

REVIEW

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Adsorption of catechol, resorcinol, hydroquinone, and their derivatives: a review

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Abstract

In recent years, there has been an increasing interest in finding innovative solutions for the efficient removal of contaminants from water, soil, and air. The present study reviews the adsorptive removal of catechol (C), resorcinol (R), hydroquinone (HQ), and their derivatives from various adsorbents. As an effective, efficient, and economic approach for water purification, adsorbents and adsorption processes have been widely studied and applied in different aspects for a long time. The role of various adsorbent materials like activated carbon, activated carbon cloth, carbon nanotubes, polymeric resins, organic clays, Fe(OH)₂, and TiO₂ was discussed together with that of other experimental parameters. In all the synthetic resins, particularly, aminated hypercrosslinked polymers have good adsorption capability for phenols. These polymeric adsorbents are suitable for industrial effluents containing C, R, HQ, and their derivatives. The adsorption capacities of the adsorbents reviewed here vary significantly depending on the characteristics of the individual adsorbent, the extent of chemical modifications, and the concentrations of solutes.

Keywords: Catechol, Resorcinol, Hydroquinone, Adsorbents, Wastewater

Review

Introduction

Phenol (P) and substituted P are important organic intermediates for the products of industry and agriculture [1]. For example, hydroxy aromatic compounds, such as catechol (C), resorcinol (R), and hydroquinone (HQ), were used widely as industrial solvents. C (1,2-dihydroxybenzene) is also widely used to produce food additive agents, hair dyes, and antioxidants [2]. R (1,3-dihydroxybenzene) is usually employed to produce dyes, plastics, and synthetic fibers [3-5]. Phenolic compounds, such as C, R, and HQ, were found in the effluents of industries such as textile, paper and pulp, steel, petrochemical, petroleum refinery, rubber, dye, plastic, pharmaceutical, cosmetic, etc. and in the wastewater of synthetic coal fuel conversion processes [6,7]. The effluents from synthetic coal fuel conversion processes may contain C and R concentrations ranging from 1 to 1,000 mg/l [8].

H. Reinsch first isolated C in 1839 by distilling catechin (from catechu, the juice of *Mimosa catechu* (*Acacia catechu* L.f)). The pyrocatechol is formed by the heating of catechin above its decomposition point. C is produced industrially by the hydroxylation of P using hydrogen peroxide [9]. C has also been produced by the hydrolysis of 2-substituted phenols, especially 2-chlorophenol, with hot aqueous solutions containing alkali metal hydroxides. Its methyl ether derivative, guaiacol, converts to C via hydrolysis of the CH₃-O bond as promoted by hydriodic acid. R is obtained by fusing many resins (galbanum, asafoetida, etc.) with potassium hydroxide or by the distillation of brazilwood extract. It may be prepared synthetically by fusing 3-iodophenol, phenol-3-sulfonic acid, or benzene-1,3-disulfonic acid with potassium carbonate; by the action of nitrous acid on 3-aminophenol; or by the action of 10% HCl on 1,3-diaminobenzene [10]. Many *ortho*- and *para*-compounds of the aromatic series (for example, the bromophenols, benzene-*para*-disulfonic acid) also yield R by fusion with potassium hydroxide. HQ is obtained from few methods: oxidation of aniline with manganese dioxide and sulfuric acid, followed by reduction with iron dust and water [11]. The second method consists of the alkylation of benzene with propylene to

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produce a mixture of diisopropylbenzene isomers from which, in a first step, the *para*-isomer is isolated. This is oxidized with oxygen to produce the corresponding dihydroperoxide, which is treated with an acid to produce HQ and acetone [12]. Finally, the oxidation of P with hydrogen peroxide can be used to produce a mixture of products from which both HQ and C (1,2-dihydroxybenzene) can be isolated [12].

C occurs freely in kino and in beechwood tar; its sulfonic acid has been detected in the urine of horses and humans [13]. HQ exists in a free state in pear leaves [14]. Arbutin, a glucoside of HQ, occurs widely in the leaves, barks, buds, and fruits of many plants [11], especially the Ericaceae [15]. C (*o*-benzenediol, 1,2-benzenediol, or 1,2-dihydroxybenzene) is a natural polyphenolic compound that widely exists in higher plants such as teas, vegetables, fruits, tobaccos, and some traditional Chinese medicines [16]. The smoke from 100 cigarettes contains about 10 mg of P and 50 mg of C besides other phenols [17]. HQ has been detected in cigarette smoke [18,19]. This compound occurs in the effluents resulting from different industrial activities such as photoprocessing [20], coal-tar production [21], and the paper industry [22].

The toxicity of C for microorganisms has been demonstrated in the past years [23-25] and has been suggested to be the reason for the difficulties in cultivating microorganisms on benzene, toluene, or chlorobenzene [23]. Several studies additionally indicated the toxicity of C for water flea, zebra fish, trout, rabbit, cat, rat, and mouse and for human cell lines [26]. C is strongly irritating to the eyes, skin, and respiratory tract, and it has been proven to cause DNA damage, vascular collapse, coma, and death. Between R and C, C is considered more toxic [11,27]. However, these compounds are considered as the primary pollutants in wastewater due to their high toxicity, high oxygen demand, and low biodegradability [8,28]. Consequently, their removal from wastewater has attracted significant environmental concerns. C, like other phenols, is of particular interest from a sanitary point of view due to its toxicity and deleterious effect on the quality of water supplies. In human medicine, HQ is used as a topical application in skin whitening to lighten the color of the skin as it does not have the same predisposition to cause dermatitis as metal does. There is inadequate evidence in humans for the carcinogenicity of HQ; however, it causes toxicity in several organs, notably the kidney and forestomach [12].

Additional file 1: Table S1 shows the hazard ranking of C, R, and HQ. The compound is readily absorbed from the gastrointestinal tract, causes hemolysis, degenerates the renal tubes, diminishes liver function, and accumulates in the bone marrow [29]. Its metabolites may initiate many cancers and neurodegenerative diseases

[30]. C is even more toxic than P since it provokes changes in the function of erythrocytes at doses as low as 50 µg/l compared to 250 µg/l of P [31].

The United States Environmental Protection Agency (USEPA) has designated granular activated carbon (GAC) adsorption as the 'best available technology' for removing organic pollutants [32]. P and associated compounds have been listed as priority pollutants by the Ministry of Environment and Forests (MoEF), Government of India, and USEPA. MoEF has prescribed that the concentration of phenols should not exceed 1.0 mg/l for their discharge into surface waters and 5.0 mg/l for their discharge into public sewers, on land for irrigation, and on marine coastal areas. These limits have generally been on the basis of the total phenols present in the effluent.

Conventional methods used in the remediation/degradation of C, R, and HQ are biodegradation [33,34], anaerobic biodegradation [35-37], anodic oxidation [38,39], photocatalysis [40], oxidative catalysis [41], etc. Adsorption techniques have gained favor in recent years because they are considered efficient for the removal of trace organic pollutants from water that cannot be removed using other treatment processes.

In addition, adsorption and desorption kinetics are technologically important because the diffusion within solid particles is a phenomenon of great importance in catalysis, metallurgy, microelectronics, materials science, and other numerous scientific and technological applications. Chemical kinetics explains how fast the rate of chemical reaction occurs and also on the factors affecting the reaction rate. The nature of the sorption process will depend on physical or chemical characteristics of the adsorbent systems and also on the system conditions.

Adsorption using GAC has been found to be an attractive process for the removal of phenols [6,42-50]. However, the cost of GAC and the loss of adsorption efficiency after regeneration of the exhausted GAC have limited its use in effluent treatment. Therefore, alternative low-cost, non-conventional adsorbents such as activated carbon cloth [51,52], waste Fe(III)/Cr(III) hydroxide [53], hypercrosslinked resin [53-55], TiO₂ surface [56,57], organoclays [58], bagasse fly ash [59], activated cashew nut shell [60], etc. have been investigated for the treatment of effluents. So, in this context, the present review is made for the adsorption of C, R, and HQ, and their derivatives onto different adsorbents.

Properties of C, R, and HQ

C, R, and HQ are available in the form of colorless crystalline and white crystals. C and R have phenolic odor and unpleasant sweet taste and become brown on exposure to air and light and pink on contact with air and light [61]. Small amounts of C occur naturally in fruits

and vegetables, along with the enzyme polyphenol oxidase. R crystallizes from benzene as colorless needles which are readily soluble in water, alcohol, and ether, but insoluble in chloroform and carbon disulfide. With concentrated nitric acid, in the presence of cold concentrated sulfuric acid, it yields trinitro-resorcin (styphnic acid), which forms yellow crystals, exploding violently on rapid heating. It reduces Fehling's solution and ammoniacal silver solutions. It does not form a precipitate with lead acetate solution, as the isomeric pyrocatechol does. Iron(III) chloride colors its aqueous solution a dark violet, and bromine water precipitates tribromoresorcin. Some of the physicochemical properties of adsorbents affect the adsorption process like water solubility, acid dissociation constant (pK_a) value, and octanol/water partition coefficient. R has more solubility (1,100 g/l) than C and HQ. The octanol/water partition value of R is 0.93, which is more than those of C and HQ. The pK_a values of C, R, and HQ were 9.25, 13; 9.4, 12.3; and 9.9, 11.6, respectively. Some more properties are given in Table 1. C can form stable complexes with various di- and trivalent metal ions, the complexes with trivalent ions being the most stable. C can also undergo redox reactions (Scheme 1 of [7]), cycling between C, semiquinone radicals, and *ortho*-benzoquinone.

