

# Sorption of bisphenol A from aqueous solutions using natural adsorbents: isotherm, kinetic and effect of temperature

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**Abstract.** Emerging organic micropollutants, such as bisphenol A (BPA), have raised concerns about their negative impact on human health and ecological safety. This review article aims to demonstrate and highlight recent advances in adsorption applications for bisphenol A, a toxic environmental pollutant commonly found in wastewater. There are many reasons to use non-toxic materials and eco-friendly technologies to remove this pollutant from sewage. [1] Several adsorbents previously used have shown significant efficiency and performance for the removal of BPAs, and current research is directed towards the development of low-cost treatment processes using materials such as clays and Chitosan. The properties of the adsorbent can be adjusted by changing their surface for an optimized performance. In addition, the efficiency of the adsorption process depends on various parameters such as solution pH, the pollutant concentration, contact time, temperature, nature and dose of the adsorbent, which are also discussed. In addition, we critically review the isothermal, kinetic and thermodynamic approaches. [2]

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## 1. Introduction

Many industries reject effluents containing high concentrations of toxic elements, which constitute visual and chemical pollution that must be treated before the discharge of these waters downstream of the industries. Indeed, industrial processes provide the quality of consumer products necessary for today's society, but are unfortunately responsible for effluent discharges leading to water pollution. Emerging organic micropollutants, such as bisphenol A (BPA), have raised concerns about their negative impact on human health and ecological safety. [2]

Bisphenol A (BPA) is the common name for 2, 2-(4, 4'-dihydroxydiphenyl) propane, 4,4'-Isopropylidenediphenol or 2, 2'-bis (4 hydroxyphenyl) propane, an organic compound with two phenolic moieties. Bisphenol A (BPA) is used widely, mainly as a monomer in the industrial production of polycarbonate plastics, epoxy resins, and as a non-polymer additive to other plastics [3, 4]. Because of their toxicity, the U.S. Environmental Protection Agency and the European Food Safety Authority define the maximum acceptable amount of BPA as 0.05 mg/kg and 4 mg/kg body weight by day [5, 6, 7 ]. Bisphenol A has been detected in all kinds of environmental waters. The maximum concentrations reached up to 17.2 mg/L in hazardous waste landfill leachates, 12 µg/L in stream water and 0.1 µg/L in drinking water [8] Among the various methods developed to remove BPA from wastewaters [9,4], adsorption has significant advantages such as its efficiency, low operating cost, high selectivity, easy handling, etc. [10,4]. Because BPA is naturally hydrophobic, adsorption is one of the suitable processes for removing it from wastewaters [11, 12].

## 2. BPA removal by adsorption

The removal of BPA by adsorption has been widely explored by many researchers using various adsorbents. [13]

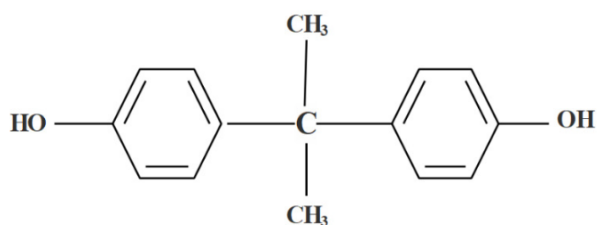


Fig. 1: Chemical structure of Bisphenol A (BPA).

