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Review

# Towards sustainable physiochemical and biological techniques for the remediation of phenol from wastewater: A review on current applications and removal mechanisms

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ABSTRACT

Phenol is a priority pollutant that presents a significant threat to human health and natural systems when discharged directly into the environment. Consequently, numerous technologies have been used and developed to eliminate phenol from wastewater streams. These technologies can be categorized into physical, chemical, and biological methods. While conventional treatment methods are highly efficient in phenol removal; some of these techniques are not environmentally friendly and others are expensive. Therefore, sustainable, and green technologies are being employed and taken into consideration in the treatment of phenol wastewater due to their effectiveness, affordability, and environmental compatibility. This review aims to highlight efficient green physiochemical and biological methods of water treatment and demonstrate the mechanisms of phenol removal in these technologies. Particular emphasis will be given to the use of low-cost adsorbents prepared from industrial and agricultural wastes for the efficient removal of phenol from wastewater as adsorption processes show the highest cost-effectiveness among all the treatment technologies.

## 1. Introduction: phenolic compounds, reactivity, toxicity, and fate

Water pollution is an increasingly pressing issue on a global scale, primarily caused by factors such as rising water demand, population growth, industrialization, urbanization, and agriculture activities. This has resulted in the degradation and pollution of the environment, adversely affecting water bodies, and ultimately affecting human health and the environment. Several organic pollutants are responsible for the reduction of water quality including phenolic compounds (Soto-hernandez et al., 2017). Phenol is an aromatic organic compound with the molecular formula $C_6H_5OH$, it comprises of an aromatic ring to which a hydroxyl group is attached. These compounds are either formed naturally by the action of different organisms or are produced by numerous industries and released to the environment as wastewater without appropriate treatment (Almasi et al., 2021; Soto-hernandez et al., 2017). Fig. 1-A shows the sources of the various phenolic compounds found in the environment. The introduced phenols are recalcitrant contaminants that are resistant to degradation through physical, chemical, and biological processes (Mohamed et al., 2020). Consequently, the US EPA and the Canadian national pollutant release inventory (NPRI) categorized phenol as one of the 129 specific priority pollutants that must be remediated before discharge (US EPA, 2014). It is estimated that over 10 million tons of phenolic compounds are discharged into the environment (Alshabib and Onaizi, 2019) by the petrochemical, pharmaceutical, leather, textile, and agrochemical industries. In addition, industrial processes such as paint, paper, pulp, and pesticide production are also believed to be responsible for phenolic compounds discharge into the environment (Alshabib and Onaizi, 2019; Deng et al., 2011). Fig. 1-B shows the percent of phenolic compounds in a variety of industrial effluents. The concentrations of phenolic compounds in industrial effluents range from 1 mg/L and could reach up to 7000 mg/L (Mohd, 2020).

It is vital to handle phenolic compounds properly since these compounds can adversely affect human health and biotic systems (Saravanan et al., 2021). In terms of their impact on human health, phenolic compounds are considered toxic, carcinogenic, and mutagenic. In addition to that, phenols can lead to various health complications and disorders including genotoxicity, muscle fatigue, dysfunction of the liver and kidneys, metabolic and eating disorders, weight loss, diarrhea, bronchoconstriction, irregular breathing, irritation of the ducts, coma,

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**Abbreviations**

| | |
|---|---|
| AC | Activated carbon |
| AGS | Aerobic granular sludge |
| AOPs | Advanced oxidation processes |
| CNTs | Carbon nanotubes |
| CWAO | Catalytic wet air oxidation |
| DESs | Deep eutectic solvents |
| DNAPLs | Dense Nonaqueous Phase Liquid |
| EPA | Environmental protection agency |
| GO | Graphene oxide |
| HRP | Horseradish peroxidase |
| ILs | Ionic liquids |
| IMO | International maritime organization |
| LAC | lignite-activated coke |
| $LC_{50}$ | Lethal concentration 50 |
| LLE | Liquid-liquid extraction |
| LNAPLs | Light Nonaqueous Phase Liquid |
| MBBR | Moving bed biofilm reactors |
| MNBs | Micro-nano-bubbles |
| NAPLs | Nonaqueous phase liquids |
| NF | Nanofiltration |
| NPRI | National pollutant release inventory |
| RGO | Reduced graphene oxide |
| RO | Reverse osmosis |
| SLM | Supported liquid membrane |
| TOC | Total organic carbon |

and central nervous system (Anku et al., 2017). The toxicity levels of phenolic compounds range between 10 and 24 mg/L, and the lethal blood concentration is ~1.5 mg/mL. Phenolic compounds can adversely affect the environment by polluting soil and water bodies including surface and groundwater (Alshabib and Onaizi, 2019; Anku et al., 2017; Mohamad Said et al., 2021). Consecutively, these pollutants induce changes in plant communities' structure and bioaccumulate in birds and fish, ultimately incorporating into food chains and adversely affecting health (Garg et al., 2020; Mandeep et al., 2020). Fig. 1-C shows the physicochemical characteristics of phenol and their maximum permissible levels in different water bodies as recommended by the US EPA (Alshabib and Onaizi, 2019). From these strict limits, it could be deduced that it is necessary to sustain low concentrations of phenolic compounds in water bodies to protect human and environmental health (Raza et al., 2019).

Many derivatives of phenol are usually found in various water bodies

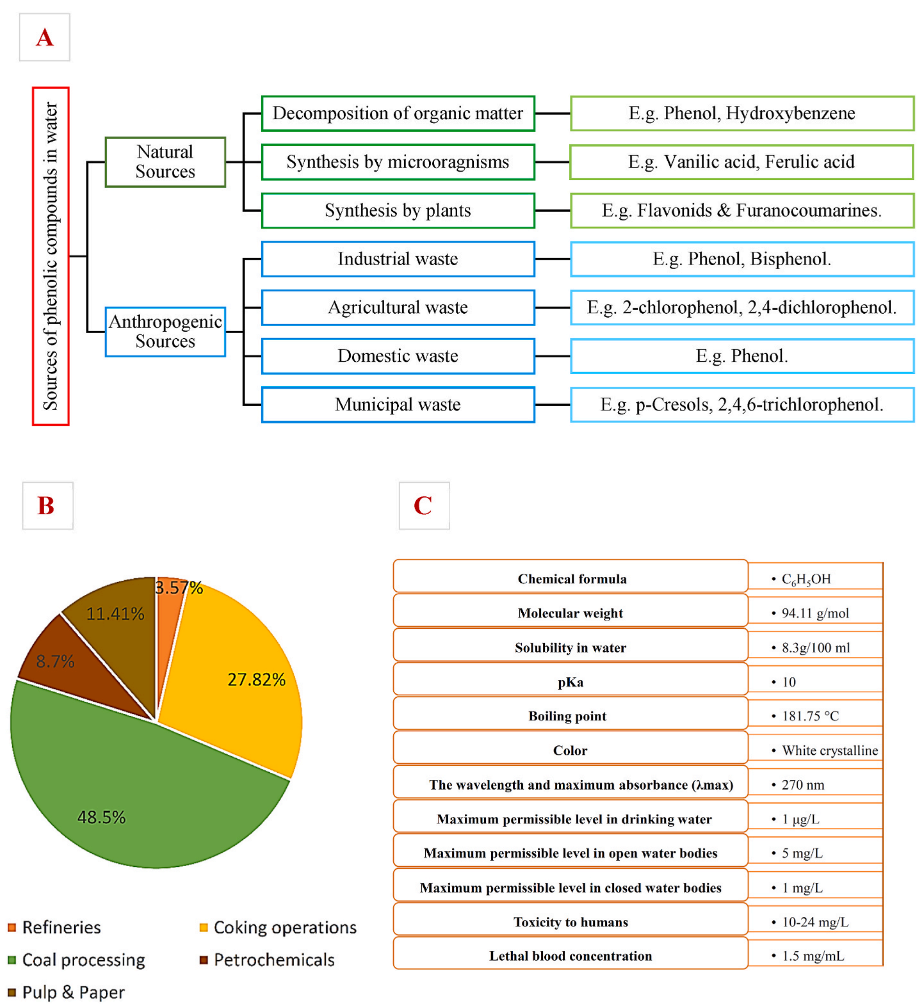


**Fig. 1.** **A**-Sources of phenolic compounds in water. **B**- Phenol concentration (%) in effluents of various industries (Raza et al., 2019). **C**- Physiochemical characteristics of phenol and maximum permissible levels of phenol in water.

including groundwater. These compounds come from nearby waste dumping sites or spill sites. Phenol has a high solubility in water (83 g/L), and has a short half-life in soils, thus it reaches groundwater easily. Phenolic compounds enter water bodies and the aquatic environment through direct discharge or runoff (Anku et al., 2017). When these compounds reach water bodies, they transform into various intermediates. The transformation of phenolic compounds in the aquatic environment is driven by various physical, chemical, and biological interactions. In terms of physical interactions, phenol can interact with nitric ions in the presence of UV radiation, leading to the formation of intermediates such as 4-nitrophenol. Additionally, phenol photolysis can result in the production of hydroquinone in the presence of charge transfer complexes (Kinney and Ivanuski, 1969). On the other hand, chemical interactions can also lead to the formation of intermediates, for example, phenol can interact with hydroxyl radicals or nitric ions, which leads to the formation of 2-nitrophenol (Moussavi, 1979). Additionally, metal cations interact with various phenolic compounds, which leads to their ionization and in turn enhances their solubility in water (Epa, 1979). Aerobic and anaerobic biological interactions also lead to alterations in the structure of phenolic compounds, for instance, microorganisms produce 4-chlorophenol upon encountering 4-chlorophenoxyacetic acid, and tetrachlorocatechol is generated as a by-product of pentachlorophenol degradation. Additionally, chlorobenzenes could also be degraded microbially, leading to the formation of chlorocatechol (Anku et al., 2017).

A significant portion of organic contaminants in nature exist as non-aqueous phase liquids (NAPLs). NAPLs are organic contaminants that are immiscible in water. NAPLs are grouped into two main categories based on their density: Light Nonaqueous Phase Liquid (LNAPLs) and Dense Nonaqueous Phase Liquid (DNAPLs). Due to their inherent properties, these compounds persist in the pores of sediments as a separate phase. Their fate in the marine environment is highly dependent on their density. In wet porous media, mass transfer of NAPLs requires the volatilization of these compounds into a gaseous phase, following that, the gaseous NAPLs will dissolute into the aqueous phase, and finally, the aqueous phase will sorb into the solid phase. In marine sediments where water coats the soil particles, NAPL mass transfer into the soil is not possible, and NAPLs need to be partitioned first to the aqueous phase, before sorbing into the sediments (Fitts, 2013; Lenhard et al., 2005). Phenolic compounds are recalcitrant pollutants; therefore, the accumulation of these contaminants and their intermediates in water bodies and marine sediments is expected. Progressively, ocean floor organisms such as bottom feeders that feed and forage near the ocean bed will start to accumulate these compounds, and thus these pollutants enter food webs and chains (Zhou et al., 2017). For example, chlorophenols distribution patterns in marine sediments indicate that these compounds could be accumulated in marine water by adsorbing the suspended organic compounds due to their lipophilic nature (Xie et al., 1986). Several freshwater and marine organisms showed high sensitivity to phenol. For example, the highest sensitivity noted from marine organisms towards phenol was with *Archaeomysis kokuboi* where the $LC_{50}$ value was found to be 0.26 mg/L after 96 h (Noszczyńska and Piotrowska-Seget, 2018). On the other hand, the freshwater organism *Cirrhinus Mmririgala* recorded the maximum sensitivity with an $LC_{50}$ of 1.55 mg/L after 96 h (Duan et al., 2018). Due to the highest sensitivity, the international maritime organization (IMO) has listed phenolic compounds as the top 20 hazardous compounds that present high risks to aquatic life (Panigrahy et al., 2022).

## 2. Phenol remediation technologies

The presence of phenol in industrial wastewater streams necessitates its treatment to prevent adverse impacts on human health and the environment. Moreover, effective treatment of these streams can potentially yield a sustainable and renewable water source. A range of physiochemical technologies, including distillation, nanofiltration, reverse osmosis, chemical oxidation, electrochemical oxidation, solvents extraction, ozonation, advanced oxidation processes, photocatalytic oxidation, adsorption, and biodegradation, have been employed for industrial wastewater treatment (Garg et al., 2020; Saravanan et al., 2021a, 2012b; Villegas et al., 2016). Table 1 summarizes the various technologies used for wastewater treatment and their advantages and disadvantages. Though these techniques have many advantages, they also have a few disadvantages mainly the high operational and maintenance costs, high energy requirements, pretreatment requirements, low removal efficiencies, scaling, fouling, limitation due to selectivity, and pH level constraints, and use of hazardous organic solvents (Gholami-Bonabi et al., 2020; Jiménez et al., 2018). Additionally, the selection of the treatment process significantly relies on pollutant concentration, volume, and cost of the treatment process (Guha Thakurta et al., 2018). Thus, in recent years, a shift towards the use of environmentally compatible technologies in wastewater treatment was noticed to reduce the operation costs and their impact on the environment (Adeniyi and Ighalo, 2019).

This review aims to provide a comprehensive overview of the conventional methods used for phenol wastewater treatment, explain the mechanism involved in phenol removal, and emphasize green technologies that support sustainable practices. Additionally, this review provides inclusive insights into the various available options and serves as a guide for selecting the most suitable technologies for treating phenol wastewater considering factors such as initial pollutant concentrations, costs, wastewater volumes, and environmental compatibility. Finally, this review highlights the importance of adsorption technologies as a cost-effective, sustainable, and environmentally friendly solution, which could be adopted for the treatment of phenol-contaminated wastewater.

### 2.1. Physical treatment methods

#### 2.1.1. Distillation

Many physical processes are used in wastewater treatment. These processes do not require the use of chemicals. Steam distillation is one of the physical recovery processes in which the steam volatile compounds are volatilized, condensed, and collected in receivers. Steam distillation separates immiscible liquids based on the volatility of the steam. In the liquid phase, phenol has limited immiscibility with water, however, the immiscibility disappears at a temperature of approximately 68 °C (Saputera et al., 2021). Phenol can be distilled from polluted water by the process, which begins when wastewater enters the distillation column, and when the temperature reaches around the boiling point of water, and the pressure of 1 atm, the water evaporates and condenses while the phenol is collected at the bottom of the column. This method of treatment is suitable for highly contaminated wastewater owing to the high operating costs and energy requirements (Gao et al., 2021).