Physicochemical characteristics of various adsorbents for removal of C, R, and HQ

Physicochemical characteristics of various adsorbents for the removal of C, R, HQ, and their derivatives from water were listed in Tables 2 and 3.

Usually, there are different processes for the preparation and pretreatment of adsorbents, namely either thermal procedures (physical) or chemical routes. In comparison with physical activation, there are two important advantages of chemical activation: One is the lower temperature in which the process is accomplished, and the other is that the global yield of the chemical activation tends to be greater since burn-off char is not required. For impregnation, the precursor with dehydrating agents was widely used as a chemical agent in the preparation of adsorbents. Knowledge of different variables during the activation process is very important in developing the porosity of materials which is sought for a given application. For example, chemical activation done by using H_2SO_4 , HCl or HNO_3 , $ZnCl_2$, $AgNO_3$, H_3PO_4 , H_2O_2 , etc. can improve the pore distribution and increase the surface area of adsorbents in the structure due to the use of different chemicals [42,44,51-53,58,70] and then heat treatment at moderate temperatures in a one-step process.

Table 1 Some of the physicochemical properties of C, R, and HQ

| Parameters | C | R | HQ | References |
|---|---|--|--|------------|
| Chemical structure | | | | - |
| | $C_6H_6O_2$ | $C_6H_6O_2$ | $C_6H_4(OH)_2$ | |
| Other names | Pyrocatechol; 1,2-benzenediol; 1,2-dihydroxybenzene | 1,3-Dihydroxybenzene; 1,3-benzenediol; 3-hydroxyphenol; resorcin | <i>p</i> -Benzenediol; 1,4-benzenediol; dihydroxybenzene; 1,4-dihydroxybenzene; quinol | - |
| λ_{max} (nm) | 275 | 273 | 289 | [63] |
| Boiling point (°C) at 101.3 kPa | 245.5 | 277 | 287 | [57] |
| Octanol/water partition coefficient (log P_{ow}) at 25°C | 0.88 | 0.79 to 0.93 | 0.59 | [64] |
| Density (g/cm ³) | 1.344 | 1.28 | 1.3 | - |
| Molecular weight (MW; g/mol) | 110.11 | 110.11 | 110.1 | - |
| Water solubility (g/l) at 25°C (C_s) | 430 | 1,100 | 59 | - |
| Molecular size (nm) | 0.55 × 0.55 | 0.56 × 0.47 | - | [57] |
| pK_a | 9.25, 13 | 9.2, 10.9 | 9.9, 11.6 | |
| Dipole moment (Debye; D) | 2.620 | 2.071 | 0.0 | [65] |
| Polarity/polarizability parameter (π ; cm ³) | $11.89 \pm 0.5 \times 10^{-24}$ | 59.11×10^{-24} | 0.21 ± 0.02 | [63,66] |
| Hydrogen-bonding donor parameter (α_s) | 0.85 | 1.10 | - | [63] |
| Hydrogen-bonding acceptor parameter (β_m) | 0.58 | 0.52 | - | [63] |

Table 2 Physicochemical characteristics of AC for removal of C, R, and HQ, and their derivatives from water

| References | Rajkumar et al. [66] | Kumar et al. [6] | Rodriguez et al. [67] | Mondal and Balomajumder [68] | Mohamed et al. [42] | Blanco-Martinez et al. [44] | Richard et al. [43] | Richard et al. [45] | Suresh et al. [46-48] |
|---|----------------------|------------------------|-----------------------|------------------------------|-----------------------------|-----------------------------------|---|---------------------|---|
| Absorbent | AC | AC | AC | GAC-SAB | Treated AC | Treated AC | AC | AC | GAC |
| Absorbates | C, R, P, and others | C and R | R | R and P | C, R, P, and others | C, R, and HQ | C and R | C | C, R, and HQ |
| Pre-treatment | - | Oven at 105°C for 72 h | - | - | - | HNO ₃ solution for 9 h | - | - | - |
| Moisture (%) | 3 to 6 | - | - | - | - | - | - | - | 9.04 |
| Ash (%) | 3 to 5 | - | - | - | - | - | - | - | - |
| Average particle size (mm) | 12/40 | 0.536 | - | 1.4 to 5 | - | - | - | - | 1.67 |
| BET surface area of pores (m ² /g) | 1,100 | 579.23 ± 17.02 | 864 | 778.12, 583.23, 393.61 | 43, 115, 152, 182, and 210 | 1,140 | 1,370 | 1,370 | 977.65 |
| BET average pore diameter (Å) | - | - | - | - | - | - | - | - | 18.79 |
| Pore volume (cm ³ /g) | - | - | 0.485 | 0.2371, 0.2044, 0.1692 | - | 0.51, 0.12 | - | - | 0.072 |
| Boehm titration (μmol/m ²) | - | - | - | - | 0.6, 2.15, 3.65, 4.05, 4.13 | 0.30, 0.60 | 0, 0.1, 0.4, and 0.02 (COOH, COO, OH, and CO, respectively) | - | 0.4, 1.8, and 3.3 (COOH, -COO, and -OH, respectively) |
| pH _{PZC} | - | - | 9.25 | - | - | 9.8 | - | - | 10.3 |

Volatile matter (%), fixed carbon (%), heating value, and ultimate analysis are not reported; AC, activated carbon; GAC, granular activated carbon; SAB, simultaneous adsorption biodegradation; C, catechol; R, resorcinol; P, phenol; HQ, hydroquinone, BET, Brunauer-Emmett-Teller.

Table 3 Physicochemical characteristics of other adsorbents for removal of C, R, HQ, and their derivatives from water

| References | Bayram et al. [69] | Huang et al. [55] | Liao et al. [70] | Shakir et al. [58] | Sun et al. [54] | Ayranci and Duman [52] | Yildiz et al. [51] | Namasivayam and Sumithra [53] | Vasudevan and Stone [56] |
|---|--------------------|--------------------|---|--------------------------------|-----------------------|--|-----------------------|---|--------------------------|
| Absorbent | ACC | HJ-1 | MWCNT | CTAB-B | AH and NDA-100 resins | ACC | ODTMA-B, HDTMA-B | Fe(III)/Cr(III) OH | TiO ₂ |
| Absorbates | C and R | C and R | C, P, and others | C | C and R | P, 4NP, HQ, and others | HQ and others | C | C and others |
| Pre-treatment | Warm (60°C) water | 1% HCl and ethanol | HNO ₃ (63%) and refluxed in oil bath at 140°C for 12 h | 85°C for 3 h and 105°C for 1 h | - | Washed with water and then dried under vacuum at 120°C | - | Washed with distilled water for three times and 60°C for 15 h | - |
| Average particle size | - | 0.4 to 0.6 mm | - | - | 0.4 to 0.6 mm | - | TOC = 19.8% to 24.28% | 250 to 500 μm | - |
| BET surface area of pores (m ² /g) | 1,870 | 750; 727 | 72; 121 | - | 934; 819; 726; 483 | 1,464 to 2,500 | 28.92 to 55.37 | 156 | 39.5 |
| BET average pore diameter (Å) | 6.51, 5.78 | - | - | - | 2.4, 2.4, 2.6, 3.0 | - | - | 7.6 | - |
| Pore volume (cm ³ /g) | 0.709 | - | 0.41; 0.49 | - | - | - | - | 0.297 | - |
| pH _{PZC} | 7.2 | - | - | - | - | 7.4 | - | 8.1 | 6.3 |

ACC, AC cloth; HJ-1, hypercrosslinked resin; MWCNT, multi-walled carbon nanotubes; CTAB-B, cetyltrimethylammonium bromide-modified bentonite; AH, aminated hypercrosslinked polymers; NDA-100, hypercrosslinked resin without amino groups; ODTMA-B, octadecyltrimethylammonium bromide; HDTMA-B, hexadecyltrimethylammonium bromide; TiO₂, TiO₂ Degussa P-25 surface; C, catechol; R, resorcinol; P, phenol; 4NP, 4-nitrophenol; HQ, hydroquinone, TOC, total organic carbon; BET, Brunauer-Emmett-Teller.

The raw sawdust was carbonized at 873 K in a nitrogen atmosphere, and concentrated H_2SO_4 was impregnated with carbonized materials in different concentrated H_2SO_4 /sawdust ratios from 3:1 to 6:1 (w/w) at 575 K for 4 h. The product was cooled to room temperature, washed with 5% H_2SO_4 and then with bi-distilled water until free from sulfate ions, and dried at 393 K for 6 h. GACs are placed in the reducing system with hydrogen under vacuum condition and then warmed at 573 K for 6 days. Finally, the AC is placed with HNO_3 solution in Soxhlet equipment for 9 h. The sample is then washed with distilled water until a constant pH value is obtained, and the AC is then dried at 383 K for 24 h. These GACs are used for the removal of C [42], HQ, and other phenols [44]. The chloromethylated PS beads were dried at 323 K in vacuum for 8 h and then swollen with nitrobenzene at 298 K overnight with anhydrous $ZnCl_2$ added into the reaction flask with the temperature at 323 K, and the beads were obtained after 12 h. The polymeric beads are washed with 1% HCl (w/w) aqueous solution and ethanol until the solution becomes transparent. Finally, they were extracted with ethanol for 8 h [55]. Shaker et al. [58] prepared the organobentonite by treating natural bentonites which are crushed and oven-heated at 85°C for 3 h and dried for 1 h at 105°C. The treated clay was washed with $AgNO_3$. Finally, the organobentonite was at last separated from water by vacuum filtration and dried for 1 h at 105°C, and a similar procedure was reported for the removal of HQ onto AC by drying under vacuum at 120°C [51,52].