## 2.1. Natural adsorbent

### 2.1.1. Chitosan

Chitosan, a natural polymer derived from natural substances and seafood waste, has received much attention as an effective biosorbant due to its low cost compared to activated carbon and its high content of amino and hydroxyl functional groups. [13] Kimura et al. investigated the combined use of the biopolymer Chitosan and the oxidoreductase polyphenol oxidase (PPO) for the removal of BPA from aqueous solutions. Optimal conditions for enzymatic oxidation of the BPA quinone are determined to be pH 7.0 and 4.0 C. The generated quinone derivatives are chemisorbed onto Chitosan beads, and BPA is completely removed in 4 to 7 hours.[14] The authors reported that the use of Chitosan in the form of porous beads is more effective than the use of Chitosan in solution or powder [13,14]. The removal efficiency of Chitosan (commercial (CC) and synthesized (SC in the laboratory)) is evaluated for the removal of BPA from an aqueous solution). The best Chitosan concentration is obtained at 0.06 g/L, the maximum removal rate of BPA adsorption is obtained by synthesized Chitosan at pH = 5, a contact time of 75 min, and adsorbent dose of 0.06 g/L, and a BPA concentration of 0.1 mg/L. The maximum adsorption capacities obtained are 34.48 and 27.02 mg/g for synthesized (SC) and commercial (CC) Chitosan, respectively [13, 15]. In another study, Chitosan and zeolites are environmentally friendly and low-cost adsorbents. Thus, the removal of organic PMs (such as bisphenol A (BPA), carbamazepine (CBZ), ketoprofen (KTF), and tonalide (TND) from aqueous solution via cross-linked Chitosan/zeolite is examined in this study. The hydraulic retention time is set at 0.8 h pH, and concentration of organic MPs ranged from 4 to 8 and 0.50 mg/L to 2.0 mg/L. They are considered as factors in optimizing the removal of pollutants via response surface methodology (RSM). Approximately 1.4560 mg/L (89.0%) of BPA, 1.4724 mg/L (90.0%) of CBZ, 1.4920 mg/L (91.2%) of KTF and 1.4118 mg/L (86.3%) of TND are removed at 5.1 pH and 1.636 mg/L initial concentration as the optimum removal efficiency based on RSM [16]. For effective BPA removal, Chitosan-immobilized zerovalent iron nanoparticles (nZVI-Chitosan) are synthesized and used as adsorbent. Since the BPA removal efficiency depends on independent process variables like nZVIChitosan dosage, initial BPA concentration, pH and contact time, experiments are conducted systematically, to characterize the interaction effect of these variables, the experimental matrix is designed by using response surface methodology (RSM). BPA removal efficiency is found to be around 97% for a solution having a BPA concentration of 6 mg/L and is removed by using the nZVI-Chitosan dosage of 2 g/L, at a pH of 3. Based on the obtained results, BPA removal using stabilized nanoparticles follows Freundlich isotherm and pseudo-first-order kinetic models.

Thermodynamic studies showed a favourable, spontaneous and endothermic adsorption process [17].

## 2.2. BPA removal by composite materials:

A composite material is a material prepared from two or more constituent materials with significantly different properties (physical or chemical) that when combined result in a material with other characteristics than the individual components. Composite materials are generally preferred because their properties create significant advantages in the products are manufactured from them. Many researchers have developed composite adsorbents for the removal of BPA from water [18]. Prepared CoFe<sub>2</sub>O<sub>4</sub>/PAC hybrid adsorbent by Li et al. in 2014 via an immersing-calcination process, using ethylenediamine tetraacetic acid (EDTA) and citric acid (CIT) ligands containing sol as the CoFe<sub>2</sub>O<sub>4</sub> precursor [13,18]. Li et al. 2015 fabricated a recyclable CNTs/Fe<sub>3</sub>O<sub>4</sub> magnetic composite via a facile hydrothermal method and used it for BPA removal; the dominant mechanism of BPA adsorption is supposed to be  $\pi$ - $\pi$  interactions between CNTs and the benzene rings in BPA molecules, as explained by the authors. The saturated CNTs/Fe<sub>3</sub>O<sub>4</sub> nanocomposite are independently regenerated by the methods of water-wash, methanol-wash and chemical oxidation (Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub>, Na<sub>2</sub>SO<sub>3</sub>/Fe<sup>2+</sup>, or UV/H<sub>2</sub>O<sub>2</sub>) [13, 19]. In another study, the use of PP-g-DMAEMA/PM composite fibre as an efficient adsorbent is demonstrated by combining graft polymerization of dimethylaminoethyl methacrylate (DMAEMA) with self-assembled modification of porous microspheres (PMs) on the surface of polypropylene (PP) fibre, and used for BPA removal. The preparation procedure of PP-g-DMAEMA/PM fibres (as reported by the authors). Authors claimed that because of the  $\pi$ - $\pi$  interactions and hydrogen bonds between BPA and PP-g-DMAEMA/PM, the resulting fibre obtained a higher adsorption amount (44.43 mg/g) of BPA. The presence of NaCl in the solution could facilitate the adsorption process, whereas the strong acid or strong alkali conditions and higher temperatures of the solution are unfavourable. Besides, the obtained fibre reusability without apparent deterioration in performance is demonstrated by seven repeated cycles [13, 20]. A hydrophobic magnetic montmorillonite composite is prepared by Salehinia et al. to remove BPA from aqueous media. The adsorption of BPA is found to increase by the increment of adsorbent dosage from 0.02 to 1 g/L and then remained constant [13, 21]. Palygorskite-montmorillonite is used as an adsorbent for BPA removal by Berhane et al. The pH (ranged 1.5-11.5), and ionic strength (0, 10, 100 mM CaCl<sub>2</sub>) are found to have a minor effect on BPA removal. The result of particle size (0.6 mm, 1.7-2.0 mm,  $\approx$ 2.8 mm) and temperature (25 C, 60 C, 75 C) indicated that the highest adsorption is observed using the smallest granule size and at the lowest temperature [13,22]. A graphene oxide cyclodextrin(GO-CD) nanocomposites is synthesized and efficiently used for the adsorption of toxic bisphenol A from wastewater. . The optimized values of effective parameters are 10 min (contact time), 25 mg (adsorbent dosage), 9 (pH) and

