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Efficient adsorption of bisphenol A from aqueous solutions using low-cost activated carbons produced from natural and synthetic carbonaceous materials

N. Jaafarzadeh[a], Z. Baboli[b,c], Z. Noorimotlagh[d,e,*], S. Silva Martínez[f], M. Ahmadi[a], S. Alavi[c], S.A. Mirzaee[e,*]

[a]*Environmental Technologies Research Center, Ahvaz Jundishapur University of Medical Sciences, Ahvaz, Iran, emails: Jaafarzadeh-n@ajums.ac.ir (N. Jaafarzadeh), ahmadi-m@ ajums.ac.ir (M. Ahmadi)*
[b]*Department of Environmental Health Engineering, Behbahan Faculty of Medical Sciences, Behbahan, Iran, email: baboliz87@gmail.com*
[c]*Department of Environmental Health Engineering, Ahvaz Jundishapur University of Medical Sciences, Ahvaz, Iran, email: sabaalavi394@yahoo.com (S. Alavi)*
[d]*Student Research Committee, Ahvaz Jundishapur University of Medical Sciences, Ahvaz, Iran, email: noorimotlagh.zahra@gmail.com*
[e]*Department of Environmental Health Engineering, School of Health, Ilam University of Medical Sciences, Ilam, Iran, Tel./Fax: +989183459155; email: Mirzaee.seyyed@gmail.com*
[f]*Centro de Investigación en Ingeniería y Ciencias Aplicadas, Av. Universidad 1001, Col. Chamilpa, Cuernavaca, Morelos, Mexico, email: ssilva@uaem.mx*

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ABSTRACT

Milk-vetch (MV) and compact discs (CDs) wastes, as natural and synthetic carbonaceous precursors, were used to produce low-cost activated carbons. The efficiency of adsorption of these adsorbents was compared with commercial activated carbon (CAC) for the adsorption of bisphenol A (BPA) from aqueous solutions. The activated carbon prepared from MV (MVAC) had a higher adsorption capacity, 33.19 mg g^{-1}, than that of both CD wastes (CDAC), 12.15 mg g^{-1}, and CAC, 29.22 mg g^{-1}. These adsorbents were characterized using X-ray diffraction, scanning electron microscope, Brunauer–Emmett–Teller method, pH_{zpc} and Fourier transform-infrared spectra techniques. The effect of operating parameters on the adsorption of BPA such as the pH, contact time, adsorbent dosages, initial BPA concentrations, temperature, and ionic strength were studied. The optimal removal of the BPA was achieved at neutral pH (pH 7) by three adsorbents. Equilibrium contact times of 90 min were achieved for all the investigated activated carbons. Based on the obtained results, the pseudo-second-order kinetic model and Langmuir isotherm fitted the experimental finding better than other isotherms and kinetics. Furthermore, thermodynamic parameters were calculated and ΔG, ΔH and ΔS indicate that the reaction is spontaneous and endothermic, and physisorption mechanism was the dominant mechanism in terms of adsorption of BPA onto the three adsorbents.

Keywords: Adsorption; Bisphenol A; CD wastes; Milk-vetch; Activated carbon

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* Corresponding authors.

1. Introduction

Bisphenol A (BPA) has been listed as one of the endocrine disrupting chemicals widely used in the plastic industry as an important monomer, and frequently detected as contaminant in water sources in recent years [1–6]. Polycarbonate plastics are used in different materials such as food storage containers, baby bottles and dental sealants [7–12]. BPA has important properties including moderate water solubility, low vapor pressure and volatility, and is in white granule form at room temperature [1,3]. The global BPA consumption in 2011 was 5.5 million tons [1,3,13]. BPA is categorized as "moderately toxic" with LC_{50} equal to 1.0 mg L^{-1}, whereas is grouped as "toxic" to environmental microorganism with 10 mg L^{-1} concentration [3]. Minnesota Department of Health (MDH) reported a guidance value of 20 ppb for BPA (below this level in drinking water would have little or no risk of any health effects). In surface water, BPA concentration ranged from no detection to microgram per liter. Slightly lower levels of BPA were reported in potable tap water samples with mean and maximum levels of nanogram per liter and microgram per liter, respectively. There is a wide variation in the values reported for BPA in different industries, but many fall within the range of 1 μg L^{-1}–150 mg L^{-1} [8,10,14–17]. Moreover, it was shown that BPA induced the reproductive disorders even at very low doses [18,19]. It has been reported that BPA may induce infertility and reproductive parameters disorders, genital tract abnormalities, type 2 diabetes, prostate and breast cancer in both humans and animals [20]. Therefore, removal of BPA from wastewater and water before being discharged into aqueous solutions is of utmost importance [11,18,16].

Biological, chemical and physical methods have been applied for degradation/removal of pollutants from contaminated water/wastewater such as ozonation, ion exchange, electrochemical treatment, chemical precipitation, advanced oxidation processes, reverse osmosis, and adsorption [1,11,14,21–26]. Among them, physical adsorption is a forcible method for various pollutants from different sample matrices [18].

In the adsorption process, one of the key factors is adsorbent. Taking this into account, activated carbon (AC) considered as one of the most important materials for adsorbent among the various adsorbent materials due to excellent adsorption capacities, ease of operation and high resistance to environmental toxicity [27–32]. The use of commercial AC is limited because of its high-cost and the need for regeneration [33,34]. Therefore, recently attention has been focused on the application of AC produced from low-cost carbonaceous materials, such as natural and synthetic material wastes for the removal of different pollutants from contaminated water [14,33–39]. Wirasnita et al. [15] tested the activated carbon prepared from the empty fruit bunch of oil palm wastes (as bio-adsorbent) and found that the maximum adsorption capacity of bio-adsorbent for the adsorption of BPA from aqueous solutions was 41.98 mg g^{-1}. Bautista et al. [40] studied the behavior of activated carbon prepared from almond shells for removal of BPA and found this adsorbent is a good candidate for the treatment of solutions contaminated. The AC prepared from locally available carbonaceous materials and the highly used digital wastes (such as waste compact discs) can be a favorable and economical approach for the removal of various pollutants in aqueous phase. Therefore, in this study, the use of milk-vetch (MV), as a locally available plant in Iran, and waste compact discs (typically CDs) have been considered as inexpensive carbonaceous precursor for AC to remove BPA from aqueous solutions [29,41].