Figures 1a,b,c and 2a,b,c show the yearly progress of publications and citations, respectively, in the late twentieth century and during the present decade. These figures clearly indicate the increasing interest in the removal of C, R, and HQ by adsorption.

Removal by adsorption onto various adsorbents

Adsorption of organic solutes from the aqueous phase is a very important application of ACs, and it has been cited by the USEPA as one of the best available environmental control technologies [72]. The removal by adsorption onto various adsorbents of C, R, HQ, and their derivatives can be found in the literature [6,43-45, 52-57,68,70,73-75].

Activated carbon

Adsorption onto AC is widely used for wastewater treatment. Various adsorbents prepared from AC and low-cost adsorbents used in wastewater treatment were reported by Lin and Juang [76]. Thus, AC is used in the control of color and odors and in the removal of organic compounds or trihalomethane precursors, chlorine, and toxic compounds in general. A large amount of work has been devoted to the study of phenolic compound adsorption onto AC for water treatment purposes. The

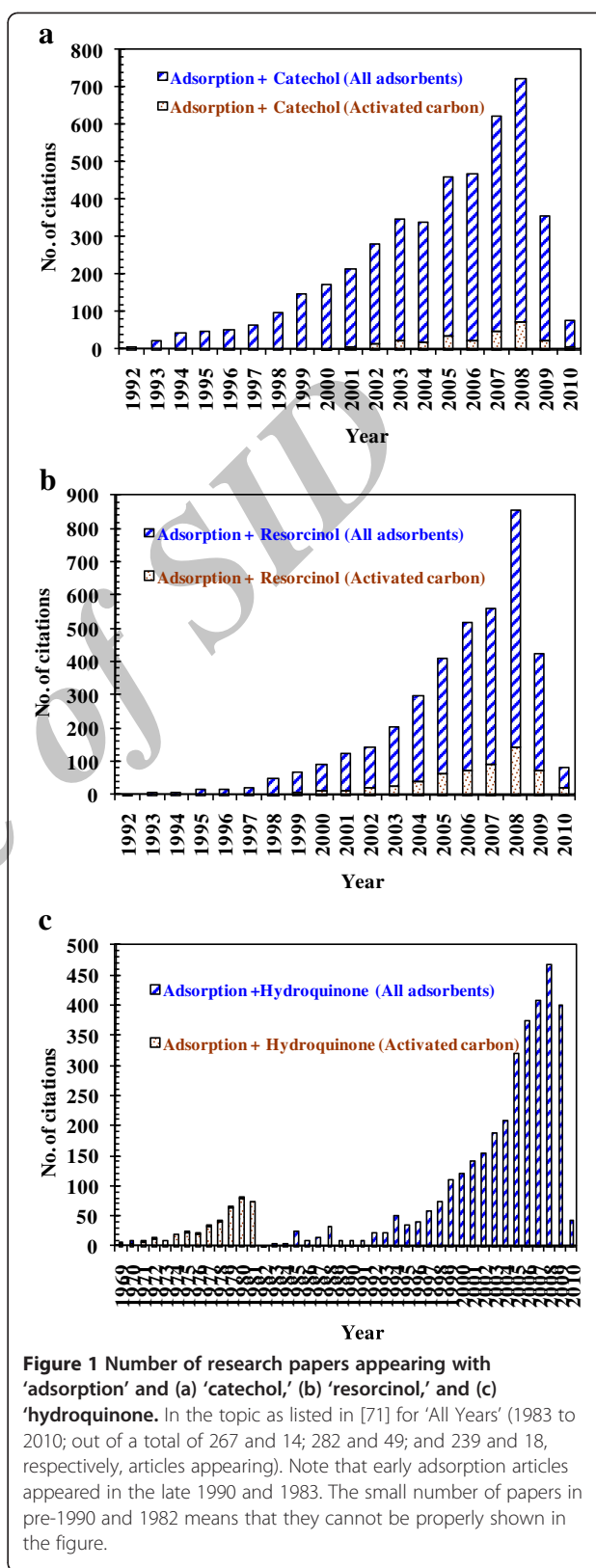
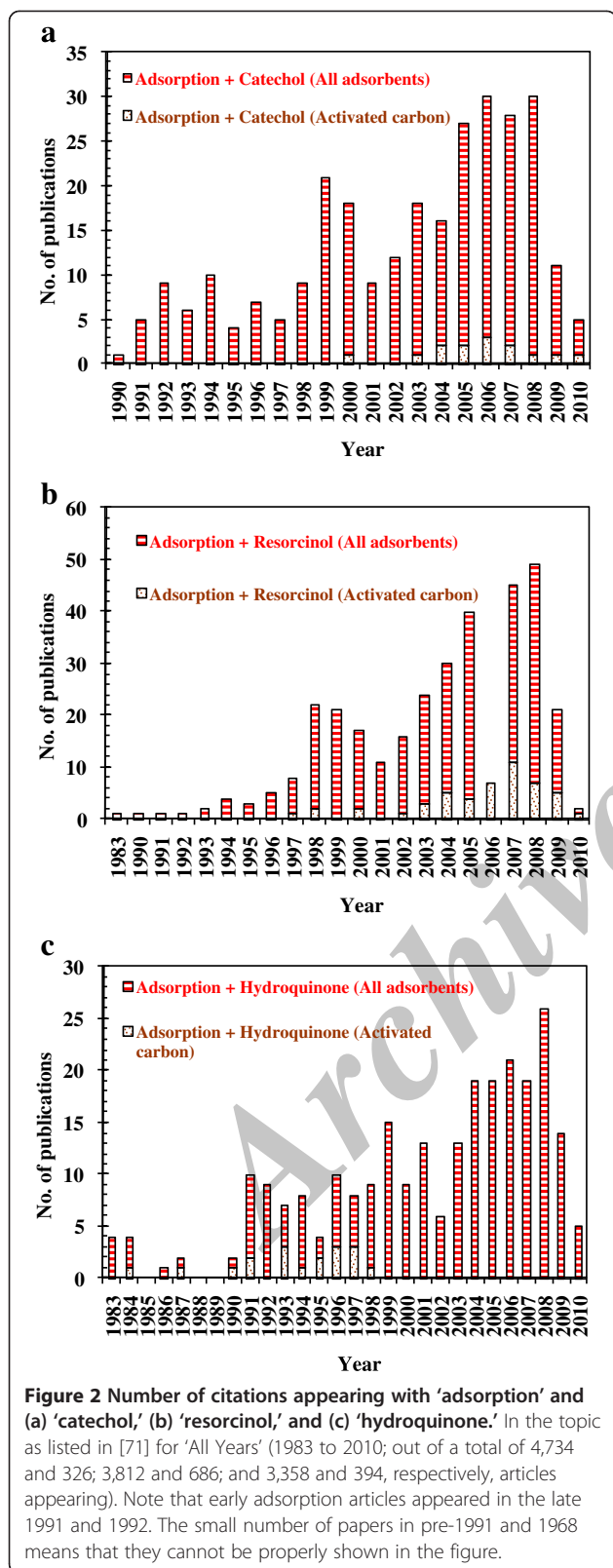


Figure 1 Number of research papers appearing with 'adsorption' and (a) 'catechol,' (b) 'resorcinol,' and (c) 'hydroquinone.' In the topic as listed in [71] for 'All Years' (1983 to 2010; out of a total of 267 and 14; 282 and 49; and 239 and 18, respectively, articles appearing). Note that early adsorption articles appeared in the late 1990 and 1983. The small number of papers in pre-1990 and 1982 means that they cannot be properly shown in the figure.



development of the investigation on the subject has been recently reviewed by Dabrowski [77]. Dobbs and Cohen [78] reported an extensive list of isotherm data of several toxic compounds. Ozkaya [79] compared different isotherm models used to describe the adsorption of phenols on AC. Garcia-Araya et al. [80] studied the adsorption of some phenolic acid on AC.

ACs present an outstanding adsorption capacity that stems from their high surface area, pore structure, and surface chemical properties. These materials are effective adsorbents for priority pollutants, therefore being suitable for the decontamination of water and wastewater. The AC surface is usually divided in three zones: basal planes, heterogeneous superficial groups (mainly oxygen-containing surface groups), and inorganic ash. For aromatic compounds, most of the adsorption sites are found on the basal planes [81]. However, heterogeneous groups have a higher activity and define the chemical characteristics of the carbon surface. The nature of the surface groups can be modified through physical, chemical, and electrochemical treatments. The most common are liquid phase treatments using HNO_3 and H_2O_2 , gas-phase oxidation with O_2 or N_2O , and heat treatment under inert gas to selectively remove some of the functional groups [42,81,82].