A few studies investigated distillation processes in phenol removal as noted in Table 2. Mohammadi & Kazemi investigated the use of a vacuum membrane distillation process for phenol wastewater treatment. It was found that the optimum condition for phenol separation was a temperature of 45 °C, a pH of 13, a concentration of 1000 mg/L, and a pressure of 60 bar (Mohammadi and Kazemi, 2014). Additionally, the distillation process could be combined with the extraction process to recover phenolic compounds. These technologies use extraction solvents to collect phenol out of the water as noted in Table 2, leading to improved recovery rates. For example, in a study, phenol was recovered using a steam distillation—extraction process using diethyl ether as the extraction solvent. High recovery rates of about ~100% were achieved at an initial phenol concentration of 30 mg/L and highly saline and acidic conditions and similar trends were noted in other studies (Saputera et al., 2021).

It could be noted from these studies that distillation processes are effective in separating various pollutants from wastewater and are particularly beneficial for pollutants with concentrations of ~1000 mg/L. However, their application in wastewater treatment is not widely

**Table 1**
Advantages and disadvantages of phenolic compounds remediation technologies.

| | Technology | Mechanism | Advantages | Disadvantages |
|---|---|---|---|---|
| Physical methods | **Distillation** | Distillation is a technique for separating the components from a miscible fluid mixture by selective evaporation and condensation according to boiling points. | High recovery rates with high purity and ability to reuse, fast separation, feasible for highly concentrated solutions. | High operational costs, energy-intensive, not appropriate for low concentration solutions, and it might have impurities. |
| | **Membranes** | Membrane technologies separate contaminants from water based on properties such as size or charge. | Operates under various conditions and concentrations, is selective, and allows the use of hybrid systems. | High capital and operational costs, scaling, and fouling of membranes. |
| | **Nanofiltration** | A pressure-driven membrane process that removes solutes with molecular weight in the range of 200–1000 g/mol, typically from aqueous mediums. | Effective in removing pollutants, simple to operate, and maintain. | Cannot treat higher loads of pollutants, membrane replacement is needed with time. |
| | **Reverse osmosis** | RO membranes allow water to pass through while rejecting solutes, such as low molecular weight organic materials. | Highly effective at removing contaminants and is energy efficient. | Cannot treat higher loads of pollutants, high capital and operational costs, and fouling. |
| Chemical methods | **Chemical oxidation** | Chemical oxidation requires the use of an oxidizing agent in the treatment of wastewater to oxidize the organic pollutants. | Oxidation of high and low concentrations of pollutants could also be paired with other technologies such as UV to improve removal efficiency. | The use of large quantities of hazardous oxidizing chemicals, and partial oxidation leads to the formation of by-products. |
| | **Electrochemical oxidation** | Electrochemical oxidation uses electric current or a potential difference between two electrodes (anode and cathode), with which hydroxyl radicals or oxidizing species can be generated and used for the oxidation of pollutants. | Use fewer chemicals and harsh materials, easy to operate and maintain. | This technology is dependent on electrical energy, and it requires electrode replacement frequently which has higher operational costs. |
| | **Ozonation** | Ozonation produces hydroxyl (OH) radicals through the decomposition of ozone ($O_3$) that are used in the oxidation of organic pollutants. | Eliminate and reduce bioactivity, toxicity, and biological effects of pollutants. | Expensive and not very effective for COD and TOC reduction leads to the formation of disinfection byproducts. |
| | **Photocatalysis** | Photocatalysis is a process in which light energy is used to generate radicals that are used for pollutant degradation | Oxidation could be done using solar irradiation which is abundant and benign. Occurs at ambient temperature and pressure conditions with cheap, nontoxic, and noncorrosive catalysts. | Most semiconductor materials are not visible light active, require high band gap energy and agglomeration of nanoparticles can occur making it difficult to separate or reuse catalyst from aqueous solution. |
| | **Extraction with solvents** | Extraction is a method that separates compounds based on their relative solubilities in two different immiscible liquids. | High concentrations of pollutants can be extracted and thus collected as a product, simple to operate. | Extraction solvents are volatile, flammable, toxic, and expensive, and require the use of large solvent volumes, multiple extraction steps, and a long extraction period. |
| | **Adsorption** | Adsorption is a process where pollutants are removed due to electrostatic interactions, π-π interactions, hydrophobic interactions, van der Wal forces, and hydrogen bonding between the adsorbent and adsorbate. | Cost-effective especially when waste materials are used. Can be used for different concentrations depending on the adsorption capacity of the adsorbent. | Less effective at high concentrations, and difficult to separate adsorbent from adsorbate. Spent adsorbents should be recycled or treated before discarding into the environment. |
| Biological methods | **Phytoremediation** | Phytoremediation includes phytoextraction and accumulation, and phytodegradation by certain resistant plants. | Eco-friendly, cost-effective, and can promote biodiversity. | Cannot operate at extremely high concentrations due to toxicity and the requirement of large spaces. |
| | **Enzymes** | Enzymes oxidize pollutants into simpler organic compounds. | Operates on low and medium concentrations, with no toxicity issues. The degradation of pollutants is rapid and selective. | Enzyme purification is expensive as the process is dependent on reaction conditions such as temperature and pH as well as possibilities of enzyme deactivation. |
| | **Biodegradation** | Biodegradation requires the use of microorganisms that consume pollutants as carbon sources leading to the decomposition of pollutants. | Eco-friendly, cost-effective, and works with low and high-strength wastewater. | Sometimes, it cannot operate at extremely high concentrations (toxicity) and requires larger space, chemical treatment, and aeration. |

reported due to the high capital and operational costs associated with them. Moreover, the use of these technologies results in the production of concentrated wastes that require further treatment or disposal. Therefore, the use of distillation processes is often not practical with larger volumes of wastewater containing low levels of pollutants.

### 2.1.2. Membrane separation

In this process, a membrane is used to separate the components in a solution by rejecting unwanted substances and allowing the others to pass. These technologies can separate various pollutants such as salts, dyes, and organic pollutants including phenol (Cevallos-Mendoza et al., 2022). Membrane performance is highly dependent on membrane properties, pollutant properties, and operating conditions as noted in Fig. 2-A. This figure also shows the different types of membranes used in water treatment and their applications. Membrane-based technologies have advantages such as the absence of by-product generation, easy installation, and low energy consumption. However, membrane-based technologies also have a few barriers including stability and reusability as these membranes have short lifetimes due to fouling (Obotey Ezugbe and Rathilal, 2020).

Reverse osmosis (RO), nanofiltration (NF), and ultrafiltration are all different levels of membrane-based technologies that are being used for phenolic wastewater treatment as reported in Table 2. Among these, nanofiltration is most used for the separation of phenol. In a study, several nano-filtration membranes were tested for the removal of phenol with an initial concentration of 1000 mg/L, it was noted that the highest removal rate was achieved using DSS-HR98PP polymeric membrane with 80% rejection at neutral pH levels (Bódalo et al., 2009). Nanofiltration could also be coupled with adsorption to improve the separation of phenol, especially with membranes that have larger pore sizes where phenol can pass on easily. For example, a study showed that phenol removal using adsorption/nanofiltration was around 31% in the presence of nanoparticles and only achieved 4% removal in the absence of adsorption i.e., around 675% improvement compared to using

**Table 2**
Examples of green physical, chemical, and biological technologies for phenol remediation.

| Type of remediation technique | Example | Pollutant | Initial Pollutant concentration | Removal effectiveness | Remediation mechanism(s) | Conditions | Reference |
|---|---|---|---|---|---|---|---|
| **Physical technologies** | | | | | | | |
| Filtration | Double filtration with AC | Phenol | 0.3–0.1 mg/L | 99% retention | AC porosity, surface area, and chemistry play important role in the adsorption of phenol. | – | Fuentes et al. (2018) |
| RO-membrane | Polyamide thin film composite RO | Phenol | 1000 mg/L | ~70% retention | The electrostatic repulsion between phenol and membrane | pH 6.5 | Mnif et al. (2015) |
| **Chemical technologies** | | | | | | | |
| Extraction | DESs-Choline chloride (ChCl)-glycerol | Phenol | – | 98.3% | H-bonds between the DES and phenolic compounds | T 30 °C | Yi et al. (2019) |
| Ozonation | Ozone reactor | Phenol | 2000 | ~35% | Oxidation by ozone and hydroxyl radicals | pH 9 T 25 °C | Haag and Hoigne (1983) |
| Electrochemical oxidation | Sn-doped Ti/$PbO_2$ | Phenol | 500 | 89% | The electric current generates hydroxyl radicals and other oxidizing species to degrade phenol | pH 5.5 T 30 °C | Li et al. (2013) |
| Electrochemical oxidation | Ti/SnO2–Sb2O3–Nb2O5/PbO2 | Phenol | 500 | 78% | The electric current generates hydroxyl radicals and other oxidizing species to degrade phenol | pH 7 T 20 °C | Yang et al. (2008). |
| Extraction | Terpenoids and hydrophobic eutectic solvents | Phenol | 500 mg/L | >95% separation | The presence of acceptor H-bond regions in the solvent would increase phenols affinity. | T40 °C | Rodríguez-Llorente et al. (2020) |
| Adsorption | AC | Phenol | 100 mg/L | 434 mg/g | Π-π interaction, electron-donor–acceptor complex formation, and H-bonding | pH 6, T 25 °C | Mojoudi et al. (2019) |
| Extraction by Ionic liquid | 1-ethyl-3-methyl imidazolium cyanoborohydride, | Phenol | 100 mg/L | Selectively extracted 95% of the phenol | Intermolecular interaction between [$BH_3CN$] anion and phenol molecules. | T 30–80 °C | Mathews et al. (2019) |
| Ionic liquid | 4-butyl-1-methyl pyridinium bis (trifluoromethyl sulfonyl) imide | Phenol | 15,000 mg/L | 96% extraction of phenol | Hydrophobic interactions between ILs and Phenol | pH 6, 25 °C | Sas et al. (2020) |
| AOPs Photocatalysis | Polymer, CNT, $TiO_2$–$NH_2$, and UV | Phenol | 10 mg/L | 99% photodegradation in 7 min | An increase in electron-hole pairs on the catalyst surface leads to higher concentrations of reactive hydroxyl radicals, which lead to phenol degradation. | pH 5 | Mohamed et al. (2020) |
| AOPs Photocatalysis | Acid-modified $TiO_2$ nanoparticles | Phenol | 55 mg/L | 99% photodegradation in 23 h | Phenol can be hydroxylated by OH radicals and the formation of Lewis acid $Ti^{3+}$ sites on the $TiO_2$ surface via hydrogenation leading to higher phenol degradation | T 20 °C | Ling et al. (2015) |
| AOPs | $O_3$-calcium peroxide | Phenol | 5 mg/L | 97% | Calcium peroxide produces hydrogen peroxide that degrades phenol | pH 3 T 25 °C | Honarmandrad et al. (2021) |
| AOPs - Fenton | Fe (II)/$H_2O_2$ | Phenol | 100 mg/L | 100% degradation in 9 min | Phenol oxidation is carried out by hydroxyl radicals generated from a reaction between hydrogen peroxide and iron (II) salts | pH~3, T25 °C | Esplugas et al. (2002) |
| **Biological methods** | | | | | | | |
| Phytoremediation | *Hydrilla verticillata* | Phenol | 100 mg/L | 99% removal in 7 days | Transformation and detoxification by peroxidases and ROS | 12-h photoperiod at 21/16 °C | Chang et al. (2020) |
| Enzymatic | Laccase | Phenol | 376.44 mg/L | 96% removal in 30 min | Catalytic oxidation of phenol | pH 5, T50 °C | Asadgol et al. (2014) |
| Enzymatic | Peroxidase | Phenol | 100 mg/L | 97.4% | Catalytic oxidation of phenol | pH (4.0–9.0), T (20–60 °C). | González et al. (2006) |
| Biodegradation | *Pseudomonas fredriksbergsis* | Phenol | 700 mg/L | 90% removal in 96 h | Hydroxylase and oxygenase enzymes are used to biodegrade phenol and use it as a carbon source | pH 7, T28 °C | Aljbour et al. (2021) |
| Biodegradation | *Acinetobacter lwoffii* | Phenol | 500 mg/L | 100% removal in 12 h | Enzymes such as Hydroxylase and catechol 1,2-dioxygenase break down phenol via the ortho-cleavage pathway. | pH8, 33 °C | Xu et al. (2021) |

(*continued on next page*)

**Table 2** (*continued*)

| Type of remediation technique | Example | Pollutant | Initial Pollutant concentration | Removal effectiveness | Remediation mechanism(s) | Conditions | Reference |
|---|---|---|---|---|---|---|---|
| **Hybrid technologies** | | | | | | | |
| Adsorption + nanofiltration | Silver nanoparticles-Nanofiltration | Phenol | 180 mg/L | ~100% | First adsorption of phenol by nanoparticles (increase in the particle size) followed by filtration with an NF membrane, | pH7, - | Naidu et al. (2016) |
| Adsorption + UV | GO-UV | Phenol | 100 mg/L | 95.95% | hydrogen bonding, π-π interactions, electrostatic interaction, $H_2O_2$ oxidation. | pH6, T 35 °C | Al-Ghouti et al. (2022) |
| Distillation + extraction | Steam distillation with diethyl ether | Phenol | – | 91.8% | Water evaporates leaving phenol to be extracted | -, T 50 °C | Barták et al. (2000) |
| Adsorption + photocatalysis | $AC/TiO_2/CeO_2$ | Phenol | 763 mg/L | 50.91% | Photocatalysis and adsorption by the negatively charged surface. | pH 8, T30 °C | Dalanta and Kusworo (2022) |
| Adsorption + UV | AC-UV | Phenol | – | 99% | AC accelerated the degradation of organic compounds by catalyzing $O_3$ to generate hydroxyl radicals (·OH). | pH7, T25 °C | Xiong et al. (2020) |

