

## Adsorptive removal of endocrine disrupting bisphenol A from aqueous solution using chitosan



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### ABSTRACT

Bisphenol-A (BPA) is an important industrial chemical generally introduced into the environment through the wastes of factories producing, for example, paper, plastics and plastic products-making industries. Consequently, factory effluents and sewage containing BPA are sources of contamination in the aquatic environments. The objective of this research is to study the efficiency of BPA removal from aqueous solutions using two chitosans, (1) synthesized chitosan; a waste derived adsorbent from waste seafood shell and which was synthesized in our laboratory and (2) commercial chitosan a chitosan sample obtained commercially. The effects of process parameters such as solution pH, adsorbent dose, contact time, and initial concentration of BPA have been examined and compared. The results obtained from this study indicated that the efficiency of removal declines with increased contaminant concentration and decreased adsorbent dose. The optimal pH for maximum adsorption for both chitosan adsorbents was 5. The adsorption data were fitted into six isotherm models namely Langmuir, Freundlich, Temkin, Dubinin-Radushkevich, Redlich–Peterson and Sips and the isotherm parameters were determined. Furthermore, the kinetics of the process were correlated by three kinetic models, namely, the pseudo-first-order, pseudo-second-order and intra-particle diffusion models. The kinetic experiments revealed that both chitosan adsorbents follow the pseudo-second-order kinetic model. The isotherm studies demonstrated that both adsorbents follow the Langmuir model. Based on the results, chitosan, synthesized (SC) and commercial (CC), is an efficient, novel and inexpensive alternative for the removal of BPA from aqueous solution.

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### 1. Introduction

Bisphenol-A (BPA) is one of the derivatives of phenol. The chemical name of BPA is 2, 2-(4,4-dihydroxy phenyl) propane and it consists of two mols phenol plus one mol acetone. The formula of bisphenol-A is  $C_{15}H_{16}O_2$  and BPA is a solid with low volatility properties at normal environmental temperatures. Its solubility in water is greater in an alkaline pH and ranges from 120 to 300 mg/L

[1]. Based on these properties, around 50% of BPA available in the environment has the potential to bind to sediments or soil in water systems. BPA is an estrogen-like endocrine disrupting chemical (EEDC) with two unsaturated phenolic rings [1]. It is an important industrial chemical widely used in polycarbonated plastic as well as epoxy resin, both of which are very practical materials [2]. A high concentration of BPA is observed in the wastewaters produced by factories producing this material (the primary BPA concentration in wastewater of apolycarbonate factory is about 100 mg/L) [3,4]. Since only a small amount of BPA is removed during wastewater treatment, BPA-containing wastewater can be a source of contamination in aquatic environments. This material can enter the environment during the factory processing and thereby pollute the rivers and groundwater, sediments in the soil and accumulates there [5]. BPA is a non-degradable biological antioxidant which

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shows great resistance against chemical decomposition, making it a hazardous material for human and animal health [6]. Important effects of low doses of BPA in experimental animals have recently been reported in the form of blood disorders at the same or lower level than the mean blood levels in humans [7]. Very low doses of BPA cause proliferation of prostate cancer cells in humans, heart disease, diabetes Type 2, and abnormalities in liver enzymes [8]. It is a contaminant of aquatic ecosystems whose solubility in water is far greater than  $EC_{50}$  [9]. BPA is found in natural water in response to the discharge of untreated waste from industries and is highly dangerous in aquatic ecosystems since BPA disrupts the endocrine glands (they have the role of estrogen receptor) [10]. The presence of even very low concentrations of BPA in water threatens the life of aquaculture through the disruption of endocrine glands [11]. Consequently, BPA removal from aquatic environments is absolutely crucial and various methods are available, including physical [12], chemical, and biological methods [13], ultrafiltration [14], reverse osmosis [14], photoelectrocatalytic [15] and biological reduction [16], for the treatment of this type of wastewater. Most of these methods have certain disadvantages including: high cost, the need for further treatment, the formation of hazardous by-products, low efficiency, and applicability for a limited concentration range. Among the diverse treatment methods, the efficiency of biological approaches is not satisfactory due to the BPA toxicity for aerobic and anaerobic bacteria [17]. Chemical sedimentation is another method but less popular due to the production of a large amount of sludge and the relatively low efficiency of common chemical precipitation methods. One of the effective treatment processes for the removal of phenolic compounds is adsorption [18]. The most widely used adsorbent for many years is activated carbon [19] but activated carbon is relatively expensive [20]. In this regard, researchers have been trying to develop inexpensive adsorbents for the substitution of activated carbon. Wastes have become the focus of attention for producing inexpensive adsorbents for assisting in the minimization of recycling and reuse [21]. Chitosan has been employed as an effective coagulating agent of organic compounds and has also been used as a chelating polymer for binding of heavy and toxic metals [22]. It has been used as an adsorbent for the removal of organic compounds including phenol and its derivatives together with dyes and other compounds in the water and wastewater.