Therefore, the aim of the present study is to survey the efficiency of adsorption of BPA from aqueous solutions with two types of AC prepared from MV (MVAC) and CD wastes (CDAC), both compared with the commercial AC (CAC), as reference. The influence of different experimental factors such as pH, contact time, adsorbent dosage, initial BPA concentrations, temperature and competing anions on the efficiency of adsorption of BPA was investigated. Furthermore, isotherm and kinetic studies were carried out to find the adsorption mechanism.

2. Materials and methods

2.1. Chemicals

Bisphenol A (BPA, $C_{15}H_{16}O_2$, >99% purity), CAC, potassium hydroxide (KOH), hydrochloric acid (HCL), phosphoric acid (H_3PO_4) and sodium hydroxide (NaOH). All chemical reagents were supplied from Merck, Germany. The BPA stock solution was processed by dissolving 1 g of BPA in 1 L of distilled water; the solution was shaked for an hour and then stored. Various concentrations of BPA were prepared by diluting the stock solution using distilled water. If necessary, the solution pH was adjusted with 0.1 M HCl or 0.1 M NaOH.

2.2. Adsorbent preparation

The synthesis of AC prepared from MV and CD wastes was carried out by the thermo-chemical method. Two procedures were followed to prepare the adsorbents depending on the material source as reported by Noorimotlagh et al. [29,42]. Briefly, the CD waste precursor of AC consisted of mixing 4 g of CD bits with HCl and then annealed at 500°C for 1 h. Afterwards, for 1 h were soaked in KOH. Finally, the prepared activated carbon of CDAC was washed with distilled water. The procedure for the MV precursor of AC consisted on collecting MV from the mountains of Ilam city, Iran (the Islamic republic of Iran). Then for activation, 5 g of dry MV were placed in phosphoric acid for 1 h under stirring, followed by carbonization at 600°C for 1 h. Finally, the prepared activated carbon of MVAC was washed with distilled water.

All the activated carbons (CDAC and MVAC, including the CAC) were crushed by hand and passed through a bolter of 60 and 120 mesh (particle size of 0.15–0.5 mm). Then, the remaining carbon was washed with distilled water, dried at 100°C for 6 h, and stored in a sealed glass bottle for subsequent experiments [43].

2.3. Characterization and apparatus

The structural features of MVAC, CDAC and CAC were determined using a scanning electron microscope (SEM, Tescan, Mira3, Czech Republic). The X-ray diffraction (XRD) was performed to determine the crystalline phases of these

ACs at 40 kV and 30 mA with Cu Kα (λ = 1.54 Å) radiation on a Philips Xpert diffractometer (Netherlands) over the 2θ of 10°–80°. The Fourier transform-infrared (FT-IR) spectra for all AC were performed from 4,000 to 500 cm^{-1} at a resolution of 1 cm^{-1} by St. Jean Baptiste Bomem 450 (Canada). The Brunauer–Emmett–Teller method (BET-Micromeritics Tristar 3000, USA) was used for the determination of the adsorbent specific surface area. The concentration of BPA was obtained from a calibration curve built with the absorbance measured at 241 nm from BPA standard solutions of different content of BPA using an UV–Vis spectrophotometer (DR-5000, Hach Co, USA) [44]. The potentiometric method was used to determination of the zero charge point (pH_{zpc}) inside the pH range of 2–12 to find out of the adsorbent surface charge [29]. The solution pH was measured by a Metrohm pH meter (Model: E532, Germany).

2.4. Method of experiments

For adsorption of BPA onto the prepared activated carbons at 25°C temperature, 100 mL of high-density polyethylene bottles were used and batch experiments were applied to investigate the influence of the solution pH (3, 5, 7, 9 and 11), contact time (15, 30, 60, 90, 120, 150 and 180 min), initial concentration of BPA (25, 50, 75, 100 and 150 mg L^{-1}), adsorbent dosage (0.02, 0.05, 0.075 and 0.1 g 0.05 L^{-1}), ionic strength (20, 40, 60, 80 and 100 mg L^{-1} as Ca^{2+} ion) and temperature (15°C, 25°C, 35°C and 45°C). The solutions were agitated using a shaker with 250 rpm for an effective interaction between the adsorbent with BPA. After the adsorption time, the solutions were filtrated through 0.22 μm syringe filters and the concentration of BPA was measured. Eqs. (1) and (2) were used for efficiency of BPA removal and the adsorption capacity (q_e), respectively.

$$\mathrm{RE}(\%) = \left(\frac{C_0 - C_e}{C_0}\right) \times 100 \tag{1}$$

$$q_e = \frac{(C_0 - C_e) \times V}{m} \tag{2}$$

where RE(%) and q_e (mg g^{-1}) are the removal efficiency and the amount of adsorbed BPA onto the unit amount of the adsorbent, respectively. C_0 (mg L^{-1}) and C_e (mg L^{-1}) are the initial and the equilibrium concentration of BPA, respectively. V (L) and m (g) are the volume of solution and the mass of adsorbent, respectively [25,45]. Experimental tests were performed three times and the mean values are reported.

2.5. Isotherm and reaction kinetics

Freundlich, Dubinin–Radushkevich (D-R) and Langmuir adsorption isotherm models were applied to investigate the behavior of BPA adsorption onto MVAC, CDAC and CAC. The isotherm equations were linearized as the following equations:

Langmuir equation

$$\frac{C_e}{q_e} = \frac{1}{bq_m} + \frac{C_e}{q_m} \tag{3}$$

Freundlich equation

$$\log q_e = \log K_f + \frac{1}{n}\log C_e \tag{4}$$

D-R equation

$$\log q_e = \log q_m - \beta\varepsilon^2 \tag{5}$$

where C_e (mg L^{-1}) is BPA concentration, q_e and q_m (mg g^{-1}) are the amount of sorbed BPA and the maximum sorption capacity at the equilibrium time, respectively. K_f, n and b (L mg^{-1}) are the Freundlich and Langmuir constants, respectively. At D-R equation β (kJ mol^{-1}) is the mean adsorption energy and ε is the Polanyi potential [43,15].