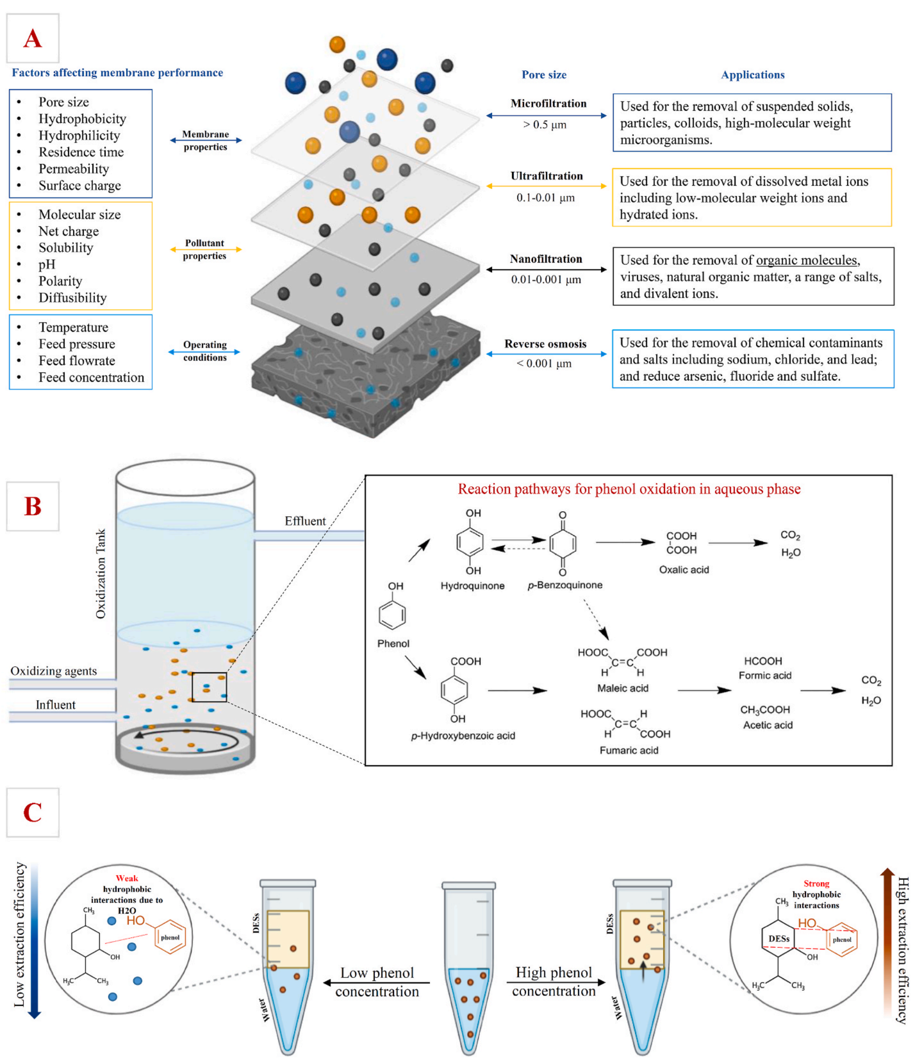


**Fig. 2.** **A-** Overview of filtration technology and its role in the removal of pollutants. **B-** Overview of Phenol chemical oxidation process, and **C-** Extraction of phenol using DESs.

nanofiltration alone (Naidu et al., 2016).

Reverse osmosis is another emerging membrane-based technology that removes organic impurities including phenolic compounds from water by using pressure to force water molecules through a semi-permeable membrane (Srinivasan et al., 2011). Since organic matter can cause clogging in the reverse osmosis membranes, nanofiltration technologies are used in advance. The coupling of reverse osmosis and nanofiltration is essential for the stabilization of pressure fluctuations that occur when using reverse osmosis systems alone, leading to higher treatment efficiencies. Several studies investigated the use of RO in phenolic compound removal (Srinivasan et al., 2011). In a study, phenol-contaminated wastewater with an initial concentration of 10 mg/L was subjected to RO. It was found that with RO alone, the phenol concentrations in the effluent reached around 1.30 mg/L at pH 4, with a removal efficiency of 87%. To further improve the treatment process, the wastewater stream was subjected to pretreatment using granular activated carbon. It was noted that the overall efficiency of the RO system increased to 99% (Ipek, 2004). Li et al. conducted a study to compare the effectiveness of nanofiltration and reverse osmosis membranes in removing phenol from synthetic wastewater. Different types of nanofiltration membranes (NF-90, NF-97, and NF-99) and reverse osmosis membranes (RO-98pHt and RO-99) were tested on wastewater containing phenol with levels below 1000 mg/L. The researchers found that both nanofiltration and reverse osmosis membranes had advantages and disadvantages. Nanofiltration showed a low rejection rate of phenol (between 0.41 and 0.72) but had a high maximum flux rate (up to 180 $L/m^2/hr$), while reverse osmosis had a high rejection rate (0.81) but a low minimum flux rate (only 60 $L/m^2/hr$). Generally, the study showed that NF and RO could be considered effective methods for removing phenol from wastewater (Li et al., 2010).

It could be implied from these studies that membrane technologies could be used in the treatment of phenolic wastewater with concentrations of approximately 1000 mg/L with retention levels of ~95–100%. However, similarly to distillation technologies, membrane technologies have high operational costs due to fouling and maintenance and high rates of energy consumption, therefore their application could be hindered.

### 2.2. Chemical treatment methods

#### 2.2.1. Chemical oxidation

Phenol-contaminated wastewater could also be treated chemically. Chemical oxidation is a destructive method that involves the use of oxidizing agents in the treatment process to oxidize organic pollutants. Many chemicals are used for the oxidation of organic pollutants including hydrogen peroxide, chlorine, chlorine dioxide, permanganate, and ferrate, these oxidants degrade organic compounds such as phenol into simpler compounds or water and carbon dioxide as illustrated in Fig. 2-B. These oxidants generate radicals that rapidly and non-selectively react with organic compounds leading to their degradation (Pal, 2017). Chemical oxidation processes are very advantageous owing to their effectiveness, ability to operate under various conditions of pH and temperature, and low operating costs, however, these processes form recalcitrant pollutants during the process of oxidation, leading to the generation of by-products, mostly when the oxidation process is incomplete (Peings et al., 2015).

A comparative study investigated sulfatoferrate, potassium permanganate, and calcium hypochlorite's ability to oxidize phenol. In this study, the initial phenol concentration was 30 mg/L, and the experiment was conducted at pH 9. After an hour, it was found that sulfatoferrate was able to degrade 57% of phenol, potassium permanganate degraded 70% of phenol, and calcium hypochlorite had a removal efficiency of 61%. It is worth mentioning that the use of permanganate could increase manganese concentration in water whereas using hypochlorite could lead to the formation of several chlorinated by-products. The authors stated that using sulfatoferrate is safer due to the lack of by-product generation (Peings et al., 2015). Matta et al. reported that synthetic chloride green rust-$H_2O_2$ was able to degrade 100% of phenol with an initial phenol concentration of 50 mg/L in 1 min by hydroxylation/oxidation. Additionally, these compounds degraded 62% of total organic carbon (TOC) in 24 h at neutral pH of 7 (Matta et al., 2008). Chamberlin et al. used potassium permanganate to oxidize phenol with an initial concentration of 125 mg/L at a high temperature of 95 °C. This oxidation process achieved a maximum removal of 62%. Due to this partial removal, another experiment was conduction for the degradation of phenol using hypochlorite while maintaining alkaline conditions to sustain the oxidation process. It was noted that the maximum removal was achieved by adding 5000 mg/L of hypochlorite to a solution containing 100 mg/L of phenol (Chamberlin et al., 1952). It could be determined from the studies that it is vital to ensure that complete oxidation is achieved especially in chlorination at lower pH levels since partial chlorination could lead to the formation of chlorophenols, which are more recalcitrant than phenol itself.

The Fenton oxidation process is another type of chemical oxidation process in which the ferrous or ferric cation decomposes hydrogen peroxide to generate strong oxidizing agents capable of degrading organic and inorganic substances including phenolic compounds. Yavuz et al. investigated the ability of the Fenton process to degrade phenol. Around 98% removal was achieved at an initial phenol concentration of 500 mg/L, hydrogen peroxide ($H_2O_2$) concentration of 3000 mg/L, ferrous sulfate ($FeSO_4$) concentration of 1500 mg/L, and pH of 2.3. The authors stated that under acidic conditions, the reaction between $H_2O_2$ and $Fe^{2+}$ generated hydroxyl radicals that oxidized phenol molecules. Although phenol removal was sufficient, the COD removal efficiency was limited (Yavuz et al., 2007).

Catalytic wet air oxidation (CWAO) is a process that involves oxidizing a liquid waste stream with air or oxygen at elevated temperatures and pressures in the presence of a catalyst. One potential application of CWAO is in conjunction with trickle bed reactors (TBRs), which are often used for the treatment of wastewater. The combination of CWAO and TBRs can enhance the efficiency of wastewater treatment, particularly when dealing with complex organic compounds and other contaminants (Candan and Ayten, 2021). A trickle-bed reactor is a type of fixed-bed reactor that can be used to remove organic compounds from wastewater (Makatsa et al., 2019). The reactor consists of a vessel filled with a bed of catalyst particles over which the wastewater is passed. The reactor operates in a continuous mode and the wastewater flows downward over the catalyst bed while air or oxygen is introduced to facilitate the oxidation of phenol and its derivatives. The effectiveness of a trickle bed reactor for removing phenol and its derivatives depends on several factors, including the catalyst type, catalyst loading, hydraulic retention time, and initial concentration of the wastewater (Al-Huwaidi et al., 2021). They offer several advantages including high contact efficiency between the wastewater and catalyst, low catalyst cost, and low energy requirements. Additionally, A trickle-bed reactor (TBR) can be easily scaled up to meet the requirements of different wastewater treatment applications (Mohammed et al., 2016). In a study, phenol was oxidized in a TBR over Al–Zr- a pillared clay catalyst under various experimental conditions. It was found that a complete conversion of phenol with an initial concentration of 1000 mg/L was achieved in 180 min, with a temperature of 160, a gas velocity of 0.012 m/s, and a pressure of 10 bar (Makatsa et al., 2019). Other studies also showed that combining TBR technologies with other processes such as reverse osmosis can significantly improve the treatment process (Al-Obaidi et al., 2018).

Chemical oxidation processes offer a solution for treating wastewater contaminated with initial concentrations of up to 500 mg/L. These processes have been shown to achieve oxidation efficiencies ranging from approximately 55%–100%. However, several challenges hinder their application including the need for large quantities of oxidants and the generation of by-products. In addition, these methods have safety and environmental concerns due to the use of strong chemical agents.

### 2.2.2. Electrochemical oxidation

Electrochemical oxidation is another method of wastewater treatment that does not require the use of chemical reagents in the oxidation process. Direct electrochemical oxidation oxidizes pollutants by their adsorption to the anode surface by charge transfer reaction. On the other hand, indirect oxidation uses an intermediate redox reagent present in the solution that prevents the electron transfer between the electrode and the pollutant preventing electrodes from fouling (Martín-Pozo et al., 2022).

Many studies have investigated the use of electrochemical oxidation of phenol as noted in Table 2. Saratale Rijuta et al. investigated phenol oxidation using a Ti/$PbO_2$ electrode with the addition of $Fe^{2+}$. Complete removal of phenol with an initial concentration of 250 mg/L was observed at 50 °C, pH 2, and a potential difference of 5 V (Saratale Rijuta et al., 2016). Abou-Talab et al. recently investigated phenol removal from petroleum wastewater using graphite electrodes as an anode and stainless-steel electrodes as a cathode. Complete phenol removal from an initial concentration of about 6.8 mg/L was achieved within 15 min and under a current density of 3 mA/$cm^2$ (Abou-Taleb et al., 2021).

It is clear that electrochemical oxidation processes are a viable option for treating phenol-contaminated wastewater with initial concentrations of up to 500 mg/L. Such processes can achieve removal efficiencies ranging from 78% to 100%. However, limitations regarding their application include the high equipment costs and energy requirements, and the selection of appropriate anodic materials.

### 2.2.3. Ozonation

Ozonation is a common method of water treatment in which hydroxyl radicals (•OH) generated from ozone ($O_3$) decomposition are used to oxidize pollutants including phenolic compounds (Manasfi, 2021; Pavithra et al., 2017). In alkaline mediums, ozone can act as a strong oxidizing agent, with a redox potential higher than hypochlorite, and it has higher solubility in water when compared to oxygen. Ozone can interact with pollutants and degrade them through two pathways. Directly, where contaminants interact with ozone or indirectly through hydroxyl radicals that are generated from the decomposition of ozone in the aqueous medium as illustrated in Fig. 3-A. Wastewater treatment with ozonate is preferred to low phenol concentrations to reduce costs, therefore it is used as a final disinfection step in wastewater treatment facilities (Sorokhaibam and Ahmaruzzaman, 2014). Numerous variables such as ozone dose, pH, and temperature impact the removal and degradation of phenolic compounds in such treatments. Many studies investigated the use of ozone in phenol removal. Turban & Uzman used an ozone bubble column containing phenol with initial concentrations ranging from 50 mg/L – 100 mg/L and stated that ozone was capable of complete phenol oxidation within 40 min (Turhan and Uzman, 2008). Wang et al. investigated the use of a self-design ozone generator for phenol degradation. It was noted that the degradation rate relied on the initial phenol concentration and reaction times. For example, 99% removal was achieved in 30 min when the initial phenol concentration was 100 mg/L, however, at an initial concentration of 3000 mg/L, around 480 min were needed to completely degrade phenol (Wang et al.,