The most important property of chitosan is that it is a safe, environment-friendly, polysaccharide, waste-derived, non-toxic and natural occurring polymeric compound. Therefore, the aim of this study is to examine and compare the efficiency of BPA removal from aquatic solutions using a waste derived adsorbent, chitosan in two types: (1) synthesized chitosan (SC), synthesized in our laboratory from waste shrimp shells, and (2) commercial chitosan (CC), purchased as a commercial product.

## 2. Materials and methods

### 2.1. Materials

The molecular structure of chitosan is shown in Fig. 1.

The experiments were conducted at laboratory scale in a batch system. All the chemicals used in this study including BPA were supplied by Merck, Germany. Table 1 shows the physicochemical properties of BPA. Solution pH adjustment was carried out by 1 N HCl and 1 N NaOH.

### 2.2. Adsorption studies

The experimental parameters which were examined included contact time, concentration of the contaminant (BPA), the amount of adsorbent, the type of chitosan and the pH. The adsorbents used

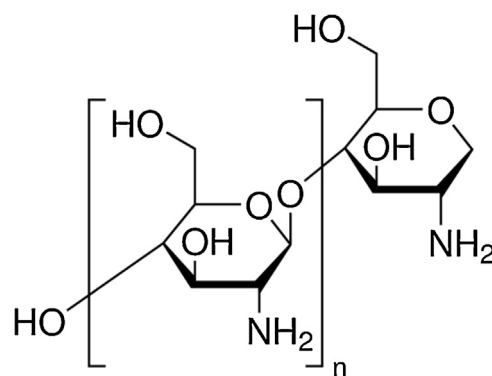


Fig. 1. Molecular structure of chitosan.

in this study were two chitosans, one synthesized chitosan (SC) obtained from shrimp shell synthesized in our laboratory and the other commercial chitosan was supplied by Sigma Aldrich and is designated as “commercial” chitosan (CC) throughout this study. Chitosan was first sieved through a 50 mesh sieve to achieve a control size of particles. For the investigation of the degree of adsorption of BPA, 0.02 g chitosan was weighed, transferred into a 250 mL flask with a BPA concentration of 0.5 mg/L; the flask was then placed onto a shaker-equipped stirrer with rotation of 100 rpm. Samples were taken every 15 min, followed by measuring the absorbance at the wavelength of 504 nm using a UV-visible spectrophotometer Lambda 25 (PerkinElmer, Norwalk, CT, USA) to determine the contact time to reach equilibrium [15]. After establishing the equilibrium time, in order to obtain the optimal pH for maximum adsorption, 0.02 g chitosan with a concentration of 0.5 mg/L was dispensed into four flasks, followed by adjustment of the pH at 3, 5, 7, and 9, respectively. In order to obtain the kinetic and adsorption isotherm data, the experiments were carried out at BPA concentrations of 0.1–1 ppm with adsorbent doses of 0.01–0.08 g/L at the four different pH values and contact times of 15–120 min.

### 2.3. Preparation of synthesized chitosan (SC)

In this study, discarded shrimp shells were used for the extraction of chitin. After washing the shrimp skins with amount water, they were dried under exposure to sunlight for one week at room temperature to avoid any thermal degradation. They were then powdered using a ball mill and prepared for the next stages of chitin and chitosan extraction. First, shrimp shell powder was added to 1 N HCl with a ratio of 1:20, then stirred for 2 h at 125 rpm, and kept at room temperature for 24 h. This process causes the removal of minerals including  $CaCO_3$ . The slurry was then filtered, and kept at room temperature for 24 h until dry. At this stage, the material obtained from the previous stage was placed in an agitated beaker with stirring at 60 rpm and a ratio of 1:20 powder and 1 N NaOH for the extraction of chitin for 4 h reaction time. This process removed protein and produced chitin. The next stage, involving drying the chitin and then reacting the dried chitin with 50% w/w NaOH for the deacetylation of chitin to produce chitosan. This reaction was performed with agitation at 100 °C for 2 h to complete the deacetylation of chitin and the conversion to synthesized chitosan (SC) [22].

### 2.4. Analytical methods

Firstly, 15 mL of the prepared sample was poured into a falkon tube, then placed in a 6000-rpm centrifuge for 15 min, and filtered. An amount, 0.25 mL  $NH_4OH$  (2.5 mL per 100 mL of the sample), was

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