ACs possess excellent adsorption ability for relatively low molecular weight organic compounds such as phenols [82]. From a general point of view, an AC to be used in such processes must have adequate adsorptive capacity, mechanical strength, and chemical purity. Furthermore, all of these specifications should coexist with a low production cost. The most common materials used for the preparation of ACs are wood, coal, lignin, petroleum coke, and polymers [83]. Recently, many investigators have studied the feasibility of using many agricultural by-products and wastes, which are available at very little or no cost to prepare AC [84]. Both the texture and surface chemistry of ACs determine performance, and the final application of the carbon material will depend on its characteristics. The porous structure of ACs is a function of the precursor used in the preparation, the activation method followed, and the extent of activation. AC types produced by applying chemical activation processes contain mainly mesopores, while carbons obtained via gas activation are of the microporous type [85], although, depending on the nature of the parent material and by adjustment of the process conditions, different pore sizes that cover the micro-, meso-, and macropore ranges can be obtained [86]. Therefore, in recent years, it has prompted a growing research interest in the production of AC from renewable and cheaper precursors which are mainly agricultural by-products, such as corncob [87], rattan sawdust [88], rice straw [59,89], apricot shell [90], jute fiber [91], rubber wood sawdust [33,92],

bamboo [93], oil palm fiber [94], bagasse fly ash, coconut shell [59], and activated cashew shell nut [60].

Table 4 compares various parameters like adsorbent dosage (m), time (t), pH, etc. for the adsorption of C, R, HQ, and their derivatives onto GAC. Kumar et al. [6] investigated in a batch mode the adsorption behavior of R and C on GAC from a basic salt medium at optimum pH \approx 7.1, optimum temperature \approx 30°C, and optimum $m \approx$ 10 g/l for both R and C with 48 h as equilibrium time. However, Mohamed et al. [42] found 5 g/l as the optimum dosage of modified AC with 48 h as equilibrium time for the removal of C and R. Mondal and Balomajumder [68] observed adsorption and simultaneous adsorption biodegradation (SAB) for the removal of R and P from two separate synthetic wastewater over a 2- to 4-mm particle size at 28°C with 10 h as equilibrium time. They conducted the experiments over various adsorbent doses (2 to 12 g/l). They found that the removal of R reaches its constant value at the dose of around 8 g/l. Richard et al. [43] have reported that the adsorption of polyfunctional phenols encountered in olive oil mill wastewater was carried out onto AC at 20°C. They studied the effect of m in the range of 2 to 15 g/l and the effect of pH in the range of 5 to 9. They found that 16 g/l and 6.25 are the optimum dosage and pH, respectively, for the removal of C from olive oil mill wastewater. Huang et al. [55] reported the adsorption of C and HQ on modified GAC at pH 7, 9, and 11 with 48 h as equilibrium time. They observed that as the pH of the solution increases from 7 to 11, the amount of the phenols adsorbed diminishes. This indicates that adsorption increases when more protonated species are present in the solution. The adsorption isotherms show a linear behavior at $C_0 < 200$ mg/l, and later, the retained quantity increases slightly at pH 9. The adsorbed species are the protonated and monobasic anions, and due to the positive charge of the carbon surface, attractive and dispersive electrostatic forces begin to interact between the surface and the anion, favoring their accumulation. At pH 11, the surface is loaded negatively, the pH $>$ pH_{PZC} and the species present are the anionic monohydroxylated phenols. Therefore, the anionic species present in the solution repulse the surface AC. The adsorption occurs when the repulsive electrostatic forces predominate, and the adsorption at low concentrations is less than that at high concentrations [55].

Activated carbon cloth

Generally, utilization of AC can be in the form of powder, granule, and fiber or cloth. Activated carbon cloth (ACC), having very high specific surface area and adsorption capacity, uniform pore size, and mechanical strength, has attracted attention in recent years. Not only the adsorption but also the electrosorption

characteristics of ACC when acting as a working electrode material have been investigated on various adsorbates [94-97]. Table 5 compares various parameters like m , t , pH, etc. for the adsorption of C, R, HQ, and their derivatives onto ACC.

Bayram et al. [69] investigated the removal of C and R from aqueous solutions by adsorption and electrosorption onto high-area ACC with a dosage of 0.9 g/l, with 25°C and 24 h as operating temperature and contact time, respectively. They found that the extent of electrosorption of C was higher than R. It was attributed partly to a higher dipole moment of the former. It was demonstrated that ACC can partly be regenerated by an electrodesorption process. It was predicted that adsorption and electrosorption of C and R resulted mainly from dispersion interactions between surface charges and π electrons on adsorbate molecules. Ayranci and Duman [52] reported the adsorption of P, HQ, *m*-cresol, *p*-cresol, and *p*-nitrophenol from aqueous solutions onto ACC. The effect of ionization on the adsorption of these ionizable phenolic compounds was examined by studying the adsorption from acidic, basic, and natural pH (7.6) solutions at an operating temperature of 30°C.

Carbon nanotubes

Carbon nanotubes (CNTs), because of their chemical and physical properties, attract wide applications such as in the field of polymer composites [98], field emissions [93], energy storage [99], and sensors [100]. The large specific surface area of CNTs makes them suitable for the adsorption of gas [101], metal ions [102,103], and organic compounds [104]. Recent studies suggest that CNTs can serve as good adsorbent for a variety of gas and liquid molecules, like hydrogen, butane, dioxins, dichlorobenzene, and heavy metal ions [105-110]. The considerable attentions about CNTs depend on their unique structural and mechanical properties, high thermal stability, and large specific surface area [62,90,111].

Carbon nanomaterials include fullerenes, single-walled carbon nanotubes (SWCNTs), and multi-walled carbon nanotubes (MWCNTs). The monomer structure of fullerenes is a closed graphite ball, while those of CNTs are rolled-up graphite sheets forming a coaxial tube. A single rolled-up graphite sheet forms the SWCNT structure, while several rolled-up graphite sheets form the MWCNT structure. They consist of sheets of carbon atoms covalently bonded in hexagonal arrays that are seamlessly rolled into a hollow cylindrical shape [112]. The cylindrical surface geometry of CNTs has been well defined with outer diameters in the range of approximately 1 to 100 nm and length up to several tens of micrometers. Knowledge of environmental risk assessment of both toxic chemicals and CNTs once they are released to the environment [113-116] is also important.

Table 4 Pseudo-first- and second-order kinetic constants for adsorption of C, R, HQ, and their derivatives on various adsorbents

| Adsorbent | Adsorbate | pH | T (°C) | C ₀ (mg/l) | m (g/l) | t (h) | Pseudo-first-order constants | | | Pseudo-second-order constants | | References |
|------------------|----------------|---------|----------|-----------------------|--------------|-------|------------------------------|----------------|----------------|-------------------------------|----------------|------------------------------|
| | | | | | | | k _f | q _e | s ² | q _e | k _s | |
| | | | | | | | (min ⁻¹) | (mg/g) | (%) | (mg/g) | (g/mg min) | |
| Waste Fe(III) OH | C | 6.08 | 32 | 10 | 10 | 2.7 | 0.0198 | 0.42 to 2.11 | 0.993 | 0.015 to 0.931 | 0.16 to 2.73 | Namasivaya and Sumithra [54] |
| NDA-100 | C | - | 25 to 40 | 1,000 | 1 | 24 | 0.026 to 0.042 | - | 0.985 | - | - | Sun et al. [55] |
| AH-1 | C | - | 25 to 40 | 1,000 | 1 | 24 | 0.0238 to 0.0395 | - | 0.988 | - | - | Sun et al. [55] |
| GAC | C | 2 to 12 | 30 | 50 to 1,000 | 1 to 30 | 24 | 0.036 to 0.898 | 5.17 to 100.32 | 0.944 to 0.982 | 5.06 to 99.66 | 0.024 to 2.60 | Suresh et al. [48] |
| | | (6) | | | (10) | | | | | | | |
| ACC | HQ | 7.4 | 30 | - | 0.15 | 1.3 | 0.0323 | - | 0.988 | - | - | Ayranci and Duman [53] |
| GAC | HQ | 2 to 12 | 30 | 50 to 1,000 | 1 to 30 (10) | 24 | 0.011 to 0.224 | 4.95 to 49.8 | 0.930 to 0.988 | 4.92 to 104.5 | 0.018 to 9.03 | Suresh et al. [49] |
| | | (6.1) | | | | | | | | | | |
| HJ-1 | C | - | 20 | 100 | 6 | 7.5 | 0.002 to 0.007 | - | - | - | 0.01 to 0.027 | Huang et al. [56] |
| CTAB-B | C | 5 to 12 | 30 | 88 to 330 | 5 | 4.16 | 0.1858 to 0.7359 | - | 0.988 | - | - | Shakir et al. [59] |
| | | (≥9.9) | | | | | | | | | | |
| ACC | C | 6.44 | 25 | - | 0.9 | 24 | 0.0056 | 0.004 | - | 0.006 | 2.43 | Bayram et al. [69] |
| ACC | C ^a | 6.44 | 25 | - | 0.9 | 24 | 0.0094 | 0.007 | - | 0.007 | 2.26 | Bayram et al. [69] |
| NDA-100 | R | - | 25 to 40 | 1,000 | 1 | 24 | 0.034 to 0.053 | - | 0.985 to 0.989 | - | - | Sun et al. [55] |
| AH-1 | R | - | 25 to 40 | 1,000 | 1 | 24 | 0.030 to 0.049 | - | 0.991 to 0.995 | - | - | Sun et al. [55] |
| HJ-1 | R | - | 20 | 100 | 6 | 24 | 0.0078 to 0.0249 | - | - | - | - | Huang et al. [56] |
| ACC | R | 6.55 | 25 | - | 0.9 | 7.5 | 0.0023 | 0.0024 | - | 0.0057 | 2.81 | Bayram et al. [69] |
| ACC | R ^a | 6.55 | 25 | - | 0.9 | 24 | 0.0069 | 0.0042 | - | 0.0064 | 2.11 | Bayram et al. [69] |
| GAC | R | 2 to 12 | 30 | 50 to 1,000 | 1 to 30 | 24 | 0.036 to 0.773 | 4.84 to 113.6 | 0.933 to 0.996 | 4.73 to 112.2 | 0.285 to 4.79 | Suresh et al. [48] |
| | | (6) | | | (10) | | | | | | | |