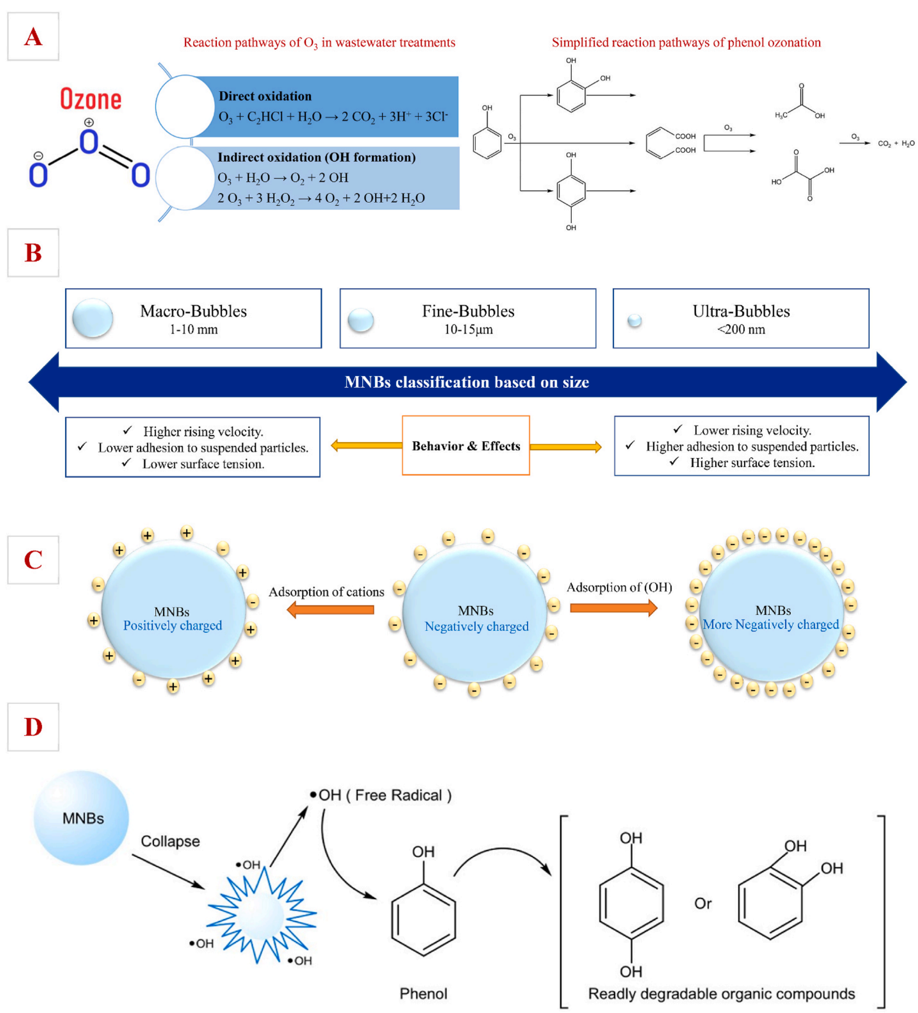


**Fig. 3.** **A**-Phenol ozonation process and reaction pathways. **B**- Classification of MNBs based on their size **C**-MNBs surface charges. **D**-Phenol degradation via free radicals generated from the collapse of MNBs.

2016). As mentioned, it is important to maintain alkaline conditions during the treatment process to ensure enough production of hydroxyl radicals for the complete oxidation of phenol.

One advancement in the ozonation process is through the use of micro-nano-bubbles (MNBs). Microbubbles are small-sized bubbles that have a diameter ranging from 10 to 15 μm whereas nano-bubbles have a diameter of fewer than 200 nm as illustrated in Fig. 3-B, collectively, these bubbles are called micro-nano-bubbles (Wang et al., 2020). MNBs have many important characteristics that could be beneficial for environmental applications. This includes their small size, large interfacial area, high internal pressure, low rising velocity, and long residence time. Additionally, MNBs demonstrate high stability, an exceptionally large surface-to-volume ratio, a high rate of oxygen dissolution, and a great ability to generate free radicals (Hu and Xia, 2018; Nirmalkar et al., 2018). In general, MNBs are negatively charged at various pH conditions as indicated by their zeta potential However, the surface charge of MNBs can be modified by the surrounding environment's composition, providing the opportunity for fine-tuning their properties for optimal performance in different applications as noted in Fig. 3-C. This feature of MNBs is important for the interactions between these bubbles and other compounds or pollutants in water as it determines the magnitude of electrostatic attraction and repulsion in water treatment systems (Jia et al., 2013; Takahashi, 2005). The bursting of the MNBs leads to the generation of free radicals (Liu and Tang, 2019). During bubble collapse, the Zeta potential usually increases and leads to the formation of free radicals (Takahashi et al., 2007; Xiong et al., 2018). These generated radicals have strong oxidizing power against a wide range of recalcitrant organic pollutants such as phenol as shown in Fig. 3-D, leading to its complete decomposition.

Many studies examined the use of MNBs in the elimination of refractory pollutants such as phenols. Wu et al. explored the use of microbubbles generated by cavitation for the degradation of phenol and it was compared between two conventional bubbles over various pH ranges. This study found that phenol degradation was more sufficient using microbubble ozonation where it required only 50% of ozone as compared to the conventional reactor. The half-time of phenol degradation using a conventional reactor was 18 min whereas, at the microbubble reactor, it took only 7 min at an initial phenol concentration of 9411 mg/L. Moreover, phenol degradation was found higher at high pH levels since phenol dissociates at such pH levels, and the ozone mass transfer rate was around 1.5 times higher compared to the conventional bubbles reactor (Wu et al., 2019). Another important feature of MNBs technology is that it could be used effectively in the in-situ treatment of groundwater. Compared to MNBs, macro-bubbles have a short lifetime and a small zone of influence, thus their use in water treatment is less efficient. Several studies have shown that MNBs can increase the dissolved $O_2$ levels in groundwater by increasing the mass transfer rate and thus lead to the oxidation of dissolved organic pollutants (Hu and Xia, 2018; Liu and Tang, 2019).

To sum up, in alkaline conditions ozone can efficiently treat many organic pollutants, including phenol at concentrations up to 9000 mg/L both in-situ and ex-situ. Many advancements have been reported in the literature, yet further improvements are required to optimize the use of MNBs with air instead of pure $O_2$ or $O_3$ to make the process more economically feasible while maintaining adequate removal efficiencies.

### 2.2.4. Advanced oxidation processes

Advanced oxidation processes (AOPs) are chemical technologies that remove and oxidize soluble organic effluents from water based on the in-situ generations of strong oxidants such as hydroxyl and sulfate radicals. AOPs have been developed to overcome the limitations of ozone treatment alone. These processes combine ozone with UV, catalysts, and hydrogen peroxide to optimize the degradation of pollutants into water and $CO_2$ (Ghime and Ghosh, 2020). In acidic and neutral environments, ozone has low solubility and stability, thus the formation of hydroxyl radicals becomes slow, and by using hydrogen peroxide or UV with ozone, more hydroxyl radicals are produced to accomplish higher oxidation levels. However, studies are required to understand the different scenarios in terms of by-product generation, pollutant decomposition pathways, and toxicity of end products.

An extensive study investigated several AOPs including $O_3$, $O_3/H_2O_2$, UV, UV/$H_2O_2$, UV/$O_3$, $O_3$/UV/$H_2O_2$, $Fe^{2+}/H_2O_2$ and photocatalysis processes for the oxidation of phenol in aqueous medium with an initial concentration of around 100 mg/L. Among all, the Fenton-based process demonstrated the fastest removal rates for phenol in wastewater with removal efficiency ranging from 32 to 100% within 9 min. It was noted that the photocatalysis took the longest period of degradation (150 min) where the removal efficiency ranged between 42% and 77%. However, the authors stated that from an economical point of view, ozonation was found to be more cost-effective (Esplugas et al., 2002). In another study, phenol with an initial concentration of 1000 mg/L was photo-oxidated using UV-$H_2O_2$. At ambient temperature and pressure and a pH of 3.5, it was found that 99% of phenol was degraded within 90 min with an oxidant concentration of 34,000 mg/L (Primo et al., 2007). It is evident that some AOPs could treat higher concentrations of pollutants compared to ozonation alone; however, it also requires large quantities of oxidants and has higher energy requirements.

### 2.2.5. Photocatalysis

Photocatalysis is an advanced oxidation process in which light energy is used to drive chemical reactions. When the catalyst absorbs energy either from UV light or visible light, an excited electron (−)/hole (+) pair is formed with photons of energy greater than or equal to the band gap energy. Due to their activated state, the electron and hole perform chemical reduction and oxidation of oxygen and water leading to the production of reactive species as illustrated in Fig. 4. Holes can oxidize adsorbed organic matter or water, producing hydroxyl radicals HO•. Electrons, on the other hand, reduce $O_2$ to the superoxide radical anion ($O_2$•). These $O_2$• and OH• radicals will in turn degrade organic and inorganic pollutants including phenol into $CO_2$ and $H_2O$. Commonly used catalysts include zinc oxide and titanium dioxide. Photocatalysis is a promising cost-effective technology that can effectively degrade a wide range of toxic compounds including phenol. One of the main advantages of this technology is that it requires low energy to operate. Since it uses sunlight as a source of energy, it significantly reduces the cost of operation compared to traditional treatment methods. Additionally, photocatalysis is a recyclable process, which means that the catalyst can be recovered, and reused, thus reducing the need for additional materials and minimizing waste generation. This results in lower operating costs and makes the technology more sustainable in the long run (Ansari et al., 2019; Mohamed et al., 2020; Pawar and Lee, 2015).

Many studies have investigated the use of photocatalysis in the degradation of phenol as noted in Table 2. Belekbir et al. photocatalyzed phenol using nanosized metal-impregnated $TiO_2$ under near-UV light. It was found that complete phenol degradation under NUV-Vis light irradiation using $TiO_2$–Cu was achieved with an initial phenol concentration of 50 mg/L, neutral pH, and ambient temperatures. This process was found to be slower when compared to the use of UV light, however, economically, a longer time using near UV–Vis radiation counter-weighted the expenses of utilizing UV irradiation sources (Belekbir et al., 2020). Even though many studies have analyzed the effects of photocatalysis pollutant degradation but faced an issue with how to accelerate the interfacial charge transfer economically. Thus, Pardeshi & Patil used a slurry batch reactor to degrade phenol using sunlight instead of UV lamps (approximately 4% UV light and 43% visible light) as an energy source to reduce operational costs. The experiments were performed at ambient temperatures in the presence of ZnO (250 mg/100 mL), and it was found that phenol solution with an initial concentration of 75 mg/L was entirely degraded to $CO_2$ and $H_2O$ within 8 h (Pardeshi and Patil, 2008). From these studies, it is evident that photocatalysis

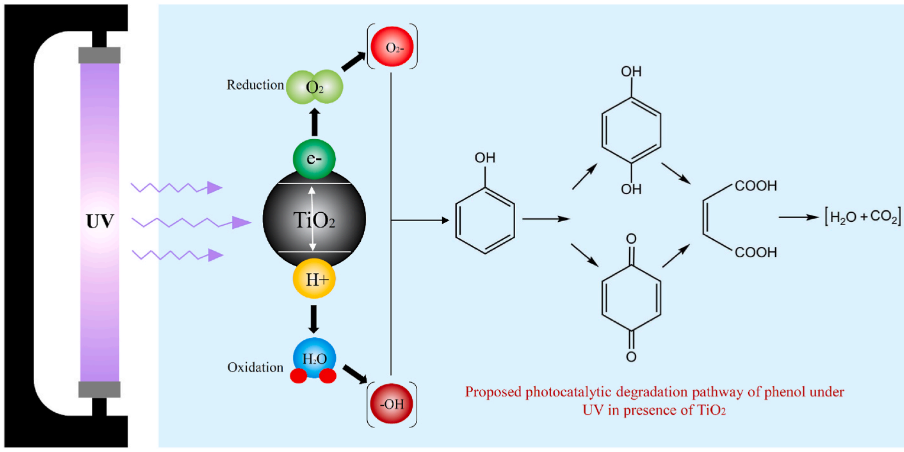


**Fig. 4.** UV photocatalysis of phenol using $TiO_2$.

could be used to treat low-strength wastewater with high efficiency.

### *2.2.6. Liquid-liquid extraction*

Liquid-liquid extraction (LLE) is another chemical method of separation that is based on the compound's relative solubility. LLE methods have been used traditionally for the removal of organic compounds from wastewater. The advantages of these methods are contributed to their non-destructive nature, high selectivity and solubility, and their ability to treat different concentrations of pollutants (Mohamad Said et al., 2021). Organic compounds including phenol can dissolve in organic solvents such as butanol, octanol, dichloromethane, alcohols, esters, ethyl acetate, butanone, benzene, and toluene (Cablé et al., 2022; Patel and Desai, 2022). In addition, a mixture of extraction solvents such as 2-octanone and n-octanol could also efficiently recover phenol from water (Wang et al., 2022). Many studies have shown that different solvents are capable of efficient separation of phenol from wastewater. Recently, Patel and Desai extracted phenol from synthetic and pharmaceutical wastewater using toluene, it was found that 20% toluene removed 60% and 68% of phenol synthetic and real wastewater respectively, with an initial phenol concentration of 50,000 mg/L and a pH of 7 (Patel and Desai, 2022). Jiang et al. also extracted phenol with an initial concentration of 6000 mg/L from wastewater using octanol as a solvent with an extraction efficiency of 99%; moreover, it also can recycle the extracting solution, leading to reduced operating costs. The authors noted that phenol extraction efficiency was high due to higher interaction energy between the phenol and the extractant, leading to stronger hydrogen bond formation and more stable complex formation (Jiang et al., 2003).

Even though extraction processes have been used efficiently for many years in the recovery of various organic compounds, these processes have several disadvantages mainly the toxic nature of the organic solvents. Therefore, research has been focusing on deep eutectic solvents (DESs) as a group of solvents that have a lower impact on the environment. Deep eutectic solvents are considered green solvents that could be used for the extraction of various compounds including phenol (Cablé et al., 2022). These solvents are non-volatile with high thermal stability and can readily dissolve many organic and inorganic compounds. Many studies investigated deep eutectic solvent (DESs) abilities to extract phenol as noted in Table 3. Sas et al. recently studied DESs based on (menthol, thymol, and organic acids) for the extraction of phenolic compounds. It was noted that DES based on menthol had an extraction efficiency of 70% with various initial phenol concentrations ranging from 5 mg/L to 1500 mg/L. In this study, it was mentioned that the higher extraction efficiencies were due to higher hydrophobic interactions between the pollutants and the DESs as illustrated in Fig. 2-C (Sas et al., 2019).

Ionic liquids (ILs) are another group of green solvents that are composed of ions with melting points below 100 °C (Sood et al., 2021). Ionic liquids are composed of organic cations and inorganic anions (Shang et al., 2021). Ionic liquids are considered promising green solvents to many organic and inorganic compounds due to their low vapor pressures, thermal stability, and non-volatile nature (Shang et al., 2021; Welton, 2004). Due to their benign nature, the usage of ILs in wastewater treatment is considered viable, environmentally friendly, and an alternate approach to toxic chemical solvents (Khraisheh et al., 2021). Over the years, ILs have been used in water treatment as a part of Liquid-liquid extraction technologies, in membrane technologies, and finally in adsorption processes. These studies focused on identifying the effects of ILs in the separation of various compounds including phenolic compounds.

