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A Review on the Adsorption of Phenol Derivatives from Wastewater

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ABSTRACT

Several environmental companies consider phenols compounds to be very dangerous pollutants because they are highly toxic and non-biodegradable, notably their high toxicity in water. For this reason, several processes have been studied by researchers to understand the mechanisms of elimination of phenolic compounds. Adsorption remains the best technique due to its characteristics, in fact, it is non-destructive and simple to use as well as have more other advantages, such as practicality and efficiency and low cost, Therefore, these methods need to be widely developed on an industrial scale to remove phenol derivatives and achieve wastewater quality in accordance with standards. On the other hand, the development of these adsorption methods is highly dependent on new research on materials from abundant natural resources, namely apatites or biomaterials.

Keywords: water; phenol; treatment; adsorption.

INTRODUCTION

Groundwater can be polluted by several sources of pollution namely landfills (leachate) [Benaddi et al. 2022c] WWTPs (domestic and industrial wastewater) [Osmane et al. 2023] or olive mill waste water, these effluents contain in addition to organic matter other toxic elements such as phenol derivatives. Phenol is one of the first constituents of the large family of aromatic organic compounds, it is composed of a benzene ring connected with a hydroxyl group. Its structure is relatively simple; the hydroxyl group is bonded to a carbon atom of the benzene ring. Simple phenols encompass all substituted hydroxylated phenol core molecules. According to this definition, several phenols belong to this group, namely: chlorophenols [Aktaş and Çeçen, 2007], nitrophenols [Yaacoubi et al. 2015], tyrosol, cathecol and methylphenol [El boughdiri et al. 2007]. Many industrial activities use phenol as a disinfectant, Plastics and resin manufacturing, in petroleum refining activities as extraction solvent [Rengaraj et al, 2009], which leads to the contamination of industrial wastewater by phenol and its derivatives. Phenol is a toxic product due to the fact that it is not highly biodegradable, in fact, the biodegradation of natural phenols is generally very good, on the other hand the biodegradability of synthetic phenols is very low, with too slow kinetics, because a number of 'among them have a bactericidal action [Al-khalid and El-naas, 2012], so it presents a real problem for polluted waters and also for the health of human beings, indeed, At high concentrations, phenols can destroy cell walls and denature proteins [Jakobek, 2015]. Several environmental companies consider phenols to be priority pollutants because they are harmful and toxic even at low concentrations. Various processes have been investigated by several researchers to remove phenolic compounds from wastewater [Samimi et al. 2020; Shahriari

Moghadam et al. 2016; Zhou et al. 2012; Sacco et al. 2018; Abid et al. 2019; Belaid et al. 2013; Ouabou et al. 2014; Chedeville et al. 2009; Achak et al. 2009a; Raza et al. 2019; Víctor-Ortega et al. 2016]. The adsorption technique has been widely studied by several authors including the power of several adsorbents to remove phenolic compounds as well as adsorption mechanisms [Benaddi et al. 2022b Datta et al. 2011; Aktaş and Çeçen, 2007; Kulkarni et al. 2013; Dabhade et al. 2009; Abdel-Ghani et al. 2016; Zeboudj et al. 2014; Ozkaya et al. 2006]. Several studies have been conducted to find low-cost adsorbents to encourage the use of this technique [Belaib et al. 2012; El Gaidoumi et al. 2015; Girish et al. 2012; Achak et al. 2009b; Elabbas et al. 2016; Villar da Gama et al. 2018; Sellaoui et al. 2019; Ali et al. 2012; Gupta et al. 2013; De Gisi et al. 2016; Moradi et al. 2018; Bensalah et al. 2018; Saoiabi et al. 2016; Bouyarmane et al. 2013; Bahdod et al. 2009; Alzaydien and Manasreh 2009; Bouyarmane et al. 2010] such as silicate gel and clay [Belaib et al. 2012; El Gaidoumi et al. 2015], bioadsorbents [Girish et al. 2012; Achak et al. 2009b; Elabbas et al. 2016], scoria powder [Moradi et al. 2018], or apatite [Bensalah et al. 2018; Saoiabi et al. 2016; Bouyarmane et al. 2013; Bahdod et al. 2009; Alzaydien and Manasreh 2009; Bouyarmane et al. 2010; Yaacoubi et al. 2015; Benaddi at al. 2022a]. The aim of this paper is to review the municipal phenol treatment technologies. It is envisaged that in-depth knowledge and understanding on advantages and disadvantages of each phenol treatment technique.