^aObtained from electrosorption data; intraparticle diffusion coefficients D_e (m²/s) for C and R are 2.33 to 6.14×10^{-13} and 3.19 to 3.96×10^{-13} , respectively [6]; NDA-100, hypercrosslinked resin without amino groups; AH-1, aminated hypercrosslinked polymers; GAC, granular activated carbon; ACC, activated carbon cloth; HJ-1, hypercrosslinked resin; CTAB-B, cetyltrimethylammonium bromide-modified bentonite; C, catechol; R, resorcinol; HQ, hydroquinone.

Table 5 Freundlich, Langmuir, and other constants for adsorption of C, R, HQ, and their derivatives on various adsorbents

| Adsorbent | Adsorbate | t (h) | K_F | 1/n | q_m | K_L | References |
|---|-----------|-------|------------------------------|---------------------|------------------|----------------|------------------------------|
| | | | (mg/g)/(mg/l) ^{1/n} | | (mg/g) | (l/mg) | |
| GAC | C | 48 | 42.40 | 0.205 | 143.47 | 0.08 | Kumar et al. [6] |
| Waste Fe(III) OH | C | 2.33 | 0.39 to 0.62 | 0.681 | 4.04 | 0.12 | Namasivaya and Sumithra [53] |
| NDA-100 | C | 24 | 0.51 | 0.493 | 111.43 | - | Sun et al. [54] |
| ACC | HQ | 24 | 350.9 | 0.371 | 204.6 | 0.0028 | Ayranci and Duman [52] |
| CTAB-B | HQ | - | - | - | 17 to 22 | 0.007 to 0.009 | Yildiz et al. [51] |
| GAC | HQ | 24 | 2.59 to 2.69 | 0.43 to 0.62 | 102.3 to 135.3 | 19.8 to 36.3 | Suresh et al. [46] |
| AH-1 | C | 24 | 1.015 | 0.308 | 171.33 | - | Sun et al. [54] |
| MWCNT | C | 10 | 16.49 | 0.411 | 14.20 | 0.904 | Liao et al. [70] |
| CTAB-B | C | 4.16 | 0.30 | 0.305, 0.299, 0.277 | 56.83 | 0.002 | Shakir et al. [58] |
| CTAB-B | C | 4.16 | 33.02 | - | 53.41 | 0.003 | Shakir et al. [58] |
| CTAB-B | C | 4.16 | 0.29 | - | 49.78 | 0.031 | Shakir et al. [58] |
| AC | C | 48 | - | 0.134 | 320 | 0.095 | Richard et al. [43] |
| ACC | C | 24 | 0.015 | 0.23 | 0.0013 | 0.000007 | Bayram et al. [69] |
| CZ | C | 48 | - | - | 2.202 | - | Mohamed et al. [42] |
| TiO ₂ | C | 2 | - | 0.337 | 11.172 | 0.0589 | Arana et al. [57] |
| TiO ₂ -H ₂ O ₂ | C | 2 | - | 0.38 | 11.140 | 0.0150 | Arana et al. [57] |
| HJ-1 | C | 7.5 | 1.40 to 2.55 | 0.574 to 0.630 | 125 to 133 | 0.003 to 0.004 | Huang et al. [55] |
| GAC | C | 48 | 1.49 | 0.81 | 238.1 | 0.004 | Blanco-Martinez et al. [44] |
| GAC | C | 24 | 0.59 to 1.12 | 52.8 to 62.7 | 1.09 to 1.41 | 492.8 to 749.1 | Suresh et al. [46] |
| GAC | R | 48 | 34.83 | 0.226 | 142.8 | 0.047 | Kumar et al. [6] |
| CZ | R | 48 | - | - | 0.880 | - | Mohamed et al. [42] |
| GAC | R | 10 | 34.52 | 0.236 | 140.72 | 0.046 | Mondal and Balomajumder [68] |
| GAC-SAB | R | 10 | 23.43 | 0.877 | 49.75 | 0.074 | Mondal and Balomajumder [68] |
| TiO ₂ | R | 2 | - | 0.703 | 7.870 | 0.0089 | Arana et al. [57] |
| TiO ₂ -H ₂ O ₂ | R | 2 | - | 0.781 | 7.605 | 0.0083 | Arana et al. [57] |
| ACC | R | 24 | 0.005 to 0.014 | 0.23 to 0.25 | 0.0013 to 0.0021 | 0.046 to 0.143 | Bayram et al. [69] |
| HJ-1 | R | 7.5 | 1.44 to 2.65 | 0.55 to 0.61 | 128.2 to 129.9 | 0.002 to 0.003 | Huang et al. [55] |
| GAC | R | 24 | 0.49 to 0.93 | 57.2 to 108.9 | 1.21 to 1.27 | 229.9 to 391.6 | Suresh et al. [46] |

GAC, granular activated carbon; NDA-100, hypercrosslinked resin without amino groups; ACC, activated carbon cloth; CTAB-B, cetyltrimethylammonium bromide-modified bentonite; AC, activated carbon; AH-1, aminated hypercrosslinked polymers; MWCNT, multi-walled carbon nanotube; HJ-1, hypercrosslinked resin; SAB, simultaneous adsorption biodegradation; C, catechol; R, resorcinol; HQ, hydroquinone.

Table 5 compares various parameters like m , t , pH, etc. for the adsorption of C, R, HQ, and their derivatives onto GAC by CNTs.

Liao et al. [70] studied the adsorption of P, C, R, HQ, and pyrogallol onto untreated MWCNTs and HNO₃-treated MWCNTs. The uptake of R was 19.7 mg/ml (60% of total amounts adsorbed) within 1 min, when the total contact time was 660 min. Liao et al. [70] have suggested that the adsorption mechanism of MWCNTs is not totally the same as that of AC. The adsorption onto AC is dependent on the porous structure, so it takes time for adsorbates to diffuse through pores [117]. It is

confirmed by the adsorption of N₂ to CNTs that most available spaces of CNTs for adsorption are the cylindrical external surfaces, neither the inner cavities nor the inner-wall spacings [116].

Liao et al. [70] observed a pH < 6. There is a slight increase in the uptake of R with the decrease of pH because the solubility of R is dependent on pH; due to the weak acidity of R, the solubility of R decreases with the decrease of pH. The adsorption onto AC suggested that the uptake of phenolic derivatives was inversely proportional to solubility [118]; thus, the uptake of R on MWCNTs increased with the decrease of pH. Lin and

Xing [75] have conducted experiments for the adsorption of P, C, and pyrogallol onto CNTs over a range of pH. It was found that the final solution pH (4.0 to 6.5) was lower than the pK_a of the phenolics and close to the point of zero charge (pH_{PZC}) of CNTs, which suggested that phenolics used for adsorption should be primarily in neutral form. Therefore, electrostatic interaction might not greatly influence the sorption of phenolics to CNTs in the weakly acidic environment. Sorption was found to increase sharply from P to C and then to pyrogallol, while their hydrophobicity decreased. Thus, the hydrophobic interaction can be insignificant as a major influencing factor regulating the sorption of these phenolics to CNTs.

Liao et al. [70] found that the acid-treated MWCNTs showed decreased adsorption capacity for R compared to untreated MWCNTs because of the increased electrostatic repulsion between carboxylic groups weakening the π - π interaction and water adsorption. The uptake of the adsorbates increased with the increase in the number of hydroxyl groups and their location in *meta*-position on the aromatic ring. Four possible solute-sorbent interactions are possible for the adsorption of phenolics onto CNTs as follows: (a) hydrophobic interaction, (b) electrostatic attraction or repulsion, (c) hydrogen bonding between the -OH and the tube surface -OH or -COOH groups, and (d) -OH substitution-enhanced π - π interactions between the phenolics and the CNTs. The -OH substitution on the phenolics and the hydroxy/carboxylic groups on the CNT surface may form hydrogen bonds; the hydrogen bonds may also form between the surface-adsorbed and dissolved phenolics. However, very low hydrogen and oxygen contents were detected on the CNTs, indicating that hydrogen bonding (if any) might not be significant between the phenolics and the functional groups on the CNTs [70,119,120]. An insignificant effect of hydrogen bonding on the sorption of nitroaromatics to CNTs was also recently reported [121], and hydrogen bond formation may have favorable adsorption at high concentration.