Many studies have shown that different ILs could be applied for the extraction of phenolic compounds as noted in Table 2. In one study, ILs 1-ethyl-3-methyl imidazolium cyanoborohydride were used in a liquid-liquid extraction system to separate phenol at an initial concentration of 100 mg/L. It was found that ILs extracted selectively around 95% of the phenol from the synthetically prepared organic oil mixture of toluene and benzene showed acceptable efficiencies up to 6 times of reuse (Mathews et al., 2019). In another study, several types of ILs containing hydroxyl, benzyl, and dialkyl groups were tested for their ability to extract phenolic compounds including phenol. The authors noted that the extraction efficiency of these phenols depended on factors such as pH levels, the ratio of phases, salt contents, and the structure of the IL used. For instance, around 82% extraction efficiency was achieved at pH 9 using 1-butyl-3-(8-hydroxyoctyl) imidazolium hexafluorophosphate. The authors also state that the non-ionized phenols were found to be more easily transferred into the ILs phases as a result of by hydrogen-bonding and phenols' hydrophobicity (Fan et al., 2014).

The supported liquid membrane (SLM) process is a new extraction technology used for the treatment of wastewater containing organic compounds. SLM processes have several advantages over solvent extraction including high selectivity and speed, large mass-transfer force, minimal extractant loss, and reduced capital and operating costs (Kocherginsky et al., 2007). In a study, phenols were separated and recovered from an aqueous solution by a green membrane system where vegetable oil was used as a green polypropylene-hollow-fiber supported liquid membrane. It was noted that phenols separation with an initial concentration ranging between 4800 and 5200 mg/L reached 95% (Mei et al., 2020). In another study, polypropylene hollow fiber membrane (PP-HFM), was improved by grafting heptadecafluoro-1,1,2, 2-tetradecyltrimethoxysilane (FAS) and $SiO_2$. The modified SLM showed significant enhancement in stability and achieved average phenol removal of 75% in 16 days (Sun et al., 2017).

Solvent extraction is one of the most efficient processes for treating high-strength wastewater containing up to 50,000 mg/L of phenol, with

**Table 3**
Examples of various types of adsorbents and their phenol adsorption capacities and mechanisms.

| Adsorbent | Conditions T (°C), pH | Adsorption capacity (mg/g) | Removal (%) | BET Surface area ($m^2/g$) | Proposed removal mechanisms | Reference |
|---|---|---|---|---|---|---|
| **Biopolymers** | | | | | | |
| Magnetic chitosan | 20, 3 | 51.68 | 97 | – | At low pH, less repulsion between the negative chitosan and phenols and more Van der Waals attraction, hydrophobic interaction, and hydrogen bonding | Salari et al. (2019) |
| Crab shell chitosan | – | 59.3 | – | 191 | Mesopore filling and ionic interactions | Francis et al. (2020) |
| Polyacrylamide/starch hybrid hydrogels | – | 21 | – | – | Interactions between NH/OH of the polymeric structure and OH of phenol | Dutra et al. (2021) |
| Cellulose-based hydrogel | 25,6 | 80.71 | – | – | The external layer of α-cyclodextrin is enriched for phenol by inclusion complexation, with hydrogen bonding as the driving mechanism. The internal pyridine group has a strong affinity for phenol, which encourages additional phenol adsorption. | Guo et al. (2022) |
| Bacterial cellulose nanofibers | 25,8 | 146 | 97 | 342.1 | Electrostatic interactions were found to contribute to the generation of specific recognition binding sites. | Derazshamshir et al. (2020) |
| **Carbon-based adsorbent** | | | | | | |
| AC (coconut shell) | 55,4 | 144.93 | – | 620.49 | The lower content of total oxygen-functional groups on AC strengthens the π-π dispersive force interaction between the phenol molecule and AC sample, which leads to higher phenol adsorption capacity. | Zhang et al. (2016) |
| AC (coconut shell) | 25,7 | 212.96 | – | 1025.02 | Electrostatic interactions and surface area and pore diffusion led to higher adsorption | Xie et al. (2020) |
| GO | 30,7 | 10.23 | 74 | 312.00 | Interactions between phenoxide ions and GO surface | Mukherjee et al. (2019) |
| GO-PPY | 25,6 | 201.40 | – | – | Ion exchange, π–π electron donor-acceptor (EDA) interaction, hydrophobic interaction, and Lewis's acid-base interaction. | Hu et al. (2015) |
| Nitrogen-doped Reduced graphene oxide | 30,6 | 155.82 | 99 | 245.71 | The removal efficiency was attributed to π-π and hydrophobic interactions | Zhao et al. (2021). |
| Horseradish Peroxidase Immobilized on Graphene Oxide/$Fe_3O_4$ ($H_2O_2$) | 25,7 | – | 95 | – | Peroxidases catalyze the oxidation of organic and the reduction of ($H_2O_2$) by donating electrons that bind to substrates to break them into harmless components | Chang et al. (2016) |
| Carbon nanotubes-PEG | 20,6 | 21.23 | 100 | – | The dispersive interactions between the aromatic rings of phenols and the basal plane of CNT/PEG replenished with a high π-electron density could contribute to phenol removal | Bin-Dahman and Saleh (2020). |
| Polymeric resins | 20, - | 22.0 | – | – | Ion exchange was the predominant process responsible for phenols removal | Víctor-Ortega et al. (2016) |
| Acrylic ester-based crosslinked resin | 25,7 | 1000 | – | – | Adsorption by BMS-resin is attributed to the H-bonding between the ester groups on the surface of resin beads and the OH groups of phenol | Qiu et al. (2014) |
| CNT-DESs | 25, 7 | 298 | – | – | The hydrophobic interaction/π-π interactions resulted in the adsorption of phenol | Lawal et al. (2019) |
| Poplar AC | 28, - | 625.00 | – | 2711 | Surface area and aromaticity could increase π–π dispersion interaction as well as the reaction between the active site and phenol. Mesopores also enhance mass transfer as well as pore accessibility | Hwang et al. (2017) |
| **Industrial waste materials** | | | | | | |
| AC- oily sludge | 25,6 | 434.78 | – | 2263 | π-π interaction, electron-donor acceptor complex formation, and the hydrogen bonding | Mojoudi et al. (2019) |
| Fly ash- PDDA | 25,7 | 13.05 | 95 | – | The coating of cationic polyelectrolyte film onto FA introduced surface polarity which enhanced the mass transfer of phenol to the PDDA-FA surface | Oyehan et al. (2020) |
| Sludge Based AC-MgAlFe-LDH | 35,6 | 216.76 | – | 320.58 | The main phenol adsorption mechanisms π–π interactions because of the phenol aromatic ring interaction with that of the SBAC-MgAlFe-LDH via charge transfer, dispersive force, and polar attractions. OH groups of the adsorbent and hydrophobicity of phenol also play important roles in the adsorption process | (Mu'azu et al., 2021) |
| Clarified sludge | 35,7 | 1.05 | 63 | 78.54 | The presence of oxides and silica (−) on clarified sludge surface had a high affinity towards adsorption of the OH group of phenol leading to hydrogen bonding | Mandal and Das (2019) |
| **Agricultural Waste materials** | | | | | | |
| Activated Biochar | 20, - | 303.00 | 95 | 881 | High surface and pore volume and oxygenated functional groups, especially carbonyl groups were responsible for phenol removal. | Braghiroli et al. (2018). |
| Date palm frond biochar | -, 6 | 17.38 | – | 245.82 | Electrostatic interactions between the biochar and phenol molecules | Fseha et al. (2023) |
| Sulfuric acid-treated pea shells | 25,7 | 125.77 | – | 7.07 | The electrostatic interactions were very high between phenol and the adsorbent surface | Mishra et al. (2021) |

(continued on next page)

**Table 3** (*continued*)

| Adsorbent | Conditions T (°C), pH | Adsorption capacity (mg/g) | Removal (%) | BET Surface area ($m^2/g$) | Proposed removal mechanisms | Reference |
|---|---|---|---|---|---|---|
| Neem leaves | 30,3 | 74.90 | 97 | 370 | At low pH, the $H^+$ ions suppress the ionization of phenol, leading to phenol adsorption | Mandal et al. (2020a,b) |
| Ziziphus leaves | 25,6 | 15.00 | – | – | In the acidic medium, the surface of the adsorbent is dominated by positive charges which increases the attraction between phenolate and the surface, thus, enhanced adsorption is observed | Al Bsoul et al. (2021) |
| Pomegranate Peel Carbon | 25,7 | 148.38 | 98 | – | Phenol interacts with the functional groups of the adsorbent | Afsharnia et al. (2016) |
| AC- *Ceiba speciosa* wastes | 25, 7 | 156.70 | – | 842 | π–π interactions, hydrogen bonds, surface area, and pores were responsible for phenol adsorption | Franco et al. (2021) |
| AC- lignocellulosic wastes (Sugarcane bagasse) | 25,4 | 158.96 | – | 1053 | Phenol molecules will interact with the positively charged surface of carbon through electrostatic attraction. | El-Bery et al. (2022) |
| **Clays and muds** | | | | | | |
| Red mud | 30,8 | 49.30 | 87 | 300 | Pore capture, hydrogen bond formation between the red mud surface and phenol, and electrostatic interaction between metal oxides (positive) and phenolate ions all lead to phenol removal | Mandal et al. (2020a,b) |
| Na–bentonite | 30,3 | 8.76 | – | 2.04 | Strong electrostatic interactions between its adsorption site and phenol. | Asnaoui et al. (2020) |
| Clay | 30,6.5 | 30.32 | – | – | Clay particles consist of active sites bearing negative charge which are neutralized by ions, therefore, enhancing the diffusion of phenol ions | Nayak and Singh (2007) |
| HDTMA-modified clay | 40,7 | 18.8 | – | – | The negatively charged phenolate anion is electrostatically attracted by the positively charged HDTMA–bentonite surface. | Gładysz-Płaska (2017) |
| HCL activated mud | 25,6 | 8.156 | – | 20.7 | Interaction between metal oxides and polar phenol molecules | Tor et al. (2009) |
| **Hybrid adsorbents** | | | | | | |
| Chitosan and silica | 25,8 | 149.25 | 86 | 1700 | Removal was higher because phenol was unionized, and the dispersion interaction was predominant | Fathy et al. (2020) |
| MOF/GO | 25,7 | 212.76 | – | – | The presence of GO leads to the adsorption of phenol via hydrogen-bond and π-π interactions | Karamipour et al. (2021) |
| Synthetic zeolite modified with chitosan | 25,6 | 5 | – | 53.5 | The hydroxyl groups in the chitosan chain form hydrogen bond with the –OH group present in phenol molecules | Bandura et al. (2020) |
| Graphene oxide-coated biochar | 30,7 | 23.47 | – | – | Factors like hydrogen bonding, electrostatic interaction, van der Waals force, and intra-particle diffusion all lead to phenol adsorption | Manna et al. (2019) |
| Horseradish peroxidase immobilized Chitosan–halloysite hybrid-nanotubes | 25,7 | – | 98 | CTS–HNT 55.2 | Peroxidases catalyze the reduction of $H_2O_2$ and the oxidation of organics by donating electrons that bind to other substrates such as ferricyanides and ascorbate, to break them into harmless components | Zhai et al. (2013) |
| Laccase immobilized on copolymer nanofibers | 50,5 | 40.33 | ~90 | – | π-π interaction, hydrogen bonding between the electron-donating atoms and the hydrogen atoms of phenolic pollutants, and oxidation by laccases, all contribute to phenol removal | El-Aassar et al. (2020) |

removal efficiency ranging from 60 to 99%. Therefore, it is commonly used as a pretreatment step by petroleum and coke conversion industries. However, the use of conventional organic solvents in solvent extraction is associated with the use of toxic and hazardous substances. This necessitates the search for greener alternatives such as DESs and ILs as these have shown promise as green solvents. Yet further research is needed to develop solvents with extraction efficiencies that are comparable to those of conventional organic solvents.

### 2.2.7. Adsorption of phenolic compounds

One of the common methods for the removal of phenolic compounds is adsorption. Adsorption can be defined as the process in which an ion or a molecule called adsorbate sticks or attaches to the surface of a solid called adsorbent. The process of adsorption can occur through physical or chemical mechanisms. Physical adsorption involves the adsorbate adhering to the surface of the adsorbent through weak physical forces such as van der Waals interactions. In contrast, chemical adsorption typically involves stronger covalent bond formation between the adsorbent and the adsorbate (Akeremale et al., 2023). Like other chemical reactions, the adsorption processes are influenced by many factors as summarized in Fig. 6-B. The main feature that determines the capacity of an adsorbent is the surface area per volume, thus porous substances such as activated carbon and clays are considered fundamental adsorbents (Artioli, 2008). Over the years, substantial efforts have been made to produce adsorbents with high selectivity, efficiency, environmental compatibility, and cost-efficiency. Many of these adsorbents are being used in wastewater treatment due to their broad applicability and benign nature (Pavithra et al., 2017; Thakur and Kandasubramanian, 2019). Adsorption is an easy and energy-efficient process that can remove low and high concentrations of numerous contaminants including phenol from wastewater (Rout et al., 2023). Moreover, adsorbents could be regenerated/recycled, making this method more sustainable and cost-effective (Sajid et al., 2018; Wang et al., 2019). Previous studies stated that activated carbon is an effective type of adsorbent used for phenol remediation while other adsorbents used are titanium oxide, activated alumina-modified bentonite, GO, and biopolymers (Mohamad Said et al., 2021).

It could be noted from Table 3 that phenol can be removed from wastewater using various types of adsorbents including biopolymers, carbon-based adsorbents, clays and muds, industrial and agricultural waste-based adsorbents, and hybrid adsorbents. It was also noticed that the adsorption capacity of these materials could reach up to 1000 mg/g.