PHENOL TREATMENT PROCESSES

The destructive techniques

Biological processes

These processes are not always applicable to industrial effluents due to high concentrations of pollutants, toxicity and very low biodegradability. some microorganisms are able to degrade phenolic compounds including bacteria, [Lika and Papadakis, 2009]. Several authors have studied the biodegradation of phenol derivatives by bacteria [Arutchelvan et al. 2005; Wang et al., 2007]. Problems arise when treating phenolic solutions with a high level of organic load. Indeed, for high phenol loads, phenol can inhibit the growth of microorganisms, this inhibition depends on several factors, namely the concentration of phenol, the temperature and the pH of the medium [Al-khalid and El-naas, 2012].

Oxidation reactions

Over the past decade, much research has been devoted to advanced oxidation techniques (AOT). These techniques have evolved rapidly. They belong to the latest generation of techniques developed to treat water. The principle of AOT is based on the generation of highly oxidizing radical species such as hydroxyl radicals $HO^{\cdot}$ Responsible for the degradation of organic pollutants. In the case of phenol oxidation, there are several methods in the literature, namely the Fenton process [Li et al., 2012], ozonation [Kuosa et al., 2015] and photocatalytic [olga et al., 2018].

These destructive methods have many drawbacks, in fact, the oxidation methods generate sludge and require significant energy requirements, the biological treatment does not make it possible to treat large concentrations of phenol, produce excess sludge and require large aeration basins.

Non-destructive techniques

Non-destructive techniques use an extraction method based on chemical or physical properties and thus make it possible to selectively separate one or more compounds from a mixture.

Physico-chemical methods have proven to be expensive and have inherent disadvantages due to the formation of toxic secondary materials such as chlorinated phenols [Barrios-martinez et al., 2006]. These techniques, such as extraction, membrane processes and adsorption, do not modify the nature of the species.

Liquid-liquid extraction

Liquid-liquid extraction is an effective technique that allows the separation and purification of several chemical compounds [Cote et al., 1998], it is based on the separation of compounds according to their relative solubility in two immiscible liquids, usually water and an organic solvent. This technique involves a transfer of material through a liquid phase which has the characteristics of an extraction solvent.

Membrane separation

The membrane separation method is a fluid separation process using as a separating agent a synthetic membrane which is a thin layer of material. The thickness of a membrane can varies from 100 nm to just over 1 cm. Separation takes place under the action of a driving force of transfer according to a defined separation mechanism. This method has the following advantages: High selectivity, Moderate operating cost, relatively short separation time. They are classified according to the type of membranes (solid, liquid or gaseous) and the nature of the driving forceCommonly used techniques to remove phenol derivatives from industrial wastewater are microfiltration, nanofiltration, reverse osmosis, and pervaporation [Khazaali et al., 2014; Mohammadi et al., 2015, Heng Loh et al., 2016].

Despite the many advantages already mentioned, membrane methods still suffer from certain drawbacks, namely Clogging, by particles or microorganisms and therefore the reduction of transmembrane fluxes and the need to implement washing operations, The limited lifespan membranes and a high energy cost resulting from the high pressures to be applied.

Adsorption

The use of adsorption technology for the removal of phenol from waste water is very easy and efficient from trace amounts to up to large quantities of phenol derivatives, depending on the recycling of the adsorbent. Many studies have been carried out to study the mechanisms of phenol adsorption by several types of adsorbents.

Mathematical models of adsorption

The study of mass transfer processes and heat is important to properly describe the adsorption mechanism.

Isotherm models

Freundlich and Langmuir models are best known for modeling adsorption mechanisms. Langmuir proposed in 1918 a simple model for studying the mechanism of adsorption [Langmuir et al. 1918].

$$\frac{1}{qe} = \frac{1}{q_0} + \frac{1}{b\,q_0}\frac{1}{c_e} \tag{1}$$

where: C_e – the concentration of adsorbate at equilibrium (mg/l-1);
qe – the amount of adsorption, at equilibrium (mg/g);
b and q_0 – Langmuir's constants.

In 1926, the Freundlich isotherm was introduced [Freundlich et al. 1926]

$$ln\, q_e = ln(k_F) + \frac{1}{n} ln\, Ce \tag{2}$$

where: n and k_F – Freundlich constants.