Polymeric resins

Adsorption onto polymeric resins has been widely studied for the treatment of effluents containing phenolic compounds [54,122]. In comparison to classical adsorbents such as silica gel, alumina, and AC, polymeric adsorbents have high chemical stability, easy regeneration ability, excellent selectivity, and longevity. Among them, the commercial resin Amberlite XAD-4 has been considered as one of the best for the removal of phenolic compounds from wastewater [54,123]. Many researchers have made more efforts on the chemical modification of polymeric adsorbents with functional groups such as phenolic hydroxyl, acetyl, benzoyl, and hypercrosslinked

polymers [124,125] to improve their adsorption properties by increasing the interactions between adsorbates and adsorbents [54,55,126,127]. Styrene-divinylbenzene matrix has also been used for the removal of hydrophobic organic pollutants [128-130]. Table 4 compares various parameters like m , t , pH, etc. for the adsorption of C, R, HQ, and their derivatives onto various polymeric resins.

Adsorption of C and R from an aqueous solution onto AH-1, AH-2, AH-3, NDA-100, and HJ-1 resins was reported by Sun et al. [54] and Huang et al. [55]. Sun et al. [54] found that the q_m of C and R onto aminated hypercrosslinked resins AH-1, AH-2, and AH-3 are higher than that onto hypercrosslinked resin NDA-100, and more time was required to reach the equilibrium for higher C_o and the required time for the adsorption of R is shorter than that of C [54,55]. Thus, effects of the solubility and position of the hydroxyl group at the *ortho*-position may probably account for higher adsorbability of C than R onto HJ-1. Specific surface area, micropore structure, and content of tertiary amino groups play a combined role during the adsorption of both compounds onto the aminated hypercrosslinked polymers [54,55]. Generally, the removal of these compounds mainly results from van der Waals interaction between the solute and the sorbent phase [131]. However, many hydrophilic or water-soluble organic compounds cannot be readily removed by those sorbents partly due to the strong solute-water interaction [130].

Organoclays

Over the last few decades, organoclays have gained much importance in the removal of organic pollutants from aqueous solutions. Among the various types of clays, montmorillonite, which is the main constituent of the low-cost and naturally abundant mineral bentonite, possesses several properties that make it very appropriate for organoclay preparation. Montmorillonite has a 2:1 type of layer consisting of one octahedral sheet of alumina inserted in between two silica tetrahedral sheets. In the tetrahedral sheets, Al^{3+} can substitute for Si^{4+} , and in the octahedral sheets, Mg^{2+} or Zn^{2+} can replace Al^{3+} . These isomorphous substitutions in the clay lattice result into a net negative charge on the clay surface [132]. Montmorillonite, with its lower surface charge density, has substitutions mostly in the octahedral sheet [133].

The adsorption properties of the organoclay surfaces may be significantly altered by exchange reactions, thus making the clay more organophilic in nature, and this increases its capability to remove organic pollutants from aqueous solutions [134-136]. Hydrophilic clays can be changed to organophilic clays through organic chemicals [137]. The modified organoclays have acted as a

partition medium in the sorption of organic pollutants [138-141]. Table 4 compares various parameters like m , t , pH, etc. for the adsorption of C, R, HQ, and their derivatives onto organoclays.

The removal of C from aqueous solutions onto cetyltrimethylammonium bromide-modified bentonite (CTAB-B) surfaces was reported by Shakir et al. [58]. They carried out the experiment with a wide range of pH (5 to 12), contact time (1 to 250 min), and concentration (0.8 to 15.3 mmol/l) at $30 \pm 1^\circ\text{C}$. The percentage adsorption is $\leq 11\%$ at $\text{pH} \leq 7.5$, whereas it increases sharply with pH, attaining about 100% at $\text{pH} \geq 9.9$. However, the sorption capacities increase with decreasing pH. Yildiz et al. [51] reported the removal of HQ and benzoic acid on synthesized organobentonites (ODTMA-B, HDTMA-B). Zhu et al. [134] and Smith and Grum [135] indicated that the magnitude and mechanism of sorption are functions of the cation-exchange capacity of the clay, the molecular structure of the exchanged organic cation, the extent of cation exchange, and the molecular structure of the solute. Sorption may take place either by partition or by adsorption depending mainly on the characteristics of the exchanged organic cation and the molecular structure of the solute. Adsorption of the large cationic surfactant molecule, CTAB, greatly modifies the nature of the clay surface which may exhibit both hydrophilic and hydrophobic as well as electrostatic properties and van der Waals interaction between the -R group of the surfactant and adsorbate [136-142], and it is also observed that the percentage adsorption increases from 81.5% to approximately 100% as the initial adsorbate concentration is reduced from the initial concentration of 88 mg/l. This observed increase in the percentage adsorption is due to the availability of larger sorbent surface sites for a relatively smaller amount of C at lower C_0 [58].

Metal surface

TiO₂

The adsorption of organic compounds onto oxides, hydroxides, and other minerals is known to retard their migration in soils and aquifers and to alter their susceptibility toward chemical and biological transformations [143]. Few literatures on the adsorption of organic compounds with hydroxyl or amino groups as the sole ligand donor groups are available [144]. It has been indicated that certain adsorbates can interact with active centers such as hydroxyl groups or bridging oxygen on TiO₂ surface, resulting in a different catalytic activity [145]. Also, some of them can act as poisons [146]. Table 4 compares various parameters like m , t , pH, etc. for the adsorption of C, R, HQ, and their derivatives onto TiO₂ surface.

The adsorption removal of phenols was 90% at 1 g/l onto TiO₂ surface. 4-Nitrocatechol adsorbs to a

significantly greater extent than 4-nitro-2-aminophenol over the entire pH range at 1 g/l TiO₂ dosage [56]. When $\text{pH} < \text{pH}_{\text{PZC}}$ of the surface, positive surface charge increases with decreasing pH, and when $\text{pH} > \text{pH}_{\text{PZC}}$, negative surface charge increases with increasing pH [56]. Generally, the adsorption behavior of a number of organic compounds possessing carboxylic acid groups and the effects of pH have been successfully modeled by choosing appropriate stoichiometries and equilibrium constants for adsorption [147,148].

Waste Fe(III)/Cr(III) OH. Adsorption has gained wide acceptance and popularity for the removal of phenolic compounds including industrial solid wastes [149]. Chromium(VI) compounds are used as corrosion inhibitors in cooling water systems in industries. Fe(II), which is generated electrolytically, reduces chromium(VI) in wastewater to Cr(III) under acidic conditions [150]. The Fe(III)/Cr(III) ions, produced in solution, are precipitated as Fe(III)/Cr(III) hydroxide by the use of lime. The resultant sludge is discarded as waste by the industries. Waste Fe(III)/Cr(III) hydroxide has been investigated in the laboratory for the removal of heavy metals [151], dyes, and pesticides [152]. The percentage C removal at equilibrium decreased from 80% to 65% as the C_0 increased from 10 to 40 mg/l onto 'waste' Fe(III)/Cr(III) hydroxide [53].

Which (C, R, HQ, and their derivatives) gets more adsorbed by different adsorbents?

Kumar et al. [6] found that C was adsorbed more than R, suggesting that a lesser quantity of AC would be required to remove the same amount of C as compared to R in the individual compounds. The results may differ when both R and C are simultaneously present in solution [6]. Mohamed et al. [42] found that for various carbons, the amount of adsorbate adsorbed follows the order $\text{P} > \text{HQ} > \text{R} > \text{C}$. Mondal and Balomajumder [68] concluded that the efficiency of SAB is more than that of adsorption and the percentage removal of P is more than that of R for both adsorption and SAB because of the increase of specific surface area as well as micropore volume in the SAB system. Kumar et al. [6] concluded that C is adsorbed to a greater extent than R because of the difference in adsorbability which can be explained in terms of the compound's solubility, pH, density, and the presence and position of the hydroxyl group on the aromatic benzene ring [63]. Liao et al. [70] have suggested that the uptake of R and other phenols increased with the increasing number of hydroxyl and its location in *meta*-position on the aromatic ring, which resulted in the highest adsorptive capacity. The solubility of R in water is higher than that of C [153]; thus, it has more affinity towards water, i.e., hydrophilic. A similar result was shown by Sun et al. [54] on different polymer resins

as adsorbents. C adsorption is much higher than those of the other phenolics, and its interaction occurs preferentially through the formation of a catecholate monodentate. R and the cresols interact by means of hydrogen bonds through the hydroxyl group, and their adsorption is much lower than that of C [57]. According to the Lewis acid–base theory, the benzene ring of the aromatic-based resin and the tertiary amino groups on it can be viewed as Lewis bases, while phenolic compounds can be viewed as Lewis acids [134]. Therefore, the Lewis acid–base interaction may occur between phenolic compounds and the benzene ring as well as the tertiary amino group of the resins, which leads to the formation of a hydrogen-bonded complex. In addition, the matching of polarity between adsorbent and adsorbate is also an important factor affecting adsorption of phenolic compounds. The tertiary amino nitrogen on the resins has a large dipole moment, and the dipole moment of C is larger than that of R (2.620 and 2.071 D, respectively [64,65,154]); therefore, the interaction between the resins and C is expected to be stronger than that between the resins and R. Furthermore, some researches revealed that the same functional group but at the *ortho*-position greatly enhances the adsorption energy of C [6]. These may be possible reasons for its lower adsorbability. Kumar et al. [6] have also observed that the same functional group but at the *ortho*-position greatly enhances the adsorption energies of these compounds, such as C. However, R was adsorbed more compared to C which may refer to the geometry and molecular size of the adsorbate molecule [42]. The changes induced in the texture of the prepared ACs as a result of H₂SO₄ activation do not seem to have a significant role in the adsorption of phenols (P, C, R, and HQ), although activation with sulfuric acid leads to a continuous increase in both micropore volume and surface area [42].