100 mg/l (initial concentration) these values are optimized using batch adsorption study in addition to this it is observed that. Langmuir adsorption isotherm is used to describe the adsorption equilibrium data, and the maximum adsorption capacity of 373.4 mg.g<sup>-1</sup> is obtained, while the pseudo-second-order model is used to describe the rate kinetics data of the adsorption method [23]. MWCNTs/Fe<sub>3</sub>O<sub>4</sub> magnetic composites, combined adsorption and catalytic ozonation behaviour, are synthesized by a hydrothermal process. The adsorption performance and catalytic activity of MWCNTs/Fe<sub>3</sub>O<sub>4</sub> for BPA removal in an aqueous solution are evaluated. The results showed that the synthesized MWCNTs/Fe<sub>3</sub>O<sub>4</sub> exhibited an excellent adsorption ability and catalytic ozonation activity, owing to its large specific surface areas hollow tube channels and abundant surface oxygen-containing groups, leading to high adsorption for BPA and O<sub>3</sub>, high retention and utilization of O<sub>3</sub> molecules on the composites. Moreover, BPA removal efficiency can be improved by increasing the dose of catalyst, pH value and the concentration of humic acid (HA). The fluorescence spectra and %OH radicals measurement displayed that HA could significantly increase %OH radicals via inducing more surface oxygen-containing groups on the catalyst, thus, promoting BPA removal. As a result, MWCNTs/Fe<sub>3</sub>O<sub>4</sub> showed excellent stability and durability during several reactions recycles [24]. The visible-light photocatalytic Fe doped TiO<sub>2</sub>/PSF composite ultrafiltration membranes are prepared in this study. Different parameters that affected the photocatalytic performance on BPA removal efficiency are discussed including metal variety, metal proportion, hydrothermal reaction time, and temperature and photocatalyst ratio. The resulted composite membrane, with a Fe-TiO<sub>2</sub> mass ratio of 0.20, exhibited a BPA removal rate of 90.78% within 180 min, enhanced mechanical capacity and self-cleaning ability [25]. A novel Fe-immobilized Chitosan/alginate composite is synthesized via a simple cross-linking reaction to activate PS. The PS/Fe-Chitoal system showed a higher BPA removal efficiency than the PS/Fe (II) ion system, releasing deficient amounts of Fe ions into aqueous solutions. FeChitoal possessed adsorptive properties toward BPA, but the degradation of BPA is mainly achieved via reactions with reactive species. From the quenching experiment, SO<sub>4</sub><sup>4-</sup> and •OH are confirmed as the prominent reactive radicals. Nevertheless, the PS/Fe-Chitoal system maintained a good efficiency under a wide pH range (pH 2–10) and in the presence of high concentrations of anions (Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>3-</sup>, and HCO<sub>3</sub><sup>3-</sup>) [26]. This study aimed to prepare and characterize polysulfone/graphene oxide nanocomposites membranes for the removal of BPA from water. Three membranes are synthesized using the phase inversion method: polysulfone membrane as PSF and two polysulfone/graphene oxide nanocomposites membranes with graphene oxide (GO) weight ratios of 0.4 and 1.0% as PSF/GO 0.4% and PSF/GO 1.0%, respectively. Les conditions optimales de pression d'entrée, de temps de fonctionnement, de concentration initiale de BPA et de pH pour l'efficacité de l'élimination du BPA par PSF/GO 0,4 % ont été déterminées à l'aide de la méthodologie de

