

Article

Adsorptive Behavior of an Activated Carbon for Bisphenol A Removal in Single and Binary (Bisphenol A—Heavy Metal) Solutions

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Received: 29 June 2020; Accepted: 28 July 2020; Published: 30 July 2020



Abstract: Bisphenol A (BPA) is an extensively produced and consumed chemical in the world. Due to its widespread use, contamination by this pollutant has increased in recent years, reaching a critical environmental point. This work investigates the feasibility of bisphenol A adsorption from industrial wastewater solutions, testing the reduction of bisphenol A in synthetic solutions by a commercial activated carbon, AC-40, in batch mode. Besides, mixtures of bisphenol A and different heavy metal cations were also studied. So far, no works have reported a complete study about bisphenol A removal by this activated carbon including the use of this material to remove BPA in the presence of metal cations. First, adsorption experiments were performed in batch changing pH, dose of adsorbent, initial bisphenol A concentration and contact time. Results showed greater retention of bisphenol A by increasing the acidity of the medium. Further, the percentage of bisphenol A adsorbed increased with increasing contact time. The selected conditions for the rest of the experiments were pH 5 and a contact time of 48 h. In addition, an increase in retention of bisphenol A when the dose of adsorbent increased was observed. Then, specific experiments were carried out to define the kinetics and the adsorption isotherm. Equilibrium data were adequately fitted to a Langmuir isotherm and the kinetics data fitted well to the pseudo-second-order model. The maximum adsorption capacity provided by Langmuir model was 94.34 mg/g. Finally, the effect of the presence of other heavy metals in water solution on the adsorption of bisphenol A was analyzed. Binary tests revealed competition between the adsorbates and a significant selectivity toward bisphenol A. Finally, the study of the adsorption performance in three consecutive adsorption–desorption cycles showed efficiencies higher than 90% in all cycles, indicating that the activated carbon has good reusability.

Keywords: activated carbon; adsorption; bisphenol A; equilibrium; heavy metals

1. Introduction

Bisphenol A (BPA) is recognized as a dangerous endocrine disruptor. It causes a number of hazardous effects on living beings such as fertility damage and fetal development problems [1–3]. It has been extensively used as a crude material in the industrial production of epoxy resins and polycarbonate plastics, which accounted for nearly 64% of BPA demand in 2018. These polycarbonate resins are mainly consumed in the electronics, construction and automotive industries. In terms of country distribution, high demand is concentrated in emerging consumers such as China, India,

Southeast Asia and Brazil. China is the largest player in the BPA industry, accounting for half of BPA's global consumption. Western Europe and the United States are the other important players in the BPA market [4].

As a consequence, concentrations of BPA are present both in surface waters as well as industrial wastewaters [5–7]. For all those reasons, novelty alternatives have been proposed for its removal from wastewater during recent years [8]. BPA is present in various types of water at different concentrations. For instance, it has a concentration of 17.2 mg/L in hazardous waste landfill leachate [9,10], 12 µg/L in stream water [11], between 3.5 and 59.8 ng/L in drinking water [7] and around 100 mg/L in effluents from polycarbonate factories [6,12].

Due to its high industrial yield and toxicological effect, the elimination of BPA from water solutions is of great importance and it has been listed as a priority pollutant in water treatment [13]. Several separation techniques such as adsorption, advanced oxidation, membrane separation, phytoremediation and photocatalysis, among others, have been applied to efficiently remove BPA from water effluents [14,15]. Particularly, among wastewater treatment technologies, phytoremediation is arousing attention because it is a promising ecologically friendly alternative to remove BPA from contaminated soil and water systems. For example, Phouthavong-Murphy et al. [9] investigated the potential of two United States native switchgrass varieties for BPA removal and found good BPA removals over approximately three months. Further, advanced oxidation processes are interesting due to their good performance in the degradation of BPA and other micropollutants. Recently, Luo et al. [16] studied the rapid removal of BPA and other organic micropollutants by heterogeneous peroxymonosulfate catalysis and reported that BPA had the highest removal efficiency at pH 9.0, almost 100% removal. Other researchers [17] integrated advanced oxidation processes with membrane filtration to form a catalytic membrane that demonstrated exceptional efficiency for instantaneous degradation of BPA. Moreover, the development of cheap and eco-friendly, effective and rapid remediation technologies for the removal of BPA has increased the investigations based on the use of transition metal-based nanomaterials for photocatalysis. For example, Rani et al. [18] synthesized and examined green zinc and cobalt ferrites nanoparticles for the degradation of BPA from water under direct sunlight. Results showed very good efficiencies, similar to those found with ZnO, Co₃O₄ and Fe₂O₃ nanoparticles.