The experimental data were fitted to pseudo-first-order and pseudo-second-order kinetics for understanding the adsorption mechanism of BPA onto prepared activated carbons. The linearized pseudo-first-order and pseudo-second-order kinetic models are represented through the following equations:

Pseudo-first-order model

$$\mathrm{Ln}(q_e - q_t) = \mathrm{Ln}q_e - k_1 t \tag{6}$$

Pseudo-second-order model

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{7}$$

where q_t and q_e (mg g^{-1}) are the amount of adsorbed BPA at time t (min) and the equilibrium time, respectively. k_1 (1 min^{-1}) and k_2 (g mg^{-1} min^{-1}) are the rate constant belonging to the pseudo-first-order and pseudo-second-order kinetic models [29,15].

For this purposes, a set of experiments were performed to get the kinetic and the adsorption isotherm data. These consisted of mixing (at 250 rpm) the given amounts (0.075 g) of adsorbent with 50 mL of BPA solution (C_0 = 50 mg L^{-1}) at pH = 7 (found as the optimal value from the pH-effect experiments) and room temperature (25°C) for different time intervals (30–180 min). In the adsorption isotherms, a BPA concentration interval of 25–150 mg L^{-1} was tested. Finally, liquid samples were collected and BPA concentration was analyzed.

3. Results and discussion

3.1. Characterization of adsorbents

The synthesis of AC, prepared from MV and CD wastes, was clearly confirmed by the XRD spectra. These XRD spectra are compared with CAC, used as reference AC, as shown in Figs. 1a–c. A sharp and strong peak is observed in CAC graph at 2θ = 26.75° (Fig. 1a) corresponding to the (002) diffraction of graphitic crystallites. However, broader peaks are observed for MVAC and CDAC spectra (Figs. 1b–c) indicative of a low graphitization degree in the as-prepared adsorbents. The weak diffraction peak at 2θ = 45.01° for CDAC and MVAC spectra is assigned to the characteristic reflection of carbon amorphous nature (Figs. 1b–c).

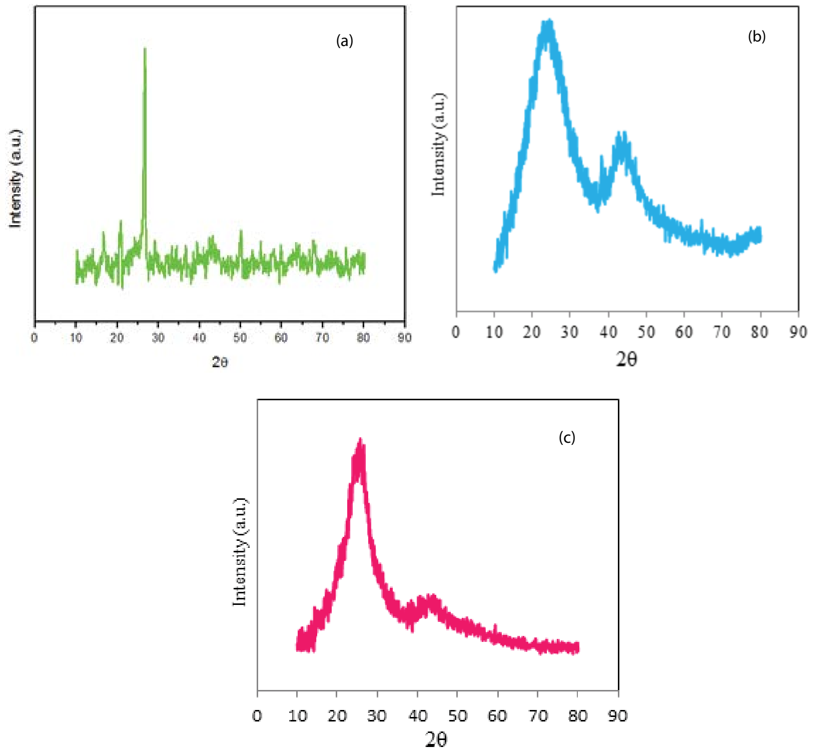

Fig. 1. XRD spectra of the activated carbons of (a) CAC, (b) CDAC, and (c) MVAC.

Fig. 2 shows the SEM images of the AC of CAC, reference AC (Fig. 2a), CDAC (Fig. 2b) and MVAC (Fig. 2c). Based on these images, the prepared ACs show uneven structure and irregular pores. The images reveal that the external surface of MVAC has more porosity compared with CDAC and CAC. This phenomenon may be related to the phosphoric acid used as an activating agent. It has been reported that phosphoric acid could change the textural properties of ACs and produce larger amounts of pores on the surface of the ACs than other activating agents, such as KOH and HCl. Similar result has been reported by Arampatzidou and Deliyanni [14] using activated carbons obtained from pyrolysis of potato peels and the activation with various activating agents (such as $ZnCl_2$, KOH and H_3PO_4) in the adsorption of BPA. It was reported that the differences in the activation agents resulted in significant changes in the morphology and chemistry of activated samples. Furthermore, the activated carbons derived from phosphoric acid showed the best adsorption capacities compared with the samples activated with KOH and $ZnCl_2$ [14]. Highly porous structure of MVAC provides higher surface area and adsorption capacity for the adsorption of BPA molecules. As demonstrated by the BET analysis, giving specific surface areas of 596, 455 and 79.71 m^2 g^{-1} for MVAC, CAC and CDAC, respectively.