Kinetics study

The study of the adsorption equilibrium and kinetics is essential to supply the basic information required for the design and operation of adsorption equipment [59]. A mass transfer occurs during the adsorption process; the first step is the solute transfer through the adsorbent external surface film, and the others are the solute fluid diffusion into the pore holes and the adsorbed molecules' migration along the pore surfaces, if it takes place. The former is characterized by the external mass transfer coefficient, and the last ones, by the internal pore and surface diffusivities. Available bulk adsorbate concentration in the liquid phase and adsorbed solute concentration in the solid phase are considered time-dependent.

Various kinetic models, namely pseudo-first-order, pseudo-second-order, and intraparticle diffusion models, have been used to test their validity with the experimental

adsorption data for C, R, and HQ onto GAC and other adsorbents. The most commonly used kinetic expressions to explain the solid/liquid adsorption processes are the pseudo-first-order and pseudo-second-order kinetic models [59,155]. The pseudo-second-order expression as proposed by Ho [156] and Srivastava et al. [59] was found to explain the kinetics of most of sorption systems very well for the entire range of sorption period. Weber and Morris had presented an intraparticle diffusion model in 1962 [47,59,157]. Recently, the intraparticle diffusion model was reviewed by Wu et al. [158]. Various models are reported for the removal of C, R, and HQ onto various adsorbents [6,43-45,52-57,68,70,73-75]. Table 5 shows a list of the first- and second-order kinetic constants for the adsorption of C, R, HQ, and their derivatives on various adsorbents.

The first-order kinetic model was fitted for the adsorption of C and R onto different polymers and ACC over a period of 80 and 90 min, respectively, at natural pH [52,54], and it was found by Sun et al. that the k_1 value of R was higher than that of C [54]. Similar results were found by Ayranci and Duman [52] in the rate constants which decreased in the order *p*-nitrophenol > *m*-cresol > *p*-cresol > HQ > P onto ACC. Various researchers have proposed and fitted the first-order kinetic model for the removal of C, R, and other phenols onto different adsorbents [54,58,94,95]. The adsorption and electrosorption of C and R onto ACC follows the pseudo-second-order model for adsorption periods of 1,042 and 1,508 min, and the electrosorption of C was higher than that of R which was attributed partly to the higher dipole moment of the former [69]. The pseudo-second-order model rather than the pseudo-first-order model was fitted by Bayram et al. [69] because of smaller error and the R^2 value was close to 1. Namasivayam and Sumithra [53] and Huang et al. [55] have reported an adsorption dynamic curve which followed the second-order rate kinetics onto metal surface and HJ-1 resin.

Isotherm and thermodynamic study

Adsorption equilibrium measurements are used to determine the maximum or ultimate adsorbed capacity. Adsorption equilibrium is established when the amount of solute being adsorbed onto the adsorbent surface is equal to that being desorbed from the surface to the bulk fluid [59,159]. At this point, the equilibrium solution concentration remains constant. Adsorption equilibrium data are formulated into an isotherm model. Six types of adsorption isotherms exist including types I to VI [59,160].

The various adsorption isotherms were reviewed [161]. Equilibrium isotherms are measured to determine the capacity of the adsorbent for the adsorbate. The equilibrium of adsorption processes is usually dependent on

the amounts adsorbed on the fluid phase composition for a constant temperature [162]. To design and optimize separations using adsorption isotherms, knowledge on the powerful concept adsorption isotherms is mandatory [163-167].

An increase in temperature increases the chemical potential of the organic molecules to penetrate through the surface pores of GAC. Also, the mobility of the adsorbate increases with an increase in temperature. Together, these result in the enhancement in the adsorptive capacity of the GAC at higher temperature. An increase in the phenol adsorption capacity of the carbonaceous adsorbents with an increase in temperature has also been reported by other investigators [59,168,169]. The investigators have ascribed different reasons for the endothermic nature of the adsorption of phenolics onto ACs. A rise in adsorption temperature weakens hydrogen bonds formed among water molecules and between water molecules and the solute or the adsorbent [170] and enhances pore diffusion [59,80,171]. Therefore, an increase in temperature favors the dehydration of adsorbate molecules, which makes them more planar and gives them a larger dipolar moment. An increase in planarity provides solute molecules a greater access to the pores of the GAC, while an increase in dipolar moment results in enhanced adsorbent-adsorbate interactions. As a result, the adsorption is found to be endothermic because of the endothermicity of dehydration adsorbate molecules. Therefore, the adsorptive uptake increases with an increase in temperature.

Adsorption isotherm modeling

Various isotherm equations like those of Freundlich, Langmuir, Temkin, Dubinin-Radushkevich, and Redlich-Peterson have been used in the literature to describe the equilibrium characteristics of adsorption for the removal of C, R, HQ, and their derivatives onto different adsorbents. Some researchers already explained the theory, assumption, and equation or models for the removal of phenolic compounds [43,59].

The R^2 values alone are not sufficient in determining the best isotherm model to represent the experimental data because they are generally found to be >0.91 for all the four models. Table 5 lists the Freundlich, Langmuir, and other constants for the adsorption of C, R, HQ, and their derivatives on various adsorbents.

The most important parameter to compare in Table 5 is the q_{\max} value of the Langmuir model because this measures the adsorption capacity of AC and others for the adsorbates. By comparing the results for the adsorption capacity of AC for C, R, or HQ, it is seen that the q_{\max} value is clearly greater for C at higher temperature (45°C), whereas it is greater for R at lower temperatures (15°C and 30°C).

The adsorption capacity as measured by K_F of the Freundlich model was best fitted for R and C onto GAC as compared to other models [6,69]. The Freundlich model is more suitable for characterizing adsorption than the Langmuir model [55], indicating that the adsorption may be a multilayer process and the adsorbent possesses a heterogeneous nature [119]. However, the adsorption was not a pure monolayer type, and the mixed model was expected for the isotherms of R and HQ as reported by Liao et al. [70].

There were higher adsorption constant (K_L) and average adsorption energy for the adsorption of C than those of R and other phenolic compounds onto Degussa P-25 TiO₂. The Langmuir model was best fitted for the adsorption of C and other phenolic compounds having K_L which was much larger than those of P and R. However, the Langmuir equation is valid up to a concentration limit, which is a characteristic of each phenolic compound [53,57,172].

The adsorption isotherms of C, R, HQ, and pyrogallol have been well fitted by both the Freundlich and Langmuir models [58,70], which means that physical adsorption occurred [173] and the adsorbed amount of phenolic derivatives follows the order $R \gg C$ onto MWCNTs [70]. Lu et al. [102] indicated that the adsorption capacity of AC was closely related with the molecular weight and boiling point of the compound. The difference in boiling point of the three phenolic derivatives (HQ, 285.2°C > R, 281°C > C, 245°C) might be the reason for the lowest adsorbability of C. However, the uptake of R is the highest, which might be due to other complex factors and need further exploration. The adsorption capacity of MWCNTs was more than that of the AC treated by Mohamed et al. [42], even though the specific surface area of AC (SSA = 210 m²/g) was much larger than that of MWCNTs (SSA = 72 m²/g) in our experiments.

Adsorption isotherms of pyrogallol, C, and P show that the effect of -OH substitution on sorption was not proportional to the number of -OH groups. The higher K_D ratios of pyrogallol/C than C/P at high equilibrium concentrations were due to the possible greater hydrogen-bonding interaction between the surface-adsorbed pyrogallol and the ones dissolved in water; the hydrogen bonding interaction may not be significant between the surface-adsorbed and dissolved C or P molecules due to fewer -OH groups on the molecules. Multilayer sorption of pyrogallol, but not C and P, on the CNTs could be the evidence for the sorption mechanism difference between these phenolics at high concentrations [75].

Adsorption capacity of C was higher than that of R [52,55,69] due to its smaller molecular size. The value of ΔH for C was more negative than that for R which can

be explained in terms of the solubility and the polarity of the two adsorbates [55]. However, the adsorption capacity decreases with increase in temperature [54]. The Langmuir isotherm was best fitted for C, and the Freundlich isotherm, for the other compounds [43]. The adsorption behavior of phenolic compounds on the same kind of AC was also studied [42], and it was also concluded that the adsorption capacities are of the same order of magnitude, but the other parameters appear to be strongly dependent upon the chemical groups around the aromatic ring. However, acidic phenols exhibit a more progressive isotherm than the non-acidic species [42,43].