réponse de surface et sont respectivement de 1,02 bar, 10,6 min, 7,5 mg/L et 5,5. En optimisant les conditions des paramètres de fonctionnement, l'efficacité expérimentale d'élimination du BPA par PSF/GO 0,4 % a atteint 93 % [27]. In this study, sulfur-doped titanium dioxide hollow spheres modified by surfactant loaded on magnetic bentonite (CST/ $\gamma$ -Fe<sub>2</sub>O<sub>3</sub>-BT) is synthesized in two steps, and bisphenol A (BPA) is chosen as the representative organic pollutant. The adsorption and photodegradation behaviour of CST/ $\gamma$ -Fe<sub>2</sub>O<sub>3</sub>-BT are examined. The Langmuir isotherm exhibited a better fit with a maximum adsorption capacity of 77.36 mg/g. At pH 7, the reaction rate constant (k) of the BPA photocatalytic degradation by-product is 0.00104 min<sup>-1</sup>, and the adsorption equilibrium constant (K) is 0.04034 L/mg [28]. Aluminium-based metal-organic framework/sodium alginate Chitosan (Al-MOF/SA-CS) composite beads are synthesized and employed as an adsorbent to remove bisphenol A (BPA). The main adsorption mechanisms between Al-MOF/SA-CS composite beads and BPA may involve  $\pi$ - $\pi$  stacking, hydrogen bonding and cation- $\pi$  interaction. At the same time, the enhancement of adsorption capacity caused by the introduction of CS can be related to obvious improvements in hydrogen bonding and newly formed cation- $\pi$  interaction. All results demonstrate that Al-MOF/SA-CS composite beads can be a promising adsorbent for BPA removal from wastewater [29]. In this study, TiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> composites are synthesized by the solvothermal method. The batch photodegradation experiments showed that TiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> displayed a highly efficient photodegradation rate of BPA (complete photodegradation within 20 min), good stability at pH 4.0–10.0, and high selectivity (superior to other organic pollutants). The •O<sup>2-</sup> radicals are responsible for highly efficient photodegradation of BPA on TiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> by quenching experiments, EPR and XPS analysis. These findings are critical to removing organic contaminants by using C<sub>3</sub>N<sub>4</sub>-based composites in environmental remediation [30]. Magnetic illite clay-composite material (Fe<sub>3</sub>O<sub>4</sub>@illite) is synthesized via the co precipitation method by loading Fe<sub>3</sub>O<sub>4</sub> nanoparticles (nanoFe<sub>3</sub>O<sub>4</sub>) onto the surfaces of illite clay. The catalytic BPA degradation of Fe<sub>3</sub>O<sub>4</sub>@illite and nano-Fe<sub>3</sub>O<sub>4</sub> confirmed the superior performance of Fe<sub>3</sub>O<sub>4</sub>@illite compared with that of nanoFe<sub>3</sub>O<sub>4</sub>. The optimum operating parameters for degradation are 0.3 mL of H<sub>2</sub>O<sub>2</sub> at pH of 3 in the presence of Fe<sub>3</sub>O<sub>4</sub>@illite, which provided a maximum degradation capacity up to 816, 364, 113, and 68 mg/g for epoxy BPA concentration of resin wastewater (266 mg/L), synthetic wastewater (80 mg/L), Hefei City swan lake (25 mg/L), and Hefei University lake wastewater (14.94 mg/L), respectively, in 180 min reaction time. The degradation data conformed to the pseudo-first-order kinetic model. The degradation pathways and mineralization study revealed that the adsorption Fenton-like reaction is the principal mechanism that demonstrated 100% degradation efficiency of Fe<sub>3</sub>O<sub>4</sub>@illite even after nine successive runs [31]. In this study, a simple water-based precipitation method is used to synthesize TiO<sub>2</sub>@nanodiamond composites. The ability of the composites to degrade bisphenol A as a model organic pollutant is investigated. It is found that