FT-IR analysis was carried out to evaluate the surface nature of the adsorbents and the involvement of the surface functional groups in the adsorption of BPA onto the ACs. The FT-IR spectra of the prepared ACs in the 4,000–400 cm^{-1} region show various absorption peaks (Fig. 3). Five absorption bands are mainly observed and appear at around: (a) 496–600 cm^{-1} and (b) 1,050–1,166 cm^{-1}, that represents the C–O stretching mode corresponding to the carbonyl group; (c) the broad and strong band at 1,383 cm^{-1}, attributed to the C–O vibrations in carboxylate groups; (d) 1,590–1,600 cm^{-1}, corresponding to a C–O–C structure and stretching of conjugated C=C in aromatic ring; and (e) 3,419–3,430 cm^{-1}, corresponding to the surface hydroxyl O–H groups, for the three adsorbents [15,46,47].

3.2. *Influence of initial pH*

The pH of the solution is a crucial parameter that influences the adsorption process in a typical batch system, since it can affect the surficial charge of the prepared adsorbents and adsorbate species within the aqueous solution [29]. The effect of the pH on the adsorption of BPA by AC of MVAC, CDAC and CAC (used as reference) is depicted in Fig. 4a. The experimental conditions were an adsorbent dosage = 0.075 g, 50 mL solution, initial BPA concentration = 50 mg L^{-1}, contact time = 120 min, various pH values = 3, 5, 7, 9 and 11. The findings imply that the efficiency of adsorption decreased with an increase in the solution pH for the three ACs. By increasing the initial pH from 3 to 11, the adsorption capacity (q_e) of BPA decreased from 15.52 to 5.75 mg g^{-1} for CDAC, from 32.21 to 16.84 mg g^{-1} for CAC, and from 36.32 to 14.21 mg g^{-1} for MVAC. The increase in the initial pH of the solution from 3 to 7 resulted in a gradual decrease in the adsorption of BPA (from 15.52 to 12.2 mg g^{-1} for CDAC, from 32.21 to 29.61 mg g^{-1} for CAC, and from 36.32 to 33.33 mg g^{-1} for MVAC); while, in the region of higher pH, which ranges between 9 and 11, there was a strong decrease in q_e of BPA for the three adsorbents. Therefore, due to the small decrease

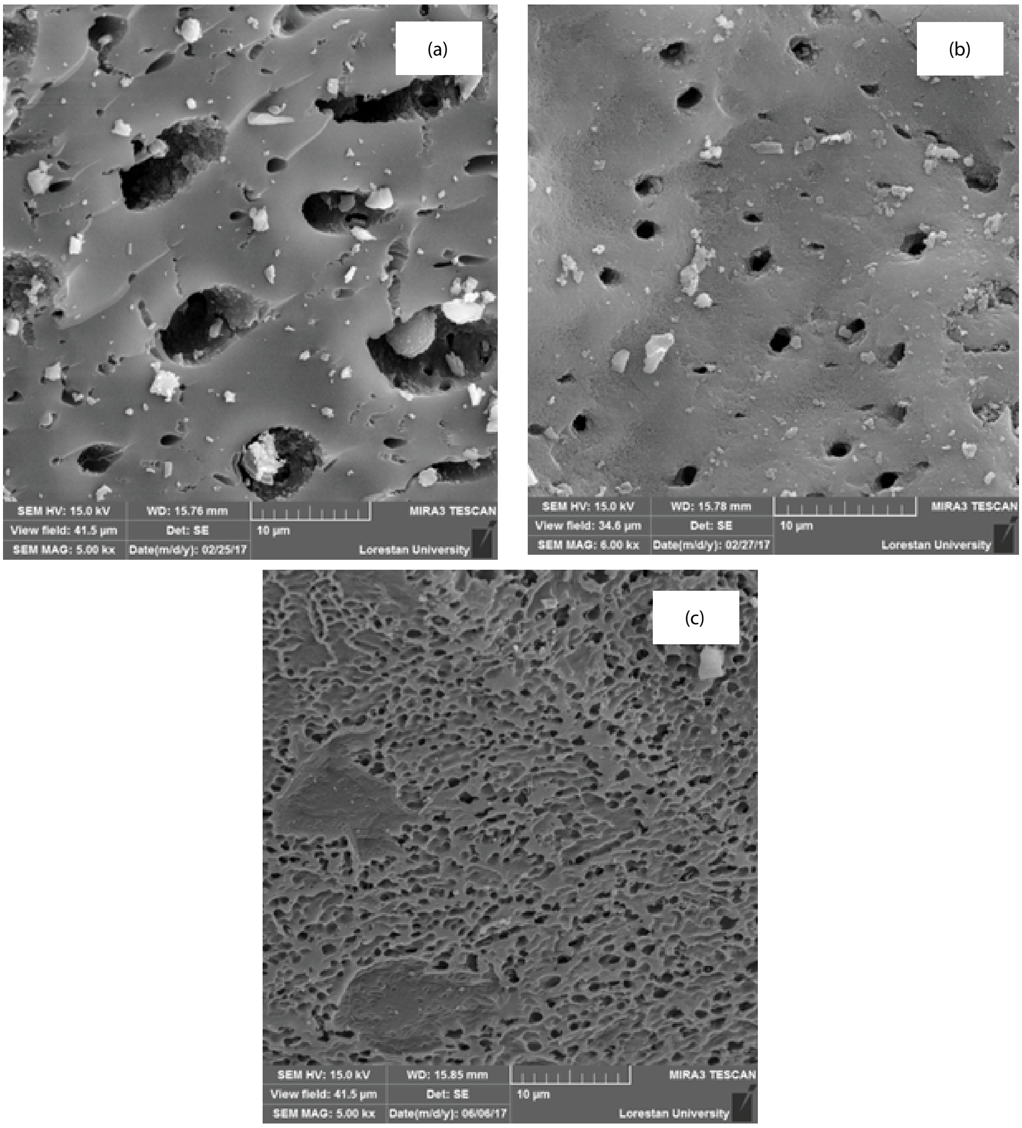

Fig. 2. Surface morphology of the adsorbents by SEM method for (a) CAC, (b) CDAC and (c) MVAC.