Adsorption thermodynamics

The Gibbs free energy change (ΔG_0) of the adsorption process is related to the equilibrium constant (K_D) by the classical Van't Hoff equation and also related to the entropy change (ΔS_0) and heat of adsorption (ΔH_0) at constant temperature. The equilibrium adsorption constant (K_D) can be related to as follows:

$$\ln K_D = \frac{-\Delta G_0}{RT} = \frac{\Delta S_0}{RT} - \frac{\Delta H_0}{R} \frac{1}{T} \quad (1)$$

where T is the absolute temperature (K), R is the universal gas constant (8.314×10^{-3} kJ/mol K), and K_D ($=q_e/C_e$) is the single point or linear sorption distribution coefficient. Thus, ΔH_0 , which is the enthalpy change (kJ/mol), can be determined from the slope of the linear Van't Hoff plot, i.e., $\ln K_D$ versus $(1/T)$. This ΔH_0 corresponds to the isosteric heat of adsorption ($\Delta H_{st,0}$) with zero surface coverage (i.e., $q_e = 0$) [174]. K_D at $q_e = 0$ can be obtained from the intercept of the $\ln q_e/C_e$ versus q_e plot [175]. The adsorption ΔH_0 , ΔS_0 , and ΔG_0 corresponding to different percentages of the adsorbent surface, adsorption free energies, and adsorption entropies are reviewed in Table 6.

Generally, the ΔG_0 value is in the range of -20 to 0 kJ/mol for physisorption and in the range of -400 to -80 kJ/mol for chemisorption [176]. ΔG_0 values were negative, indicating that the adsorption process led to a decrease in ΔG_0 and that the adsorption process is feasible and spontaneous [59,177]. The ΔG_0 values achieved at four different temperatures are negative, indicating a spontaneous physisorption process [58]. The ΔH_0 of the adsorption of organic molecules from an aqueous solution onto AC is usually within the range of 8 to 65 kJ/mol [178]. The positive values of ΔH_0 indicate the endothermic nature of the adsorption process. The ΔH_0 value decreases with increase in the percentage of the adsorbent surface which resulted from the energetic heterogeneity of the adsorbent surface [119]. In physisorption, the bond between adsorbent and adsorbate is the van der Waals interaction, and ΔH_0 is typically in the

range of 5 to 10 kJ/mol for liquid-phase adsorption. In the case of chemisorption, a chemical bond is formed between adsorbate molecules and the surface, and the chemisorption energy is, generally, in the range of 30 to 70 kJ/mol [179]. The value of ΔH_0 of C was little more negative than that of R at the same percentage of the adsorbent surface [55]. The initial adsorption enthalpy of C is greater than that of R, displaying that the interaction of the adsorbent with C is a little stronger [54]. The negative adsorption free energies imply a favorable and spontaneous process, and the adsorption free energy is independent on the occupancy percentage of the adsorbent surface. The ΔH_0 indicates exothermic adsorption, and the positive value of ΔS_0 , indicates increased randomness at the solid/solution interface with some structural changes in the adsorbates and the adsorbents [57]. The positive ΔS_0 value also corresponds to an increase in the degree of freedom of the adsorbed species [180]. The negative value of ΔS_0 reveals that a more ordered arrangement of the adsorbates is shaped on the adsorbent surface and also a weaker activity of adsorbate molecules on the adsorbents than on the aqueous solution [54].

Conclusions

Chemical contamination of water from a wide range of toxic compounds, in particular aromatic molecules, is a serious environmental problem owing to their potential human toxicity especially C, R, HQ, and their derivatives which appear to be the major organic pollutants globally. They, derived from industrial effluent discharges, present an ongoing and serious threat to human health and to natural water. Most of the researchers concluded that there is higher adsorbability of C than R and HQ because of the effects of solubility and the position of the ($-OH$) functional group at the *ortho*-position.

The role of AC and other adsorbents in the removal of C, R, HQ, and their derivatives from water and wastewater was discussed. However, it is only able to remove few milligrams of phenols per gram of AC, and there are still some problems encountered in the regeneration process. This makes AC an expensive adsorbent for this purpose. The solid waste can be converted into low-cost adsorbents for the treatment of discharged wastewater; the cost of removal might decrease. Although the organoclays revealed good adsorption capability, they were still non-economic. The ability and efficiency of the adsorption technologies in water treatment depend on the characteristics and functions of adsorbents. Therefore, to transfer the pollutants in promoting the adsorption rate, we can design and prepare some special composite adsorbents with good adsorptive functions.

Table 6 Thermodynamic properties for adsorption of C, R, HQ, and their derivatives on various adsorbents

| Adsorbent | Adsorbate | Temperature (°C) | ΔH_0 | ΔG_0 | ΔS_0 | References | | |
|--------------------------|----------------|------------------|--------------|----------------------|------------------|------------------------------|--------|--------------------|
| | | | (kJ/mol) | (kJ/mol) | (J/mol K) | | | |
| Waste Fe(III) Cr(III) OH | C | 32 to 60 | 15.43 | | 63.82 | Namasivaya and Sumithra [53] | | |
| AH-1 | C | 10 | -33.76 | -7.64 | -92.30 | Sun et al. [54] | | |
| | | 25 | -33.76 | -7.71 | -87.42 | | | |
| | | 40 | -33.76 | -6.96 | -85.62 | | | |
| AH-2 | C | 10 | -33.26 | -7.90 | -89.61 | Sun et al. [54] | | |
| | | 25 | -33.26 | -7.93 | -85.00 | | | |
| | | 40 | -33.26 | -7.60 | -81.98 | | | |
| CTAB-B | C | 30 | -7.3 ± 0.148 | -12.446 ± 0.094 | 16.985 ± 0.467 | Shakir et al. [58] | | |
| | | 40 | -7.3 ± 0.148 | -12.616 ± 0.196 | 16.985 ± 0.467 | | | |
| | | 50 | -7.3 ± 0.148 | -12.787 ± 0.199 | 16.985 ± 0.467 | | | |
| | | 60 | -7.3 ± 0.148 | -12.956 ± 0.203 | 16.985 ± 0.467 | | | |
| HJ-1 | C ^a | 20 | -22.53 | -4.2418 | -62.42 | Huang et al. [55] | | |
| | | 30 | -22.53 | -4.2639 | -60.28 | | | |
| | | 40 | -22.53 | -4.1272 | -58.89 | | | |
| | C ^b | 20 | -16.44 | -4.2418 | -41.63 | | | |
| | | 30 | -16.44 | -4.2639 | -40.19 | | | |
| | | 40 | -16.44 | -4.1272 | -39.34 | | | |
| | C ^c | 20 | -15.19 | -4.2418 | -37.37 | | | |
| | | 30 | -15.19 | -4.2639 | -36.06 | | | |
| | | 40 | -15.19 | -4.1272 | -35.34 | | | |
| | GAC | C | 15 to 45 | 17.99 | -16.84 to -20.45 | | 121.63 | Suresh et al. [46] |
| | NDA-100 | R | 10 | -24.93 | -5.38 | | -69.08 | Sun et al. [54] |
| | AH-1 | R | 10 | -31.37 | -5.99 | | -89.68 | Sun et al. [54] |
| 25 | | | -31.37 | -5.74 | -86.01 | | | |
| 40 | | | -31.37 | -5.99 | -81.09 | | | |
| AH-2 | R | 10 | -31.17 | -5.88 | -89.36 | Sun et al. [54] | | |
| | | 25 | -31.17 | -5.75 | -85.30 | | | |
| | | 40 | -31.17 | -5.25 | -82.81 | | | |
| HJ-1 | R ^a | 20 | -20.02 | -4.4591 | -53.11 | Huang et al. [55] | | |
| | | 30 | -20.02 | -4.3919 | -51.58 | | | |
| | | 40 | -20.02 | -4.2396 | -50.42 | | | |
| | R ^b | 20 | -14.40 | -4.4591 | -33.93 | | | |
| | | 30 | -14.40 | -4.3919 | -33.03 | | | |
| | | 40 | -14.40 | -4.2396 | -32.46 | | | |
| | R ^c | 20 | -13.06 | -4.4591 | -29.35 | | | |
| | | 30 | -13.06 | -4.3919 | -28.61 | | | |
| | | 40 | -13.06 | -4.2396 | -28.18 | | | |
| GAC | R | 15 to 45 | 28.32 | | 152.04 | Suresh et al. [46] | | |
| Modified bentonite | HQ | 20 | 21.1974 | -16.5869 to -17.0896 | 0.1268 | Yildiz et al. [51] | | |
| | | 40 | 8.9448 | -18.4797 to -18.3909 | 0.0873 | | | |
| GAC | HQ | 15 to 45 | 35.28 | -19.69 to -25.39 | 191.25 | Suresh et al. [48] | | |

^a20%; ^b30%; ^c40% [(q_e/q_m) × 100%]; AH-1 and AH-2, aminated hypercrosslinked polymers; CTAB-B, cetyltrimethylammonium bromide-modified bentonite; HJ-1, hypercrosslinked resin; GAC, granular activated carbon; NDA-100, hypercrosslinked resin without amino groups; C, catechol; R, resorcinol; HQ, hydroquinone.

Additional file

Additional file 1: Table S1. Hazard ranking of C, R, and HQ [181].

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

SS, VCS, and IMM conceived the concept and procedures of the analysis and drafted the manuscript. All authors read and approved the final manuscript.

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