Even novel technologies are being investigated with very good results: one of the most favorable methods is traditional adsorption onto carbonaceous materials [19]. Particularly, activated carbons have good characteristics to work as an adsorbent: high porosity and surface area (around 3000 m²/g), high degree of surface reactivity and diverse characteristics of surface chemistry [20,21]. Due to these characteristics, activated carbons have been used for many different applications: as adsorbents, catalysts or catalyst supports. They have been tested as a cleaning material for removing pollutants from gaseous or liquid phases and the purification or recovery of chemicals [22,23]. Adsorption on activated carbon is one of the first water treatments to have been used [24,25] and it is recognized by the United States Environmental Protection Agency (US EPA) as one of the best methods for removing organic and inorganic compounds from water for human consumption. The aqueous-phase adsorption of both organic and inorganic compounds has been a very important application of activated carbons. In fact, around 80% of the world production of activated carbons is used in liquid-phase applications [26,27].

The adsorption mechanism of activated carbons is due to interactions (electrostatic or non-electrostatic) between the carbon surface and the adsorbate. The nature of these interactions, which can be attractive or repulsive, depends on the: (a) charge density of the carbon surface, (b) chemical characteristics of the adsorbate and (c) ionic strength of the solution. Non-electrostatic interactions are always attractive and can include: (a) van der Waals forces, (b) hydrophobic interactions and (c) hydrogen bonding.

Activated carbons can be manufactured by physical or chemical activation processes using a wide range of raw materials. Physical activation uses high temperature for the carbonization of the original

material in absence of oxygen (usually CO₂ or steam atmosphere). During chemical activation, the material is impregnated with a chemical agent prior to its carbonization. One of the most frequent chemical agents extensively used for the production of activated carbons in industry is H₃PO₄ [28,29].

Adsorptive removal of BPA from wastewater solutions by carbon-based materials, such as activated carbon, has been extensively studied [30–35]. For example, Juhola et al. [31] prepared a biomass-based adsorbent with a BPA adsorption capacity of 41.5 mg/g. Pan et al. [33] investigated the BPA adsorption on carbon nanomaterials that showed a very good potential for the removal of BPA of waters. Redding et al. [34] studied the adsorption of BPA on various modified lignite carbons. Specifically, the work demonstrated that these modified carbons prepared using high-temperature steam or methane/steam offered better behavior than conventional activated carbons. However, there are few works available focusing on the BPA removal from water when other types of contaminants are also present in the water [36–39]. BPA is a high-volume industrial chemical used in the production of epoxy resins and polycarbonate plastics. Further, different additives are added in these materials to improve the performance of the plastics. The chemical substances used as additives can be very different depending on the objective. The additives can include metal-containing additives such as heavy metals as cadmium, copper, lead or zinc, among others [40]. Heavy metals are another important type of pollutants that have been found in waters. Some researchers have analyzed the simultaneous adsorption/biosorption of these priority pollutants (heavy metals and organic pollutants) [41–43]. The influence of heavy metals could enhance the removal of BPA or perhaps they could compete with BPA for the available active sites and decrease the removal of BPA when the removal of BPA and heavy metals is carried simultaneously.

Hence, one of the objectives of this investigation is to study the interactions between some of the most common heavy metals in wastewater environments and BPA during adsorption onto activated carbon (AC-40 commercial sample). First, the single adsorption of BPA on AC-40 activated carbon was investigated. The main factors influencing the adsorption process were analyzed. Then, the simultaneous adsorption of BPA and heavy metal cations on AC-40 activated carbon was performed in order to know the type of adsorption (competitive or cooperative) which occurs between BPA and these metal cations. Finally, desorption of BPA in three consecutive cycles was analyzed. Although the study has been performed using a commercial activated carbon, so no novel findings about the material are given, there are not a wide range of research works in the literature about the use of AC-40 for the removal of BPA from wastewaters including (1) the effect of the main parameters, (2) a kinetics study, (3) an isotherms study, (4) simultaneous adsorption of BPA and heavy metals and (5) desorption. The authors consider that specific complete studies about the removal of emergent pollutants with the current technology (adsorption on activated carbons) can be interesting since they increase the knowledge in this field and can be applied in the industrial wastewater sector.