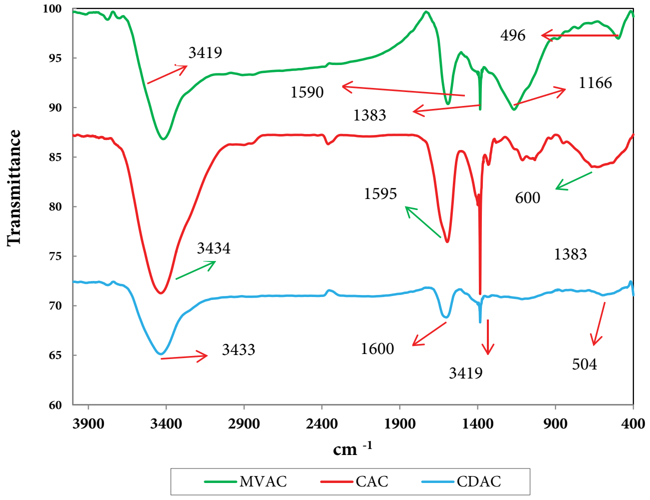

Fig. 3. FT-IR spectra of CAC, CDAC, and MVAC.

in the adsorption of BPA within the initial pH range between 3 and 7, the extreme acidity condition of the solution in pH equal 3, need to use excessive acid to reach in pH equal 3 at real (large) scale application and considering an economic point of view, a pH value of 7 was chosen as the optimum value for subsequent experiments.

The increase in the initial pH leads to increase in hydroxide ions (OH^-) that compete with BPA anions for occupying the adsorbent sites for adsorption, while the decrease in the initial pH results in the protonation of the adsorbents surface, which favors the capture of BPA anions [48]. The pH_{zpc} of the AC of MVAC, CDAC and CAC was estimated to be 3, 4.3 and 5.1, respectively (Fig. 4b. Regarding to the pH_{zpc} values, it is reported that the surface of the prepared adsorbents will be deprotonated at pH values higher than pH_{zpc}; thus, the adsorption of negatively charged BPA is decreased at pH values higher than pH_{zpc}. The deprotonation and, consequently, the negatively charged surface of the prepared adsorbents were produced by increasing the pH of the

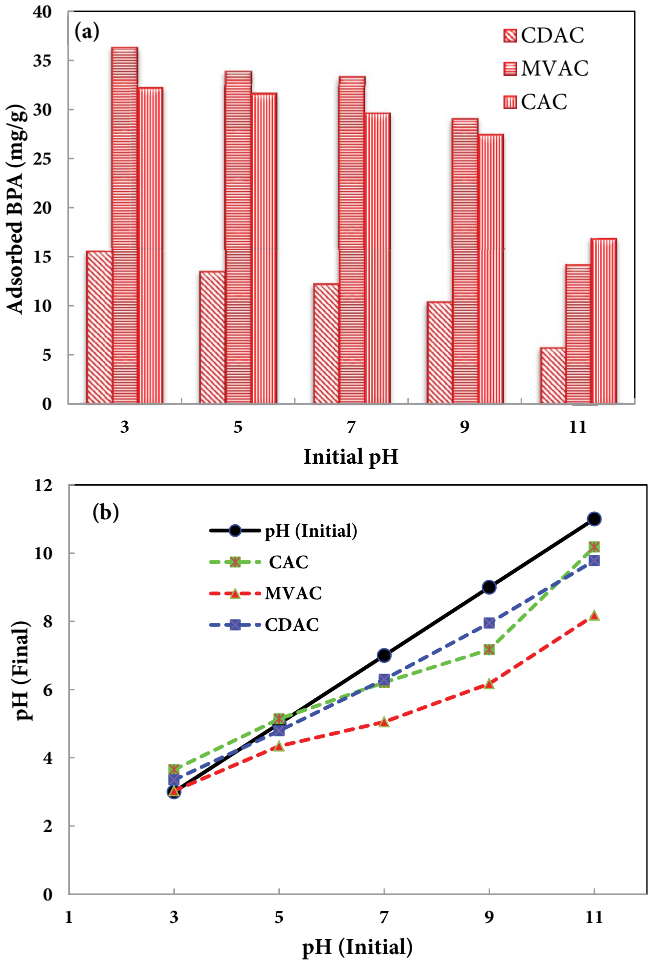

Fig. 4. (a) Effect of pH on the adsorption of BPA onto CDAC, MVAC and CAC and (b) pH_{pzc} plot for MVAC, CDAC and CAC.

solution. While, this phenomenon may reason for a decrease in BPA adsorption, in agreement with other studies [1,13,47].

Liu et al. [18] demonstrated that the maximum removal of BPA by ACs (W20 and W20N) was achieved at the pH range of 3–8. When the pH of solution increases to 8, the surface charge density of AC was negative and BPA deprotonated. So, the electrostatic interaction of repulsive was intensified [18].

3.3. Influence of contact time

For determination of the optimal contact time, the adsorption test was carried out at various times (0–180 min), and at constant pH (optimal value). The experimental conditions were using with 0.075 g of adsorbent in 50 mL solution and 50 mg $L^{-1}$of initial BPA concentration. Fig. 5 shows the amount of adsorbed BPA onto the activated carbons as a function of reaction time. As shown, the adsorption rate increased from 20.32 to 33.19 mg g^{-1} for MVAC, from 4.52 to 12.15 mg g^{-1} for CDAC and from 19.21 to 29.22 mg g^{-1} for CAC (reference AC) with an increasing contact time from 30 to 90 min. At the contact time higher than 90 min, the adsorption rate showed a negligible increase in the BPA adsorption in the three AC tested. Therefore, the amount of BPA adsorbed in the adsorbents is in a dynamic equilibrium with the molecules desorbed from the surface of the ACs in equilibrium time equal to 90 min. The higher adsorption value at the beginning of the process is attributed to the presence of a greater number of active sites in the adsorbent [25]. Arampatzidou and Deliyanni [14] reported an equilibrium time of 70 min for the adsorption of BPA onto AC prepared from potato peels.