Biopolymers are polymers that are made of biological monomers that could degrade in the environment. These materials are produced by different organisms such as microorganisms, plant biomass, and agricultural wastes, and are composed of proteins, polysaccharides, and fats as summarized in Fig. 6-A (Pandian et al., 2021). Recently, biopolymeric composites have gained great attention due to their high surface area and functionality, structural stability, environmental compatibility, diverse applicability, and durability leading to superior adsorptive removal of various pollutants including phenol as noted in Table 3 (Udayakumar et al., 2021; Yaashikaa et al., 2022). Biopolymers could be prepared either by polymerization or through the termination of raw material. This process led to the production of good-quality biopolymers with enhanced features. However, the use of chemicals such as solvents could be problematic since these chemicals could be hazardous leading to the production of by-products that would require careful handling and further treatment. Some studies also suggested the use of green solvents such as deep eutectic solvents and ionic liquids to avoid the use of large qualities of organic solvents.

Even though adsorption processes have many significant advantages over physiochemical methods of wastewater treatment. However, it is vital to take into consideration the cost-effectiveness and fate of the adsorbents after use. In terms of economic value, since biopolymeric adsorbents could be made from agricultural wastes, the initial costs of the adsorbents could be acceptable, yet surface modification and labor costs could lead to an increase in expenses (Yaashikaa et al., 2022). For instance, a cost analysis was done for a modified pectin composite that was estimated to reach up to $70/kg. Whereas other studies showed that membranes made from biopolymeric materials (Arabic gum) were more cost-effective compared to conventional membranes (Aji et al., 2020). Additionally, the recyclability and regeneration of these composites could also significantly reduce the overall treatment costs. On the other hand, the environmental fate of spent adsorbents should also be considered carefully. Most biopolymers could be degraded in the environment by the actions of microorganisms and phytoremediators that oxidize these compounds and use them as carbon and nitrogen sources for their growth and metabolism. However, the adsorbed pollutants should be desorbed before the release of these composites into the environment (Yaashikaa et al., 2022). Several studies have investigated the use of biopolymers in the removal of phenolic compounds from aqueous solutions. In one study, researchers used chitosan beads modified with sodium alginate and $CaCl_2$. In this study, the maximum phenol sorption capacity was found to be 108.7 mg/g. The modification of the biopolymer has resulted in improved stability and sorption capacity (Nadavala et al., 2009). In another study, biopolymer-based biochar was used for the adsorptive removal of phenol and 2-nitrophenol. The adsorptive composite showed monolayer removal of pollutants. In addition, 2-nitrophenol was adsorbed more effectively than phenol due to the functional group ($NO_2$) that had a strong interaction with the adsorbent. This study also investigated the simultaneous removal of both pollutants in a binary system, which also showed similar trends where higher removal was achieved with 2-nitrophenol, indicating that it hindered the adsorption of phenol due to its stronger attraction to the receptor site (Li et al., 2019).

Activated carbon is one of the most commonly used adsorbents for the removal of organic pollutants (Allahkarami et al., 2023). However, its high cost and the environmental problems related to the regeneration and disposal of its waste dictate the use of more suitable and eco-friendly alternatives. AC is generated conventionally from coal and petroleum products and unconventionally from various agricultural and industrial wastes. These unconventional sources reduced the cost tremendously and are considered renewable sources of materials, thus, making the process of adsorption economically feasible (Issabayeva et al., 2018). In a study, commercial AC-SP1000 was used for phenol removal from synthetic and real wastewater. At neutral pH, ambient temperatures, and an initial phenol concentration of 5000 mg/L, the adsorbent was able to remove about 92% of phenol in the case of real syngas scrubber wastewater and around 99% of phenol from synthetic wastewater, demonstrating an adsorption capacity of 270 mg/g (Catizzone et al., 2021). Recently, several studies were conducted to evaluate low-cost and environmentally friendly materials for wastewater treatments as noted in Table 3. Most of these studies considered low-cost materials, mainly biosorbents that were generated from agricultural wastes, which were found to be able to reduce the availability and concentration of some organic compounds mainly phenolic compounds, polycyclic aromatic hydrocarbons, and industrial dyes. Additionally, industrial wastes are also being used as a source of adsorbents with high removal efficiencies (Adeniyi and Ighalo, 2019; Frezzini et al., 2019).

As mentioned, ILs are being studied as part of liquid-liquid extraction technologies to estimate their efficiency in phenol extraction. (Mohamad Said et al., 2021). Several recent studies have investigated the use of ILs for the removal of organic compounds from wastewater. The use of ILs is environmentally compatible, however, it is not cost-effective because these solvents are not highly recyclable and thus will be used in large quantities (Gholami-Bonabi et al., 2020). To overcome this issue, researchers have investigated the use of supported ILs where various materials are used to stabilize or immobilize the ILs (Laurent et al., 2008). For instance, in a study, amine-functionalized IL-modified graphene oxide was used as an adsorbent for phthalates from water. In this study, the modified GO was packed into a fixed bed column for solid-phase extraction. The study showed that the modified composite was able to extract contaminates with a minimum recovery of 95% and with high reproducibility (reaching up to eight times) without decreasing the efficiency of the absorbent in extraction. This study also concluded that ILs-modified GO can be used as a high-quality adsorbent in low-pressure columns (Zhou et al., 2016).

To further improve the adsorptive capabilities and overcome some of the issues associated with the use of GO in adsorption studies, a magnetic nanocomposite made of graphene oxide was modified by an IL (1-amino-3-methylimidazole chloride (LI-MGO)) and used to adsorb phenol from contaminated water. GO surface modification with ILs is possible due to the existence of carboxylic groups on the GO surface. It was noted in this study at the surface area increased from 64.32 $m^2/g$ to 110.44 $m^2/g$. This is due to the fact that the surface of the magnetic graphene oxide became rougher with IL modification. Furthermore, under optimal conditions, around 95% of phenol was removed from the solution, making this adsorbent a highly efficient and cost-effective process with high environmental compatibility (Gholami-Bonabi et al., 2020). Fig. 5 summarizes the adsorption process using M-GO-ILs. It could be concluded that adsorption is a highly economical and effective method to remove phenol from wastewater (Melaibari et al., 2023).

It could be noted from these studies that many adsorbents have extremely high phenol adsorption capacities that range from 1 mg/g to 1000 mg/g and removal efficiencies ranging from 50 to 99%. Additionally, these adsorbents could be prepared from waste materials, ranking the treatment process as the most economic method of phenol treatment (Magdy et al., 2021). However, to reduce the environmental impacts of these adsorbents, further improvements and studies are required to evaluate the regeneration potential and their disposal.

### 2.3. Biological degradation of phenolic compounds

Biological remediation of phenol has also been exploited to mitigate the negative effects associated with physiochemical techniques. These biological methods have numerous vital properties including their high specificity, accessibility, and the absence of production of harmful by-products. Various plant species and microorganisms have been used to biologically remove pollutants including phenolic compounds from wastewater. The biological treatment of phenolic compounds in wastewater can be done in three different ways. First, by phytoremediation where plants are used to extract, immobilize, contain, or degrade organic and inorganic pollutants (McCutcheon and Jørgensen, 2008). Second, by using Enzyme-based methods where enzymes extracted from

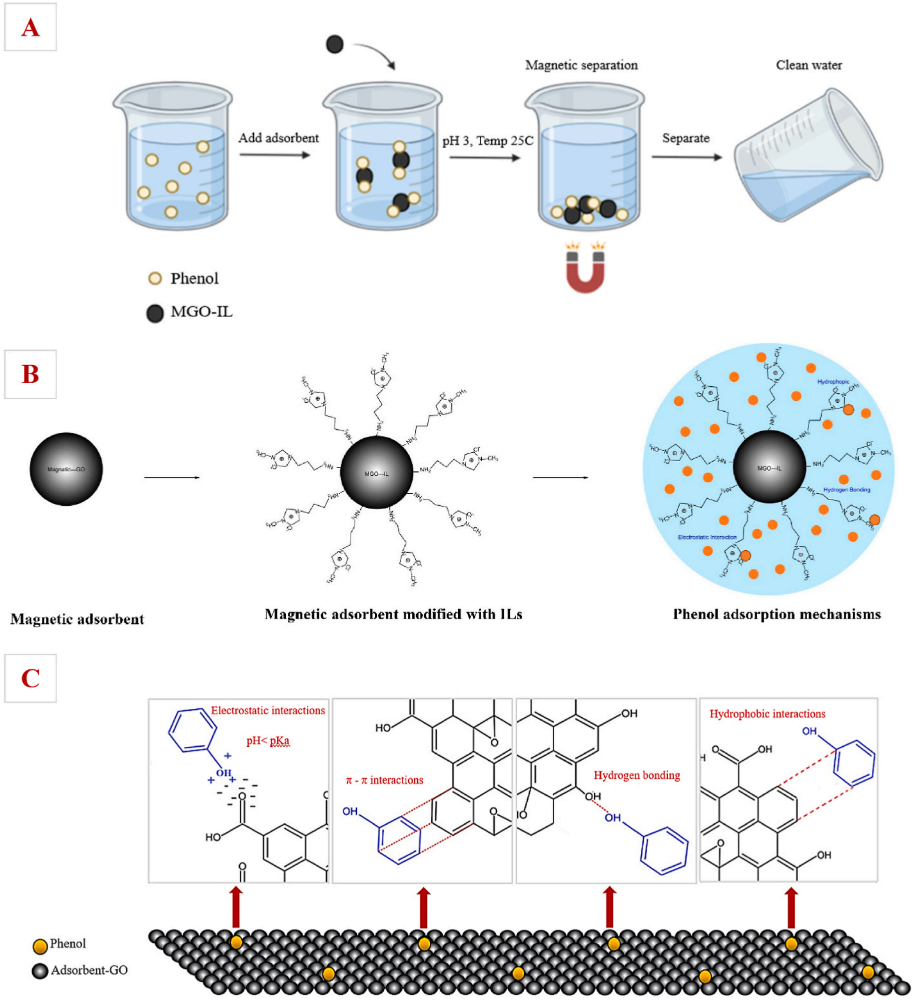


**Fig. 5. A-B**: An overview of batch adsorption of phenol using magnetic adsorbent modified with ionic liquids (Gholami-Bonabi et al., 2020), and **C**- Mechanisms of phenol adsorption onto graphene oxide (Bibi et al., 2022).

various living organisms such as plants, fungi, and bacteria are used in water treatment (Alshabib and Onaizi, 2019). Third, bioremediation where water is treated using microorganisms such as bacteria, fungi, and yeast that utilize the pollutants as their source of carbon and degrade it into $CO_2$ and $H_2O$ (Anku et al., 2017; Saravanan et al., 2021).

### 2.3.1. *Phytoremediation*

Phytoremediation is the process of degradation or accumulation of harmful pollutants into less harmful substances by plants as illustrated in Fig. 7-A. It is considered an eco-friendly, cost-effective method to remove, detoxify or immobilize different organic and inorganic pollutants. In addition to that, phytoremediation usually does not require costly inputs or expressive operational costs, and can also assist in enhancing biodiversity, thus is considered a more acceptable and preferred choice for remediation (Agostini et al., 2011). Phytoremediation of organic pollutants is usually done in two ways. First is phytoextraction and degradation where the pollutants are directly taken up and sequestered or degraded by plants. Several plant enzymes are involved in the sequestration and transformation of organic pollutants such as cytochrome P450 and glutathione-S-transferase. The second process is through plant-assisted rhizo-remediation. The pollutants are degraded by enzymes such as laccase, dehalogenase, and nitro-reductase, which are secreted by the plant or its rhizosphere microbial community. Soil microbes use plant root exudates (e.g., sugars, organic acids, etc.) as energy and carbon sources and in turn help in the degradation of pollutants. In addition, plant exudates can also increase the bioavailability of pollutants by increasing the solubility of these pollutants (Chen et al., 2013). To increase the phytoremediation efficiency and achieve higher degradation rates, especially when the contaminants are recalcitrant it is recommended to use multiple plants species, this will be a result of increased microbial functional diversity and biomass, and higher enzymatic activities (Wei and Pan, 2010). Fig. 7-A shows the various mechanisms involved in the phytoremediation of phenolic compounds. Detoxification of xenobiotics such as phenol starts with transformation where pollutants become more soluble. Enzymes such as peroxidases and laccases catalyze these reactions. Few studies showed that the crude enzymes of plants were able to oxidize phenol by cleaving the structural ring and forming muconic acid and catechol as an intermediate. Further oxidation will lead to the formation of fumaric acid. Following transformation, pollutants will be conjugated to the plant's endogenous compounds. This step is also vital because it increases pollutants' mobility and hydrophilicity. Examples of enzymes used in this step are glutathione s-transferase and N-glucosyltransferase. Conjugation can only partially reduce toxicity since the soluble pollutants will be accumulated in various plant tissues such as vacuoles with the help of ATP-binding cassette transporters. Following that, pollutants are either processed or moved out of the cell by exocytosis where they can accumulate in cell walls or apoplasts. These bound residues cannot be released from the plant matrix by solvent extraction. Finally, some pollutants including phenols can be excreted by the plant's leaves into the surrounding air (Agostini et al., 2011).