Kinetics of adsorption

To describe the adsorption kinetics, several kinetic adsorption models have been established namely pseudo-first and second-order models [Hui et al. 2009; Loredo-Cancino et al. 2014]. The form of the equation that describes the pseudo first model is:

$$\frac{dq_t}{dt} = k_1\,(q_e - q_t) \tag{3}$$

While the form of the equation that describes the pseudo second model is

$$\frac{dq_t}{dt} = k_2\,[q_e - q_t]^2 \tag{4}$$

On the other hand, to study the scattering mechanism in an adsorbent in the form of a spherical particle (radius r), another equation has been proposed [Burakova et al. 2018].

$$\frac{q_t}{q_e} = 6\sqrt{\frac{D_1\, t}{r^2 \Pi}} \tag{5}$$

Activated carbon

Activated carbon is a black powder consisting essentially of carbonaceous material with a microporous structure. Activated carbons are considered good adsorbents. It can be obtained from a large number of carbonyl materials (wood, coal, coconut, petroleum residues, etc.). Following carbonization processes followed by duly controlled activation processes. Activated carbon is part of a range of solids with very high porosity and high specific surface area varying from 500 to 1500 m^2/g, good pore size distribution and high mechanical strength [Perrich, 1981]. The mechanisms of adsorption of phenol derivatives by activated carbons have been studied by several authors, in particular the models of adsorption kinetics as well as the isotherms involved [Dabhade et al., 2009; Sunil et al., 2013; Sellaoui et al., 2019]. Other authors have studied the adsorption of phenol on granular activated carbon in the presence of the bacterium Pseudomonas aeruginosa. Analysis of the adsorption isotherm obtained shows a better fit is obtained with the Langmuir model and the kinetic study revealed that the adsorption of phenol to activated carbon well described by the pseudo-second-order model [Zeboudj et al., 2014]. The regeneration of activated carbon after adsorption of derivatives of phenol have also been studied by several authors [ozkaya, 2006, Aktaş and Çeçen, 2007]. Ozkaya compared several isotherms with the aim of studying the mechanism of desorption. The result found showed that the Langmuir model is the best to describe the phenol desorption reaction.

Clay

Clays are aluminosilicates. They consist of a basic mineral component (kaolinite, montmorillonite, etc.) and impurities such as quartz, ristobalite, calcite and organic matter [El gaidoumi, 2017]. The term clay designates not only a rock formation but it also defines a granulometric range comprising mineral particles, whose grain diameter is less than two micrometers. Several characteristics of clays, namely the size of surfaces with the presence of electric charges as well as their abundance in nature, justifies the great interest devoted by researchers to study the mechanisms of these adsorbents. The adsorption of phenol on quartz pyrophyllite from Tata (Morocco) was studied in a bach system in an aqueous medium [El gaidoumi et al., 2015]. The experimental results showed that the adsorption follows second order kinetics and the Langmuir model is more descriptive of the studied adsorption. The heat treatment of the adsorbent materials indicates that the studied clay is reusable for several adsorption tests. several physico-chemical parameters affect the adsorption power (pH, temperature, stirring rate and mass of the adsorbent).

Ion exchange resins

Synthetic resins are also widely used as adsorbent materials due to their ion exchange property. Ion exchange is a purification technique in which the ions present in a solution are adsorbed on a solid support (resin) and replaced by an identical quantity of an ion of the same electrical charge released by the solid. These resins do not have real porosity, and the ions to be exchanged must pass through the gel structure. The adsorption of phenols by the anion exchange resin (Amberlite IRA-67) was studied. Indeed, they studied the effect of many parameters on the adsorption power for a future use of this resin for industrial wastewater treatment. In particular, effects of the concentration of phenols in the feed stream and the recirculation time. [Victor-ortega et al., 2016].

Bio-adsorbents

Biomaterials have developed thanks to their low cost of preparation and the possibility of production from renewable raw materials (De et al., 2016]. The term bioadsorbent refers to a large number of products of biological or plant origin that trap organic or inorganic pollutants without prior transformation [Rocher, 2008]. Many studies have been carried out namely peanutshells [Villar da gama et al., 2018]. Girish et al carried out a comparative study between several bio adsorbents in order to show their effectiveness for the elimination of phenol and its derivatives [Girish and Ramachanda, 2012].

Nanomaterials

The development of nanomaterials (NM) capable of eliminating organic pollutants in water. They could be an effective solution in a water treatment sector to improve and develop the methods currently used. The use of non-toxic, low-cost and naturally abundant nanomaterials is part of an eco-design approach and is in accordance with the same directions of green chemistry [Ghannoum Obeid, 2014]. Magnetic nanoparticles have been developed by a new combustion technique, a method which makes it possible to obtain the magnetic nanopowder covered with organic residues generated by fuel combustion, which leads to the increase of its adsorption capacity [Mihoc et al., 2014]. Prepared NMs demonstrated greater adsorption power for p-chlorophenol than phenol, Kinetic studies indicated that the adsorption of phenol on the prepared NMs to well described by the pseudo-first order model and the Langmuir isotherm correlated with the results. The prepared NMs showed High adsorption power for p-chlorophenol and phenol as well as can easily remove using a magnetic field, which favors its potential application in the adsorption of phenolic compounds from industrial wastewater.