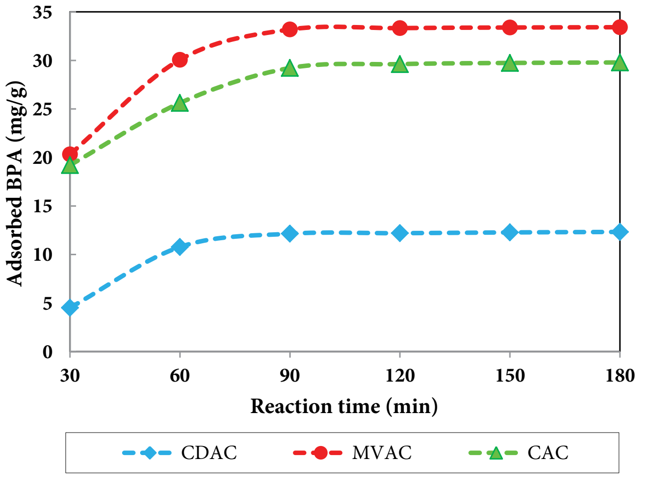

Fig. 5. Effect of contact time on the adsorption of BPA onto CDAC, MVAC and CAC.

3.4. Kinetic study

The study of adsorption kinetics is important for the prediction of the adsorption behavior of BPA onto the tested ACs. This describes the adsorbate uptake rate that controls the equilibrium time, the transfer of mass and the modeling of the sorption process [49]. Two sorption kinetic models were employed to the experimental data of the adsorption of BPA onto the tested ACs, including pseudo-first-order and pseudo-second-order kinetics. Table 1 shows the values of the adsorption kinetic parameters. The results show a good agreement between the obtained experimental findings and pseudo-second-order kinetic model. The correlation coefficients (R^2) of the pseudo-second-order model for the adsorption of BPA onto the three ACs were found to be higher than that of the pseudo-first-order kinetic model. On the other hand, the experimental q_e values ($q_{e,\mathrm{exp}}$) of 33.19 (MVAC), 29.22 (CAC) and 12.15 (CDAC) mg g^{-1} were closer to the calculated values ($q_{e,\mathrm{cal}}$) of 37.59 (MVAC), 33.33 (CAC) and 16.94 (CDAC) mg g^{-1} for the pseudo-second-order model. These results showed that the pseudo-second-order kinetic model sufficiently describes the adsorption kinetics of BPA on MVAC, CDAC and CAC (reference AC), in agreement with other studies [50]. Wirasnita et al. [15] reported that the adsorption of BPA on AC, prepared from the oil palm wastes, followed the pseudo-second-order kinetic model, and also Filippou et al. [50] reported that the adsorption of BPA in the magnetic AC followed the pseudo-second-order kinetic model.

Table 1
Parameters of the kinetic models for BPA adsorption onto CDAC, MVAC and CAC

Adsorbent	Pseudo-first-order model plot: $\ln(q_e/q_t)$ vs. Time			Pseudo-second-order model plot: (t/q_t) vs. time			$q_{e,\text{exp}}$
	K_1	q_e (mg g^{-1})	R^2	K_2	q_e (mg g^{-1})	R^2	
MVAC	0.052	55.25	0.96	0.0014	37.59	0.99	33.19
CAC	0.046	47.46	0.98	0.0016	33.33	0.99	29.22
CDAC	0.002	22.87	0.66	0.0011	16.94	0.89	12.15

3.5. Influence of adsorbent dosage

Fig. 6 shows the influence of adsorbent dose on the adsorption of BPA by varying the dose of the ACs (between 0.02 and 0.1 g) in a 50 mL aqueous solution, initial BPA concentration of 50 mg L^{-1}, the contact time of 90 min and pH 7. As shown in Fig. 6, when increasing the dose of the adsorbents from 0.02 to 0.1 g, the adsorption capacity of BPA decreased from 64.2 to 27.32 mg g^{-1} for MVAC, from 43.55 to 7.15 mg g^{-1} for CDAC and from 58.88 to 22.41 mg g^{-1} for CAC, respectively. In contrast, the opposite is observed for the removal efficiency; as the adsorbent dosage is increased from 0.02 to 0.1 g, the removal efficiency of BPA increased from 51% to 100% for MVAC, from 34.84% to 61.66% for CDAC, and from 47.12% to 89.64% for CAC, respectively. This implies that by increasing the dose of adsorbent, the number of adsorption sites increases, but the adsorption capacity (q_e) decreases. At high adsorbent doses, the available surface area increased and, subsequently, more adsorption sites are available for an efficient adsorption process [51]. As illustrated, by increasing the dose of adsorbent above 0.075 g, the rate of adsorption was continued at a slower rate and the removal of BPA has a negligible increase. Therefore, 0.075 g of adsorbent dosage was determined for implementation in subsequent experimental tests. These results show that MVAC adsorbent is better than CDAC and CAC adsorbents. Similar results were reported by Cherifi et al. [52]. These authors studied the methylene blue adsorption on AC, prepared from vegetable sponge of cylindrical loofa. It was reported that the removal of MB increased rapidly with increasing dosage of adsorbent to 0.4 g, then slowly increased with a further increase in adsorbent dose to 0.8 g and, thereafter, remain unchanged [52].

3.6. Influence of initial BPA concentration

Fig. 7 shows the amount of adsorbed BPA and BPA removal onto the surface of the ACs as a function of initial BPA concentration (25, 50, 75, 100 and 150 mg L^{-1}). The experimental conditions were at 0.075 g of adsorbent dosage, 50 mL solution, 90 min reaction time and pH 7. As can be observed, by increasing the concentration of BPA from 25 to 150 mg L^{-1}, the amount of adsorbed BPA by the three adsorbents was enhanced (from 16.4 to 74.75 mg g^{-1} for MVAC, from 6.35 to 33.2 mg g^{-1} for CDAC, and from 14.1 to 64.2 mg g^{-1} for CAC). This is attributed to the increase in the propulsion, to overcome resistance to mass transfer, between the aqueous solution and the ACs that occurs by increasing the initial BPA concentration [53,54]. Whereas, the percentage removal of BPA decreased from 99.84% to 74.75% for MVAC, from 69.06% to 33.2% for CDAC, and from 84.6% to 64.3% for CAC as the initial BPA concentration increased from 25 to 150 mg L^{-1}. This decrease in removal efficiency may be due to the lack of available active sites on the surface of the three adsorbents in high initial concentrations of BPA [55].