Enhanced phytoremediation of phenol could be achieved using

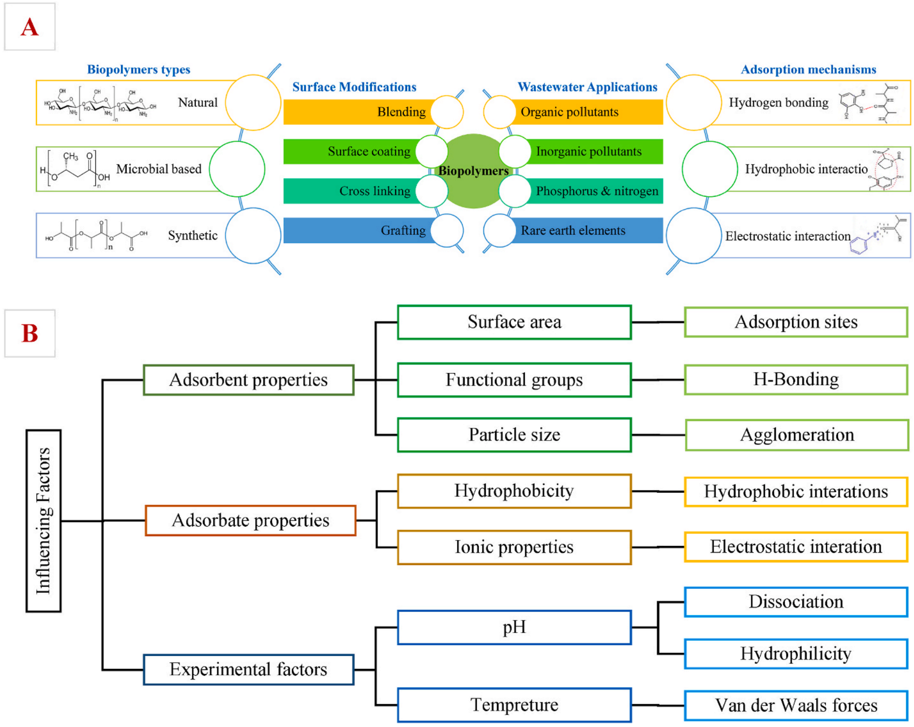


**Fig. 6. A**-An overview of the roles of biopolymers in water treatment, and **B**- Factors influencing the adsorption process.

several approaches such as studies that involve screening to identify the most suitable plant species, the use of transgenic plants with enhanced capabilities for phenol degradation, increasing soil microbial diversity, and optimization of agronomic practices to increase biomass production and subsequently, phenol degradation. Several studies have investigated the use of plants in the remediation of phenol-contaminated wastewater as noted in Table 2. Ibáñez et al. investigated a legume specie *Vicia sativa* L. ability to remediate phenol-contaminated wastewater. Phenol tolerance of the plant was assayed at different stages of growth, and it was noted that 30-day-old planets were able to tolerate phenol concentrations of 100 mg/L and with 60% removal efficiencies within 4 days. The activities of antioxidative enzymes, including peroxidase and ascorbate peroxidase, significantly improved with increased phenol concentration, whereas superoxide dismutase activity, malondialdehyde, and $H_2O_2$ levels did not change. The study suggested that *Vicia sativa* L. could be considered an exciting tool in the field of phytoremediation as it has an efficient protection mechanism against phenol-induced oxidative damage and could tolerate and remove phenol with concentrations up to 100 mg/L without phytotoxic effects (Ibáñez et al., 2012). Sosa Alderete et al. investigated the use of transgenic tobacco hairy roots (HR) system, which expressed basic peroxidase genes from tomatoes (TPX1 and TPX2) for phenol removal. The results showed that TPX1 is engaged in phenol removal not only when it was overexpressed in tomatoes, but also when it was expressed in other plants including tobacco. The removal efficiency using transgenic HR clones in the presence of $H_2O_2$ was optimal for TPX1/TPX2 clone, with a maximum value of about 90% at an initial phenol concentration of 100 mg/L, which represented an increment of about 15% compared with the wild type controls. The greater efficiency of TPX2 transgenic hairy root demonstrated that this peroxidase also participates in the removal of phenol (Sosa Alderete et al., 2009).

It could be concluded from these studies that phytoremediation could be used effectively with removal efficiencies ranging from 60 to 99%, for large volumes of wastewater with low phenol concentrations up to 100 mg/L. Further studies are required to investigate the resistant plant's ability to degrade contaminants and the possibilities of their application in constructed wetland systems. It is worth mentioning that phytoaccumulation is a slower process and it requires further treatment or disposal of plant materials to prevent any impacts on the environment.

### 2.3.2. Enzymes based remediation

Enzymes are a specialized class of proteins (catalysts) responsible for catalyzing numerous chemical reactions. Compared to inorganic catalysts, enzymes are more effective. In addition, enzymes show a greater specificity of the effect (Blanco and Blanco, 2017). The use of enzymatic-based technologies is considered a cost-effective and sustainable approach in the treatment of various pollutants. Microorganisms such as bacteria, cyanobacteria, fungi, and actinomycetes as well as several plant species are considered sources of different useful enzymes that can be used in the remediation of pollutants (Singh et al., 2021).

Several groups of enzymes are employed in the degradation of pollutants including hydrolases, oxygenase, oxidoreductases, laccase, and peroxidases (Singh et al., 2021). These enzymes can selectively and effectively degrade various pollutants at much faster rates when compared to other reactions. Another important advantage of enzymatic systems is the fact that enzymes can remove pollutants even in unfavorable conditions (e.g., temperature, pH, pollutant concentration, etc.) where bacteria might be inhibited. Additionally, enzyme-based technologies eliminate the time required for bacterial biomass generation. Compared to biodegradation, enzymes can be used in various conditions and will degrade pollutants into harmless products (Anku et al., 2017). Peroxidases are enzymes that are utilized extensively in phenol remediation due to the presence of heme cofactor or redox-active cysteine/selenocysteine residues in their active sites. Several studies investigated peroxidase from various sources for the removal of phenol from contaminated water. Kurnik et al. investigated the use of peroxidases produced from potato pulp waste by-products in the removal of phenol from synthetic and industrial wastewater. Phenol removal efficiency reached 95% with an initial phenol concentration of 94.11 mg/L where the enzymes maintained their activity at a pH range of 4–8 and were stable over a wide temperature range. Similar high efficiency was also noted with industrial effluents where 90% removal was achieved (Kurnik et al., 2015). In another study, peroxidases extracted from an

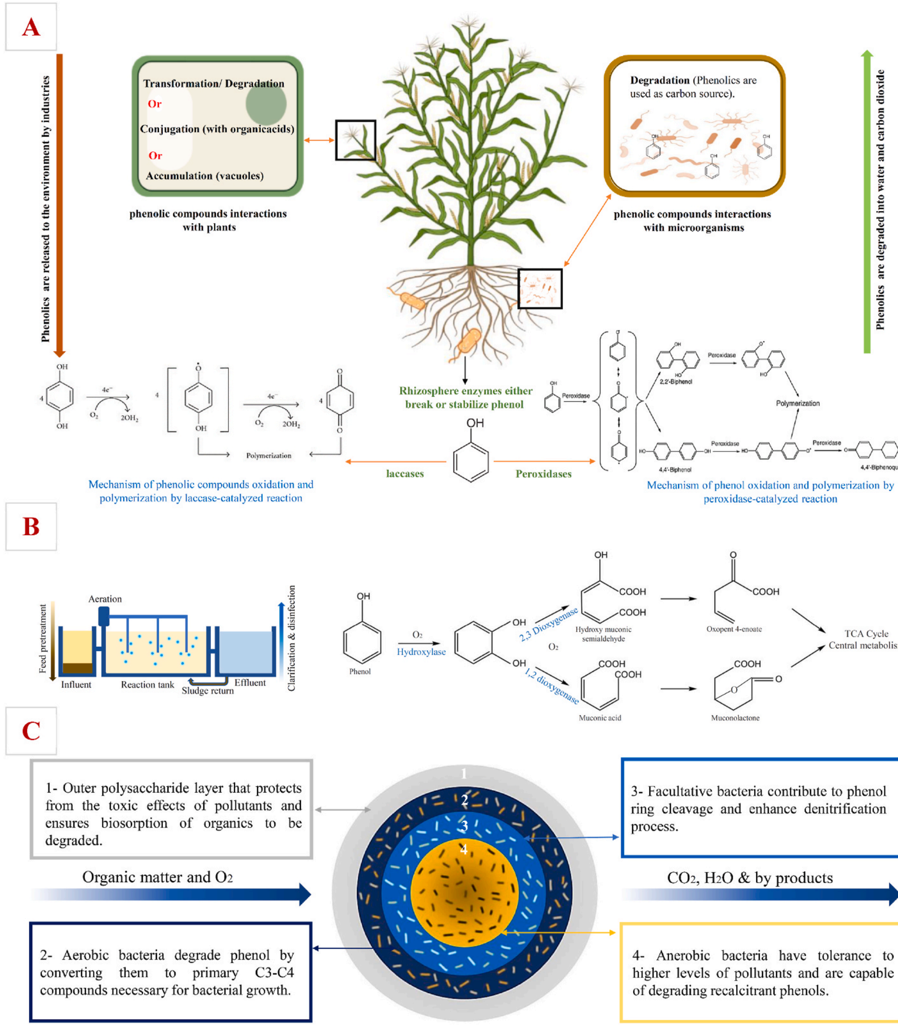


**Fig. 7.** **A**- Possible mechanisms involved in phenolic compounds phytoremediation, enzymes-based remediation, and bioremediation. **B**- Pathways of phenol degradation under aerobic conditions, and **C**-Graphical representation of granular activated sludge and the role of various microbial populations in the degradation of phenolic compounds.

invasive plant (*Prosopis juliflora*) were used for phenol remediation with an initial phenol concentration of approximately 40 mg/L, where crude peroxidases were found capable of degrading phenol within 30 min and with efficiencies higher than 90% for textile and leather industrial wastewaters. This study showed that agricultural waste materials could be used as an important source of potent enzymes that are highly efficient in the remediation of phenolic compounds (Garg et al., 2020). Such studies emphasize the importance of finding alternative sources of raw materials to promote sustainable production practices.

Enzymes could also be immobilized on carbon-based materials such as activated carbon and graphene oxide, which could be more cost-effective and can enhance their recovery, stability, and reusability in water treatment processes. Many studies investigated the use of immobilized enzymes in the removal of phenol from wastewater as noted in Table 3. In one study, laccase was immobilized on metal-chelated chitosan nanoparticles, and around 82% of phenol with an initial concentration of 20 mg/L was degraded within the initial 4 h compared to free laccase that degraded 80% of phenol after 12 h. The activity of the composite increased up to 96% in the presence of a redox mediator (ABTS). Laccase immobilization preserved the enzymatic activity over a wider pH range and showed a shift toward higher temperatures (30–40 °C) compared to the free enzymes (30 °C). In addition, the composite retained about 50% of the initial activity after eight reaction cycles (Alver and Metin, 2017). Besharati Vineh et al. immobilized horseradish peroxidase (HRP) by covalent bonding to reduced graphene oxide (RGO). The authors stated that the pH range and temperature were significantly improved compared to the free enzyme. Additionally, the removal efficiency was 100% and 55% for the immobilized HRP and free HRP respectively at an initial phenol concentration of 2500 mg/L (Besharati Vineh et al., 2018). On the other hand, Abdollahi et al. reported that tyrosinase nano-biocatalyst particles were able to degrade more than 70% of phenol with an initial concentration of 2500 mg/L within 4 h. At a lower initial concentration of 250 mg/L, the removal efficiency reached up to 100% for up to 3 cycles after which a decrease in removal efficiency was noted where it reached to about 55% removal at the 7th cycle (Abdollahi et al., 2018).

As mentioned, enzyme-based processes were developed to overcome the toxicity issues found in living systems. It was found that these processes selectively and effectively treat low concentrations of phenol up to 2500 mg/L with removal efficiencies ranging from 55% to 100%. Furthermore, the immobilization of enzymes in certain cases significantly enhanced pollutant degradation process efficiency, durability, the ability to treat higher-strength wastewater, and more importantly cost efficiency.

### 2.3.3. *Microbial bioremediation*

Many microorganisms can break down organic compounds into harmless products. These microorganisms use organic compounds such as phenol as their main carbon source and in turn, convert them into $CO_2$ and $H_2O$. The advantages of these methods include their low operational cost and environmental compatibility (Almasi et al., 2021; Anku et al., 2017). Numerous gram-negative bacteria can utilize phenol as their sole carbon source, including members of the three primary genera *Pseudomonas, Alcaligenes,* and *Acinetobacter* (Tian et al., 2017). Bacteria belonging to the genus *Acinetobacter* and Pseudomonas produce important enzymes such as phenol hydroxylase, which helps in the degradation of phenolic compounds (Cafaro et al., 2004; Gu et al., 2016). According to the literature, species belonging to the genus *Pseudomonas* and *Bacillus* are capable of phenol degradation by meta-cleavage pathway since these organisms have a broad range of catabolic enzymes involved in the process of phenol degradation (Panigrahy et al., 2022; Sarwade and Gawai, 2014). Both aerobic and anaerobic microorganisms are capable of phenol degradation (Almasi et al., 2018; Dargahi et al., 2017). In aerobic degradation, phenol hydroxylase enzymes catalyze the oxygenation of phenol to form catechol. Following that, a ring cleavage adjacent to or in between the two hydroxyl groups of catechol is created. Phenol hydroxylases can be simple flavoprotein monooxygenases or multicomponent hydroxylases. Catechol is oxidized via the ortho-cleavage pathway by catechol 1,2-dioxygenase (carbon bond between two hydroxyl groups). The final product of the pathway is a molecule that can enter the tricarboxylic acid cycle. Once the ring is opened, the degradation of the phenol can proceed as noted in Fig. 7-B (Mahiudddin et al., 2012). In the absence of oxygen, phenol can be degraded by the carboxylation in the *para*-position and the formation of 4-hydroxybenzoate. After that, thioesterification of 4-hydroxybenzoate to co-enzyme A allows consequent ring reduction, hydration, and fission. Para-carboxylation appears to be involved in the anaerobic degradation of several aromatic composites (van Schie and Young, 2000).

