes namely Blue Remazol R160 (BR 160), Rubi S2G (R S2G), Red Remazol 5R (RR 5), Violet Remazol 5R (VR 5) and Indanthrene Olive Green (IOG) dye. It was found that the sorption capacity decreases in order of BR 160 > VR 5 > RR 5 > R S2G > IOG. [18]. Ibrahim *et al* 2010 modified barley straw with NaOH and a cationic surfactant hexadecylpyridinium chloride monohydrate (CPC) to remove two anionic dyes; Acid Blue (AB40) and Reactive Blue 4 (RB4) from aqueous solution. The CPC was observed strongly attached to straw surface with good removal percentage of AB40 and RB4. The adsorption of these dyes on modified straw was favorable at high acidic condition but the desorption process was found relatively less efficient [19]. Dong *et al* 2010 synthesized zeolite from coal fly ash (ZFA), modified with hexadecyltrimethylammonium (HDTMA) and used for the adsorption of bisphenol A (BPA) from water.

HDTMA forms bilayer micelles on external surfaces of zeolites and, thus, greatly enhanced adsorption capacity [20].
Ademiluyi *et al* 2009 investigated the adsorption and treatment of organic contaminants using activated carbon from waste Nigerian bamboo. Waste Nigerian bamboo was carbonized at 400 °C-500 °C and activated with acid at 800 °C to produce granular activated carbon (GAC). Adsorption of organics from the refinery waste on the activated carbon produced was examined at 28 °C. The experimental batch equilibrium data was correlated by Freundlich and Langmuir isotherms. The adsorption data fitted well into the Freundlich isotherm. Breakthrough time of about 1.5 hours was observed for the fixed bed adsorption process. The organic concentration expressed as chemical oxygen demand (COD) was reduced from an initial value of 378 mg/l to 142 mg/l for the first hour, 143 mg/l for the second hour, 152 mg/l for the third and fourth hours, and 156 mg/l for the final hour., which also compare favorably with the refinery effluent specification of 150 mg/l [21].
Different types of nanomaterial like nanosorbents such as CNTs, polymeric materials (e.g., dendrimers), and zeolites have exceptional adsorption properties and are applied for removal of organics from water/wastewater [22]. CNTs have received special attention due to their exceptional water treatment capabilities and adsorption of organics on CNTs is widely studied. CNTs are also effective to remove polycyclic aromatic organic compounds [23]. Nanoporous activated carbon fibers, prepared by electrospinning of CNTs, showed much higher organic sorption equilibrium constants for benzene, toluene, xylene, and ethylbenzene than granular activated carbon [24]. Multiwalled CNTs have been used as sorbents for chlorophenols, herbicides, DDTs, and so forth [25].

Membrane processes

Membrane process has been used to remove organic pollutants, among these technologies the liquid, anion exchange, nanofiltration, reverse osmosis and pervaporation membranes are precedents of this technology [26]. Membrane processes like microfiltration, ultrafiltration, NF, and RO, which are pressure-driven filtration processes, are considered as some highly effective processes. Water/wastewater treatment by membrane techniques is cost-effective and technically feasible and can be better alternatives for the traditional treatment systems since their high efficiency in removal of pollutants meets the high environmental standards. RO is relatively more effective than NF but higher energy consumption in RO makes it less attractive than NF where removal of pollutants is caused by different mechanisms including convection, diffusion (sieving), and charge effects [27].
There are several types of membrane based on it pore size, such as: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). MF Membrane pore diameter ranged between 0.1 to 10 μm and 10-150 μm for the thickness, whereas UF membrane pore diameter ranged between 0.1 to 1 nm, UF membrane give good result for separation of large molecule but not with small molecule. RO membrane has the better result for separation of small molecule until ionic molecule, caused it has smallest pore diameter compared with MF, UF and NF. Although efficient dye removal can be achieved by RO, the high-pressure requirement of RO process (50-70 bar, for some process can reach until 120 bar) jeopardizes its potential due to expensive cost of utilizing high pressure. NF is categorized between UF and RO, therefore NF provide significant advantages such as, lower osmotic pressure gradient, higher permeate flux, multivalent salt retention and can reject organic component with molecular weight 200 until 1000 Da such as dye and salt divalent. Furthermore, NF membrane operating pressure is not high as RO membrane, which is about 2-40 bar. Although NF retention is not good as RO, NF considered more advantageous because its operating cost are lower than RO; the higher the operating pressure, the higher the operating cost [28, 29, 30].
Arcadio *et al* 2013 investigated two commercial thin film composite polyamide reverse osmosis (BW-30) and nanofiltration (NF-90) membranes for sorption of 2-nitrophenol and 2-chlorophenol. Adsorption kinetics and equilibrium on the surface of both membranes were studied. The rate of adsorption was adequately modeled by a linearized form of the pseudosecond-order kinetics in all cases. Adsorption isotherms obtained from equilibrium experiments were quite similar, except for the 2-nitrophenol/BW-30 system, which showed a higher maximum adsorption capacity. Experimental adsorption isotherms were compared with Freundlich and Langmuir models; the Langmuir model yields the best fit in all cases, suggesting the formation of a fouling monolayer onto the membrane surface [31].
Nada Mustafa 2013 studied the removal of dyes from wastewater by reverse osmosis process. Two dyes were used direct blue 6, and direct yellow. Experiments were performed with feed concentration (75 – 450 ppm), operation temperature (30 – 50 ºC) and time (0.2 – 2.0 hr). The membrane used is thin film composite membrane (TFC). The maximum rejection for direct blue 6 and direct yellow are 98.89% and 98.30% respectively. The maximum recovery percentage for direct blue 6 and direct yellow are 17.84% and 18.20% respectively. The maximum concentration factor of direct blue 6 is 1.227 and for direct yellow is 1.272 [32].

Conclusion

Safe water has become a competitive resource in many parts of the world due to increasing population, prolonged droughts, climate change, and so forth. To address the challenges of providing the fresh water in adequate amounts free from organic contaminants the treatment methods need to be stable, economical, and more effective.

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