10 ppm of bisphenol A is wholly degraded in 100 min by the TiO<sub>2</sub>@nanodiamond photocatalyst under ultraviolet illumination [32]. In this work, T-ZnO-rGO-PEI composite is synthesized employing the hydrothermal method and the obtained composite samples. The catalytic activity and adsorption capacity of the synthesized T-ZnO-rGO-PEI composite is successfully explored using 4-nitrophenol and bisphenol-A as model pollutants. T-ZnO-rGO-PEI composite and found that 4-NP reduction reaction were completed within 10 min with the rate of 0.224 min<sup>-1</sup>. The BPA adsorption over T-ZnO-rGO-PEI exhibited a high adsorption rate of 0.0210 min<sup>-1</sup>. In addition, the detailed 4-NP reduction and BPA adsorption mechanism is demonstrated. Hence the synthesized T-ZnO-rGO-PEI composite is a promising catalyst for removing micropollutants in an aqueous medium [33].

### 3. Equilibrium and kinetic modelling and thermodynamic studies:

The adsorption isotherms are applied to indicate the correlation between the pollutant adsorbed on the adsorbent and the pollutant concentration in the solution. The study of isothermal models is an essential part of adsorption studies to explain the interactions between contaminants and adsorbents. [34] In most studies focusing on BPA removal, only two isotherm models are tested [13], Langmuir isotherm fitted best to the experimental adsorption data. Kinetic studies are essential for the prediction of optimum conditions in the full-scale adsorption processes [35]. Kinetic modelling provides valuable information about adsorption mechanisms and possible rate-controlling steps [35]. In most examined BPA removal studies, only pseudo-first and pseudo-second-order models are used to represent the kinetic results, and there is a lack of mechanism explanation using other models mentioned above. [13] Pseudo-second-order interprets the kinetic experimental data satisfactorily. Thermodynamic studies are used to estimate the thermodynamic parameters, such as Gibbs free energy ( $\Delta G^\circ$ ), enthalpy change ( $\Delta H^\circ$ ) and entropy change ( $\Delta S^\circ$ ) [13]. Moreover, thermodynamic parameters may be used as a base to explain the adsorption mechanism [13].

### 4. Conclusion

This review centres on removing BPA from waters and wastewater by various sorts of adsorbents like clays and clays minerals, and composite adsorbents. The previously mentioned adsorbents seem to have sufficiently adsorptive attributes in a wide range of different examined parameters. The viability of the adsorbents, for the most part, relies upon characteristics and characteristics of the matrix. It is being discovered that the chemical modification can improve the adsorption limit of adsorbents when contrasted with the unmodified materials. However the utilization of chemical substances in alteration steps could prompt



secondary contamination and pollution (because of the filtering and the leaching of synthetic compounds utilized in the process). Additionally, the use of chemical substances for alterations can likewise build the treatment cost. Critical discussion about the pollution caused by modified adsorbents was seldom reported in the literature reviewed herein [13]. Notwithstanding, some issues should be mulled over for a better understanding of the adsorptive characteristics of these materials. It is essential to take note that in the more significant part of the examinations, the Langmuir isotherm model and pseudo-second-order model are found to represent best the adsorption data. Regarding equilibrium modelling, it was recommended to examine three parameters models also besides two parameters isotherms (Langmuir, Freundlich etc.) for a better understanding of the adsorption mechanism. Assessment of thermodynamic parameters, based on a set of temperature-dependent equilibrium adsorption data, should be viewed with caution since enthalpy-entropy compensation is observed [13]. Adsorption generates a large amount of BPA-loaded waste, and as a result, its disposal is a crucial environmental issue. Attention is chiefly on defining maximum adsorption capacities in batch mode with engineered BPA solution. Because of the absence of column-, pilot-, and full-scale examines, it is hard to appraise the capability of these adsorbents in real applications of BPA removal under natural ecological conditions. [13]

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