Fig. 7-B illustrates a typical aerobic-activated sludge reactor which is commonly used for organic waste treatment including phenol. These technologies use recircled microbial communities to degrade phenol in aerated systems and are capable of withstanding phenol concentrations up to approximately 2000 mg/L (Hussain et al., 2015). Many studies suggest that microbial consortium application for the remediation of phenolic compounds is a promising technique since mixed microbial populations can lead to higher tolerance to toxic pollutants, further, it also increases the efficiency of the degradation process by improving the synergetic activities of various microbial organisms that secrete various metabolites and enzymes. In such cases of co-metabolism, less toxic intermediate by-products are accumulated, unlike in the case of pure cultures (Patel & Kumar, 2016). Poi et al. used bacterial consortia consisting of 22 cultures including species belonging to *Pseudomonas* sp. *Bacillus* sp., and *Acinetobacter* sp. It was noted from this study that the biofilm-producing community was capable of remediating phenol-contaminated wastewater with a concentration of 407 mg/L using a bio-trickling reactor (Poi et al., 2017). Currently, many studies are conducted to investigate the ability of sequential anaerobic anoxic aerobic processes. Studies also show the significance of moving bed biofilm reactors (MBBRs) that are carbon-based in the fact they provide a protected surface where diverse groups of microorganisms can accumulate. The coupling of activated carbon with activated sludge has many benefits including the adsorption of pollutants and superior shock resistance. According to a study, two MBBRs were operated using different carriers' lignite-activated coke (LAC) and activated carbon (AC) to estimate phenol removal. In this system, phenol was used as the main carbon source in the first 3 stages of treatment, an initial decrease in degradation was noticed, however, the performance was stabilized probably due to the biofilm formation and the adsorption capacities of the used carriers reaching up to 96% removal of phenol. It was noticed that the LAC-based MBBR performed more efficiently when phenol levels were increased; nonetheless, both reactors had a similar tolerance limit to phenols (around 450 mg/L). LAC-based MBBR also demonstrated improved shock loading resistance at higher ammonia levels, leading to removal that is more efficient on phenols (88.68% vs 94.61%) when compared to the AC-based MBBR. The higher impact resistance was formed due to the resilient adsorption capabilities of LAC. In addition, the sludge flocs were enhanced in terms of compactness, stability, size, and size distribution, all leading to improved and enhanced resistance against high-concentration shocks and toxicity. In LAC-based MBBR, the degradation of phenols was dominated by excellent cooperation among core microbes. Facultative anaerobes *Cloacibacterium* and *Hydrogenophag*a contributed to phenol ring cleavage and enhanced denitrification. The predatory bacteria *Bdellovibrio* had a role in nitrogen fixation and biomass conversion by converting complex biopolymers to extracellular substances leading to more compacted flocs. As anaerobes on biofilm, *Tissierella* exhibited tolerance to higher levels of ammonia and stimulated methanogens. For archaea, *Thaumarchaeota* proportion of LAC was double the AC, especially for *Nitrososphaera*, which are exceptional nitrifiers. Due to the adaptability of *Comamonas* belonging to *Burkholderiales,* these species were found to be vital because it was capable of phenol biodegradation, nitrification, and denitrification. Moreover, they demonstrated interspecific cooperation with other bacteria. These species also produced biopolymers and created flocs for protection from toxicants and predators. Similar to *Comamonas*, *Pseudomonadales* were also of importance in phenols and ammonia degradation (Zheng et al., 2019). It could be concluded from these studies that co-metabolism is essential for the complete degradation of phenolics and for achieving higher toxicity tolerances (Tran et al., 2013).

Another promising technology in the field of phenolic wastewater treatments using aerobic bacteria is aerobic granular sludge (AGS) (Nancharaiah and Sarvajith, 2019). AGS can promote the degradation of phenolic compounds and increase toxicity tolerance due to the fact that these granules consist of highly dense, physiologically diverse, and spatially heterogenic microbial communities. The micro-environment of AGS is protected by the secretion of polysaccharides on the surface, thus reducing the toxic effects of pollutants as illustrated in Fig. 7-C. Exo-polysaccharides have many functional groups such as C=C, OH, and C=O that reduce phenol toxicity by enhancing the aggregation of AGS. In addition to that, polysaccharides also help in the biosorption of pollutants by providing efficient electrostatic forces, thus ensuring the degradation process. He et al. investigated the use of an AGS sequencing batch reactor for the simultaneous removal of phenol, nitrogen, and phosphorus from saline wastewater. It was noted that initial phenol concentration affected the removal of other pollutants by inhibiting the activities of the heterotrophic denitrifiers and stimulating phosphorus removal indicating the importance of co-metabolic activities in the removal of pollutants. It was noted that the reactor was able to degrade phenol completely with a concentration of up to 100 mg/L by the action of multiple bacterial species and their roles in the production of extracellular polymeric substances (He et al., 2021). Ho et al. investigated the use of AGS for high-strength phenol wastewater. It was noted that conventional activated sludge's ability to degrade phenol was inhibited at phenol concentrations above 3000 mg/L, however, when acclimated granules were used, effective degradation of phenol was achieved without severe inhibitory effects at a concentration up to 5000 mg/L. The authors also noted that by using acclimated granules, a shorter lag phase or faster degradation rate can be achieved compared to unacclimatized sludge (Ho et al., 2010).

It is evident that microbial biodegradation could be used effectively to treat medium and high-strength wastewater containing up to 5000 mg/L of phenol. Compared to phytoremediation and enzyme-based remediation this technology is more effective and could achieve 88–100% removal efficiencies especially when acclimatized microbes are used simultaneously in the treatment process.

## 3. Efficiency, sustainability, and economic feasibility of phenol treatment technologies

Various technologies can be used for the removal of phenol from

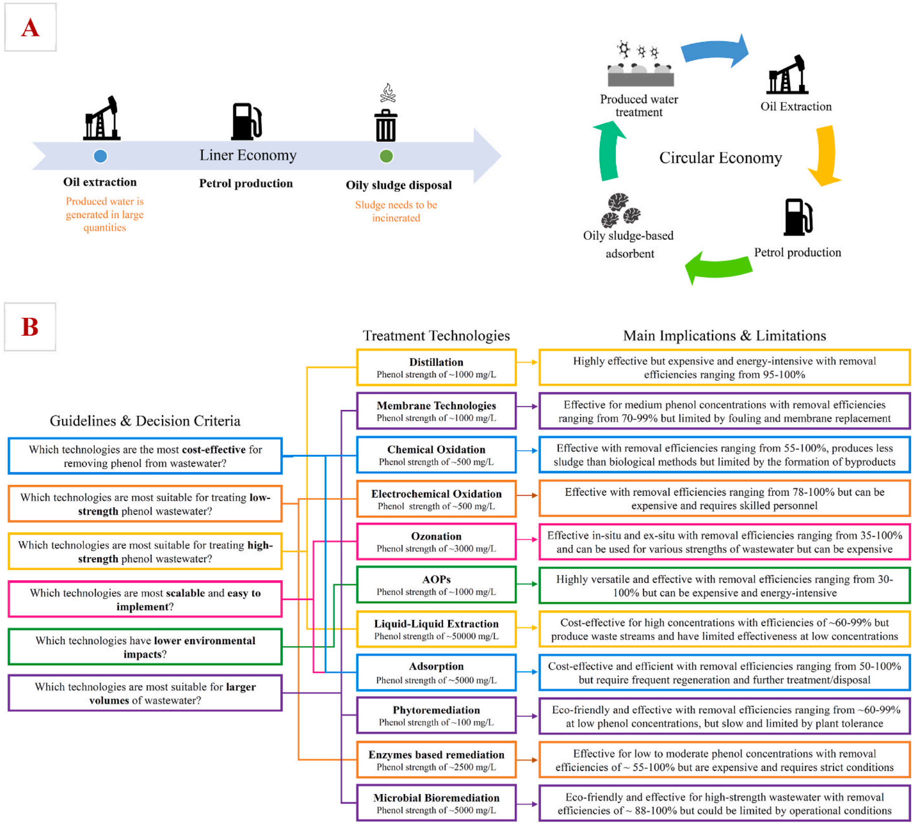


**Fig. 8.** **A-** An example of converting wastes into adsorbents and their roles in promoting sustainability and circular economy, and **B-** An illustration of guidelines and decision criteria used to select treatment technologies and their main implications, limitation, and efficiencies.

wastewater, each with its unique advantages, implications, and limitations as summarized in Fig. 8-B. For instance, technologies such as distillation, membrane filtration, chemical oxidation, electrochemical oxidation, ozonation, and advanced oxidation processes can be highly effective, but also expensive and energy intensive. On the other hand, technologies such as liquid-liquid extraction and adsorption can be more cost-effective but can produce additional waste streams. Additionally, approaches such as phytoremediation and microbial bioremediation can be environmentally friendly and sustainable but can be slow and limited by factors such as pollutant concentration and operating conditions. Overall, the choice of technology will depend on the specific characteristics of the wastewater, costs, and the desired treatment outcomes.

The sustainability of wastewater treatment technologies is becoming increasingly important as society continues to recognize the value of conserving natural resources and protecting the environment. Traditional wastewater treatment processes often rely on energy-intensive and resource-consuming methods, which can have negative impacts on the environment and contribute to climate change. Sustainable wastewater treatment technologies aim to reduce the environmental impact of wastewater treatment by utilizing renewable energy sources, minimizing waste generation, and optimizing resource use (Kadam et al., 2023). The use of agricultural and industrial waste for adsorption processes in wastewater treatment is an example of a sustainable technology that can provide both economic and environmental benefits (Steiger et al., 2023). By reducing the need for costly synthetic adsorbents and diverting waste from landfills, the use of waste materials in wastewater treatment can contribute to a more sustainable future.

The cost of any treatment method is an important factor that determines its feasibility and applicability in environmental applications. It is also important for decision-making. This review included many technologies that could be used in the treatment of phenolic wastewater, each with its cost. For instance, distillation and membrane filtration require higher capital costs for the equipment, membranes, energy consumption, and maintenance costs than other conventional treatment methods. Similarly, the use of ozone for phenol removal can be considered a relatively expensive treatment option due to the high capital costs of the ozone generator and the high energy required to produce ozone. Additionally, ozone treatment requires a high level of operator expertise and maintenance, which can add to the overall cost. In comparison, the cost of using biological treatment methods such as phytoremediation and microbial biodegradation is typically lower due to lower capital and maintenance costs. However, the cost of operation may be higher for biodegradation due to the energy required to maintain the ideal environmental conditions for microorganisms. In contrast to these technologies, the use of agricultural and industrial wastes as adsorbents for phenol removal is one of the most economical approaches that should be developed and used for wastewater treatment with costs

**Table 4**
Examples of costs of phenol removal using a few conventional technologies.

| Technology | Pollutant | Cost (unit) | Reference |
|---|---|---|---|
| Distillation | Phenol | 2185 \$/($m^3$/day) Estimate based on multi-effect distillation plant with 2531 $m^3$/day capacity | Fan et al. (2014) |
| Electro-Fenton | Phenol | 6.12 (US\$/$m^3$) | Magdy et al. (2021) |
| Photo-Fenton | | 1.55 (US\$/$m^3$) | |
| Photocatalysis | | 1.66 (US\$/$m^3$) | |
| Adsorption (AC) | | 0.74 (US\$/$m^3$) | |
| Photocatalysis/ Adsorption | | 2.19 (US\$/$m^3$) | |
| AOP | Phenol | 108 (US\$/$m^3$) | Krichevskaya et al. (2011) |
| Catalytic wet air oxidation | Phenol | 1.20 (US\$/kg phenol) | Mohammed et al. (2017) |

around $0.74/$m^3$ as noted in Table 4. Many adsorbents could be recycled and regenerated to be used for several cycles before being discarded, making them more cost-effective. In the literature, only a few reviews have conducted cost analyses for the treatment of phenolic wastewater. Magdy et al. conducted an economic analysis of five different technologies that are used in phenol removal. It stated that adsorption is the most cost-effective one as noted in Table 4 with costs less than $1/$m^3$. Another recent review has investigated the costs associated with the various adsorbents for the removal of a wide range of pollutants. This study stated that there is a broad variation in adsorbent costs, but the majority of adsorbents fall between 1 and 200 $/mol. Adsorbents at $<$ 1 $/mol threshold can be considered very cheap for the intended application, while those at $>$ 200 $/mol are believed to be highly costly (Ighalo et al., 2022).

The use of adsorption processes especially in wastewater treatment is significant due to their agreement with the concept of cleaner production and circular economies mainly when these adsorbents are generated from waste materials and used again as illustrated in Fig. 8-A. These materials have been gaining great attention due to their effectiveness in converting wastes into values, where various raw materials that are otherwise discarded are being efficiently used in the purification processes, with high efficiencies and low costs. Thus, turning waste into a renewable source of material for the removal of various pollutants such as nitrogen and phosphorus, heavy metals, and toxic organic compounds for effective treatment of wastewater. It is evident from Table 3 that waste-based adsorbents are highly efficient in removing phenol from water with adsorption capacities ranging from 13 mg/g and reaching 434 mg/g. Interestingly, waste-based adsorbents could also be used directly for water treatment without any modification or processing needed, as in the case of using Ziziphus and Neem leaves with capacities competing with many expensive carbon-based adsorbents (Sieradzka et al., 2022).

In conclusion, the choice of technology for the treatment of phenolic wastewater depends on various factors, including the characteristics of the wastewater, treatment outcomes, and cost considerations. Sustainable wastewater treatment technologies that utilize renewable energy sources, minimize waste generation, and optimize resource use are becoming increasingly important to reduce the negative impact of traditional wastewater treatment processes on the environment. While some technologies may be highly effective, they may also be expensive and energy intensive. Therefore, it is essential to conduct further research and a cost-benefit analysis of each technology before deciding on the appropriate treatment approach.

## 4. Concluding remarks

To sum up, wastewater contains various recalcitrant pollutants including phenol with different concentrations. According to the literature, many methods can be used for the treatment of such influents. Physiochemical technologies are very effective; however, these technologies could be expensive and not compatible with sustainable development goals. Therefore, green technologies should be developed to achieve the needed levels of treatment and to accommodate various types of wastewater.

Depending on the concentration of the pollutants, the type of treatment method can be selected, keeping in mind the cost-effectiveness of the selected treatment systems. From this review, it is clear that the concentration of phenol plays an important role in determining the appropriate treatment method. Consequently, it is recommended that reverse osmosis/nanofiltration chemical, electrochemical, and photocatalytic oxidation, ozonation, and biodegradation are used to treat phenolic wastewaters with low and moderate concentrations, whereas liquid-liquid extraction, and distillation, are suggested for higher phenol concentrations. It is worth noting that the majority of these technologies have demonstrated a remarkable effectiveness of over 90% in removing phenol from wastewater. Furthermore, with some alterations, many of these technologies have the potential to become even more environmentally friendly and sustainable by incorporating alternative materials, selecting non-conventional resources, and optimizing their recyclability. Integrated water treatment systems where more than one technology is used are also a great option for sustainable wastewater treatment since they showed their ability to handle various levels of concentration. Accordingly, a case-by-case study should be done to address the various limiting factors and select the appropriate technology.

Adsorption is an effective method for treating phenolic wastewater with various initial concentrations. Using adsorbents produced from agricultural and industrial wastes is a great solution, as they could be environmentally friendly, biodegradable, cost-effective, and highly efficient. However, there is still a lack of research on the recovery and recyclability of these adsorbents, which must be addressed to ensure cost-effectiveness. Scaling up the use of these adsorbents is also necessary, as most of the research is conducted as batch adsorption studies at laboratory scales. Additionally, it is important to explore alternative sustainable and green methods for synthesizing adsorbents with high removal capacities to promote environmental compatibility. With the use of low-cost and environmentally friendly adsorbents, adsorption processes have the potential to become a key solution in addressing water pollution and ensuring access to clean water.

## Declaration of competing interest

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

## Data availability

No data was used for the research described in the article.

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