3.7. Isotherm study

The most widely used isotherm equations such as D–R, Freundlich and Langmuir isotherm models have been applied to model the isotherms in the present study [1,56]. The results of the isotherm parameters and the correlation coefficients

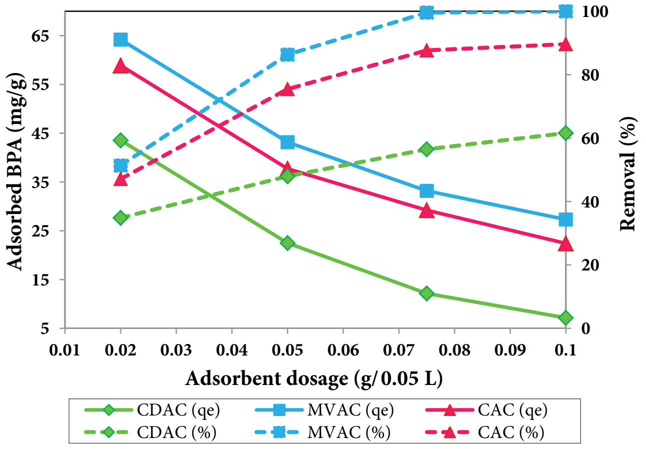

Fig. 6. Effect of adsorbent dosage on the amount of adsorbed BPA (mg g^{-1}) and removal efficiency of BPA by CAC, CDAC and MVAC.

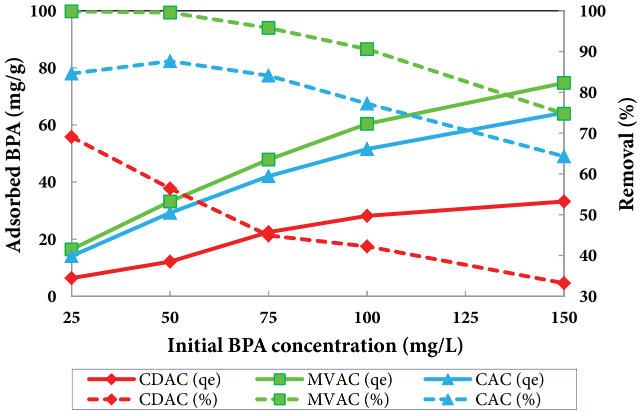

Fig. 7. Effect of initial BPA concentration on the adsorption capacity (mg g^{-1}) and removal efficiency of BPA by CAC, CDAC and MVAC.

(R^2) are provided in Table 2. Hence, the R^2 value obtained for Langmuir model was higher than that of the Freundlich and D-R models, which indicates that the Langmuir isotherm was better suited to the experimental data than other isotherm models, and the BPA adsorption onto prepared ACs well fitted with Langmuir model with homogeneous adsorption sites to be occupied by monolayer of BPA molecules [33]. Table 2 shows the maximum adsorption capacity ($q_{\max}$) of BPA by MVAC was higher than that of CDAC and CAC (used as reference AC). It has been observed that the adsorption of BPA on the ACs prepared from waste carbonaceous materials can be happened through physical adsorption. In a similar study, Wirasnita et al. [15] found that the adsorption of BPA onto AC (prepared from the oil palm wastes) can be well explained by a monolayer isotherm model [15]. Ozer et al. [57] found that the adsorption of MB onto AC was reasonably well represented by the Langmuir model.

3.8. Influence of solution ionic strength

The influence of the ionic strength on the adsorption of BPA onto CAC, CDAC and MVAC was studied at different $CaCl_2$ concentrations (20, 40, 60, 80 and 100 mg L^{-1} as Ca^{2+} ion). The experimental conditions used are as follows: adsorbent dosage 0.075 g, 50 mg L^{-1} initial BPA concentration, pH equal 7 and the reaction time 90 min. As shown in Fig. 8, negligible changes are observed in the removal of BPA by increasing the ionic strength of the solution from 0 to 100 mg L^{-1} as Ca^{2+} ion. This indicates that the tested ACs could be used efficiently to remove BPA from aqueous solutions of high ionic strength. These findings are similar to those reported by Nourmoradi et al. [43] and Li et al. [58], who examined different ionic strength solutions and found no significant effect on the removal of textile dye compounds (such as reactive red 24, MB and AO7) using ACs.

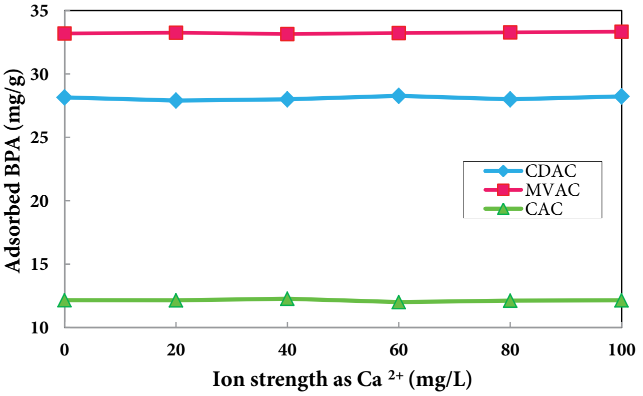

Fig. 8. Effect of ionic strength on BPA adsorption onto CDAC, MVAC and CAC.

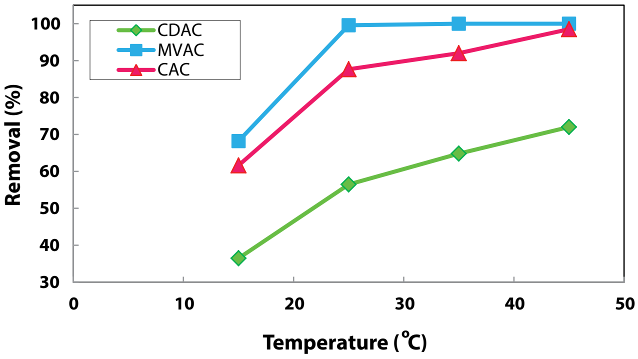

Fig. 9. Effect of temperature on BPA adsorption onto CDAC, MVAC and CAC.