Apatites

Several studies have studied the power of apatites for the elimination of heavy metals [Bailliez, et al., 2004; Yaacoubi et al., 2014; Saoiabi et al., 2016], however the use of apatites for the elimination of phenolic compounds is very low

[El asri et al., 2009; Lin et al., 2009; Bouamrane et al., 2014]. The results of the adsorption reaction of phenol and its derivatives by natural phosphates showed that the adsorption capacity of these adsorbents depends on several parameters, namely, initial concentration of phenol, temperature and pH of the phenolic solution [Alzaydien and Manasreh 2009; Bouyamrane et al., 2010, Yaacoubi et al., 2015]. Bouyamrane et al studied the phenol retention capacity of crude natural Benguerir phosphate (NBP). The experimental results proved that the kinetics of phenol adsorption on PNB could be described by a pseudo first order model and for the adsorption isotherm, the Freundlich model showed a better fit [Bouyamrane et al., 2010]. The adsorption reaction of 2-nitrophenol on El Gantour phosphate (Morocco) was studied by Yaacoubi et al., the results found showed very rapid adsorption kinetics (2h) and a very high retention rate at values of PH equal 2 and 4. the result found also showed that the Dubinin _Radushkevich isotherm is the best for the description of the adsorption reaction and the intra diffusion model which controls the adsorption kinetics well [Yaacoubi et al., 2015]. Other authors have studied the effect of several parameters on the adsorption of a phenol solution on a phosphate rock activated by nitric acid. They found that adsorption power increases significantly with initial phenol concentration and decreases with temperature (exothermic reaction). They also found that increasing the adsorbent content leads to an increase in the amount of phenol adsorbed. Modeling showed that the reaction is irreversible and governed by a second-order equation [Alzaydien and Manasreh, 2009].

A study relating to the retention of phenol in a static regime on natural (NP) and modified (MP) phosphate was carried out by El Asri, the experimental results proved that:

- The kinetics of phenol adsorption on NP and MP could be considered as pseudo second order;
- The adsorption capacities obtained on the two materials NP and MP are respectively 3.75 mg/g and 6.46 mg/g;
- Lower adsorption capacity obtained at neutral pH values.The best modeling of the experimental results is obtained with the Langmuir model [El asri et al., 2009].

Bahdod et al conducted a comparative study between three synthetic apatites including their phenol adsorption capacity. The best performance was obtained for porous hydroxyapatite. The results found showed that the pseudo first order model and the intra diffusion model which control the adsorption kinetics well for the three apatites [Bahdod et al., 2009].

Alginate beads

Tests concerning the adsorption of p-nitrophenol by magnetic alginate beads having an organophilic and hydrophobic character due to the encapsulation of a bridged clay were carried out by Obeid et al., the results found showed that the adsorption increases with the amount of clay introduced into the beads. A slight decrease in adsorption is observed in a basic medium linked to electrostatic repulsions between the phenolate form of p-nitrophenol and the negatively charged beads. The adsorption of p-nitrophenol does not modify the concentration of calcium ions in the beads, thus indicating interactions of a hydrophobic nature between p-nitrophenol and the encapsulated clay. The equilibrium time is relatively long, however 50% of the p-nitrophenol is adsorbed in 10 minutes for an initial p-nitrophenol concentration of 100 mg/L [Ghannoum Obeid, 2014].

Table 1. Comparison of adsorption capacity of several adsorbents towards phenol derivatives

Adsorbent	Phenol initial Concentration (mg/L)	Adsorption Capacity (mg /g)	Reference
Natural phosphate (NP)	40	23	[Alzaydien and Manasreh 2009]
Hydroxyapatite (Hap)	100	8	[Bouyarmane et al. 2010]
Sedimentary phosphate (SP)	100	40	[Yaacoubi et al. 2015]
Pecan shells (AC)	35	18	[Shawabkeh et al. 2007]
Coffee residue (AC)	170	84.02	[Khenniche et al. 2010]
Black stone cherries (AC)	500	133.33	[Rodriguez Arana et al. 2010]
Natural phosphate (NP)	100	12	[Benaddi et al. 2020]
Hydroxyapatite-calcium alginate composite (HA/CA)	100	244	[Benaddi et al. 2021]

CONCLUSION

Several techniques have been studied for the removal of phenolic compounds from industrial wastewater. Adsorption remains the best technique due to its characteristics, in fact, it is non-destructive and simple to use as well as have more other advantages, such as practicality and efficiency and low cost, Therefore, these methods need to be widely developed on an industrial scale to remove phenol derivatives and achieve wastewater quality in accordance with standards. On the other hand, the development of these adsorption methods is highly dependent on new research on materials from abundant natural resources, namely apatites or biomaterials.

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