3.9. Influence of temperature

The influence of the reaction temperature on the adsorption of BPA onto the tested ACs was investigated in temperature range from 15°C to 45°C. The experimental conditions were 50 mL of solution, 0.075 g of adsorbent, 50 mg L^{-1} of initial BPA concentration, 90 min for reaction time, pH 7 and the obtained results are presented in Fig. 9. These results indicate that with increasing temperature from 15°C to 25°C, the RE increased, but no significant increase in the RE was observed from the temperature range between 25°C and 45°C. Therefore, the maximum RE for all the adsorbents was observed at 25°C indicating that lower temperatures are more suitable for the removal of BPA. The decrease in temperature may increase the adsorption forces between the various BPA species and the active sites on the surface of the adsorbent as a result of the increase in the adsorption efficiency [59].

To indicate the parameters of thermodynamic such as enthalpy change (ΔH), entropy change (ΔS) and Gibbs free energy (ΔG), Eqs. (8)–(10) were used [45]. These parameters could determine the mechanism of adsorption process.

$$\Delta G = -RT\ln K \tag{8}$$

$$\Delta G = \Delta H - T\Delta S \tag{9}$$

$$\ln K = \left(\frac{\Delta S}{R}\right) - \left(\frac{\Delta H}{RT}\right) \tag{10}$$

where K is the equilibrium constant of Langmuir isotherm. The values of ΔG were calculated from Eq. (8), while the

Table 2
Langmuir, Freundlich and D–R isotherm parameters for the removal of BPA on CDAC, MVAC and CAC

Adsorbent	Langmuir isotherm plot: (C_e/q_e) vs. C_e			Freundlich isotherm plot: $\ln q_e$ vs. $\ln C_e$			D–R isotherm plot: $\ln q_e$ vs. ϵ^2		
	Q_m (mg g^{-1})	b (L mg^{-1})	R^2	K_f (L mg^{-1})	n	R^2	q_m (mg g^{-1})	E (kJ mol^{-1})	R^2
MVAC	98.03	0.67	0.99	33.11	4.25	0.72	57.39	5	0.90
CDAC	54.5	0.45	0.90	6.35	1.54	0.85	79.04	2.76	0.83
CAC	81.30	0.73	0.97	9.02	1.89	0.86	83.96	3.33	0.81

Table 3
Thermodynamic parameters for the removal of BPA onto CDAC, MVAC and CAC

Adsorbent	q_e (mg g^{-1})				ΔG (kJ mol^{-1})				ΔH	ΔS
	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	(kJ mol^{-1})	(J mol^{-1} K^{-1})
MVAC	22.73	33.19	33.33	33.33	–1.82	–13.48	–17.29	–20.67	22.64	78.82
CDAC	5.33	12.15	13.60	15.00	–1.05	–4.19	–5.07	–7.14	4.55	14.11
CAC	20.43	29.22	30.66	32.83	–1.13	–4.85	–6.11	–11.05	11.61	40.02

values of ΔH (kJ mol^{-1}) and ΔS (kJ mol^{-1}) of the sorption are evaluated from the slope and the intercept of linear plot between ln (K) vs. T^{-1}, respectively [14].

Table 3 demonstrates the thermodynamic parameters. The negative values of ΔG, inside the temperature range studied (15°C–45°C), indicate that the adsorption of the BPA by the prepared ACs is a physical adsorption process, spontaneous process and feasible [14]. The positive values of ΔH and ΔS show that the adsorption of BPA by the adsorbents is of endothermic nature [14,43,45]. The positive values of ΔS change indicate increased randomness at solid–solution relation during the adsorption of BPA on tested ACs [45]. In this study, the values of the ΔG for the BPA adsorption by the three prepared ACs, was between 1 and 20 kJ mol^{-1} which indicates that the physisorption is the dominant mechanism of adsorption [60].

Balarak [45] showed that the BPA adsorption on barley husk was spontaneous (by the negative values of the Gibbs free energy) and of endothermic nature (by the positive values of ΔH change). The results of the study of Liu et al. [18] indicated that BPA adsorption by activated carbon samples mostly followed the mechanism of physical adsorption (exothermic).

3.10. Comparison of other adsorbents with MVAC in the BPA adsorption

A comparative study was conducted between MVAC and other adsorbents to evaluate the adsorption capacity of the as-prepared MVAC as an efficient adsorbent to remove BPA from aqueous samples. The maximum adsorption capacity of BPA on MVAC is 33.19 mg g^{-1}, which is higher than that of different adsorbents used in the literature [61–63]. This indicates that the MVAC has a relatively good adsorption capacity for BPA adsorption, even when compared with the CAC used in this study (29.22 mg g^{-1}). Although Wirasnita et al. [15] and Bautista-Toledo et al. [40] reported a greater capacity for adsorption of BPA with ACs prepared in their work than those found in this study.

4. Conclusions

This research examined the BPA removal, as an endocrine disruptor compound, from aqueous samples using AC prepared from CD wastes (a synthetic carbonaceous precursor) and MV (as natural carbonaceous precursor) which were compared with the commercial AC. The morphology of the adsorbents showed that the MVAC was more porous than the CDAC and the commercial AC. The results showed that MVAC had higher adsorption capacity (33.19 mg g^{-1}) than CDAC (12.15 mg g^{-1}) and CAC (29.22 mg g^{-1}). The amount of BPA adsorbed per unit mass of the prepared ACs increased with increasing the dosage of the three adsorbents, the initial BPA concentration and the temperature. The pseudo-second-order model and the Langmuir isotherm were adjusted to the experimental data of all tested adsorbents. The thermodynamic study showed that the BPA adsorption by the adsorbents is feasible, spontaneous and proceeds with the mechanism of physisorption. In general, MVAC and CDAC were efficient adsorbents with a favorable adsorption capacity for the treatment of solutions contaminated with BPA.

Conflict of interest

The authors confirm that this article content has no conflict of interest.

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