2. Materials and Methods

2.1. Adsorbent

Activated carbon trademark CECA S.L., known as AC-40, was used as an adsorbent in all the experiments. It is an extruded, thermally activated carbon from bituminous coal with a high surface area (1200 m²/g). The main features of this adsorbent have been determined by Méndez Díaz et al. [44], Abdel Daiem et al. [45] and Velo-Gala et al. [46]. Table 1 shows a summary of the properties.

Table 1. Main physic-chemical characteristics of activated carbon AC-40.

Median particle diameter (MPD) (mm)	3
Density (g/L)	450
Moisture content (%)	4
Specific surface area (m ² /g)	1201
Pore volumes (mean widths of micropores 0.71 nm) (cm ³ /g)	0.406
Pore volumes (diameter between 6.6 and 50 nm) (cm ³ /g)	0.046
Pore volumes (diameter > 50 nm) (cm ³ /g)	0.409
Concentration of acidic groups (µeq/g)	907.7
Concentration of basic groups (µeq/g)	96.0
Point of zero charge (pH _{pzc})	7.5
%C	68.97
%H	1.03
%N	0.45
%S	1.02
%O	20.23
%Ash	8.30

2.2. Reagents

BPA, C₁₅H₁₆O₁₂, with 100% purity of Sigma-Aldrich company was used. Solutions of BPA were prepared by dissolving BPA in ultrapure water obtained from Milli-Q[®] equipment (Millipore). In adsorption tests conducted with mixtures of BPA and metal cations, copper (II) nitrate, Cu(NO₃)₂·3H₂O, lead (II) nitrate, Pb(NO₃)₂, nickel (II) nitrate, Ni(NO₃)₂·6H₂O, and cadmium (II) nitrate, Cd(NO₃)₂·4H₂O, were used. All of them had purity higher than 98% and they were supplied by Panreac Quimica S.A. company.

2.3. Methodology

All adsorption experiments were conducted in batch mode in a stirred tank. BPA solutions were prepared in ultrapure water using a mild ultrasonic bath with a temperature of approximately 50 °C (323 K).

In the preparation of mixtures of BPA and metal cations, the proper amount of metal reagent was added to the previously prepared solution of BPA.

Each solution was located in a jacketed reactor with a capacity of 250 mL and maintained at a constant temperature (298 K) with stirring throughout the test by using a magnetic stirrer. When the operation time finished, a liquid sample was taken from the reactor by using an automated pipette. Moreover, a sample of the original solution was taken. The concentrations of initial and final contaminants were determined. Consequently, it was possible to determine the percentage of contaminant removed by the activated carbon.

The effect of main experimental conditions such as adsorbent dose, pH, contact time and initial BPA concentration was studied changing the variables between the following ranges: adsorbent dose (0.1–1.0 g/L), pH (3–10), contact time (0.5–48 h) and initial BPA concentration (1–160 mg/L) at a constant temperature of 25 °C. The desired value of pH was obtained by adding HCl solution (0.1 M) or NaOH solution (0.1 M) to the BPA solution. The real initial concentrations of BPA obtained in a pH range of 3–10 were measured in absence of the adsorbent.

To test the effect of pH on the adsorption process of BPA with activated carbon AC-40, an initial concentration of BPA of 20 mg/L, a contact time of 48 h and an adsorbent concentration of 0.5 g/L were selected. The pH of the medium was changed from 3 to 10. The temperature remained constant at 25 °C. Then, experiments were performed at pH values between 4 and 8 at different contact times in order to analyze the influence of contact time on the BPA removal efficiency.

To determine the minimum amount of activated carbon necessary to achieve maximum removal of BPA, experiments were carried out with a BPA initial concentration of 20 mg/L, three contact times

5.5, 22 and 48 h and a pH of 5, changing the concentration of the adsorbent from 0.1 to 1 g/L. The temperature remained constant during the tests at 25 °C (298 K).

The initial concentration of BPA is one of the most influential parameters in determining the contaminant removal on activated carbon from the aqueous solution. Experiments were carried out to test the effect of the BPA concentration in the adsorption process. A pH value of 5 with an adsorbent concentration of 0.5 g/L, and a contact time of 48 h were selected. The initial concentration of BPA was changed from 1 to 160 mg/L.

To analyze the kinetics of BPA adsorption, experiments were performed with different contact times (until a contact time of 48 h), at pH = 5 and a BPA initial concentration of 20 mg/L, with an adsorbent dose of 0.5 g/L and at a temperature of 298 K.

BPA adsorption isotherms of the activated carbon AC-40 were carried out at pH 5 and a temperature of 298 K. For this purpose, 100 mL of solutions containing different concentrations (1–160 mg/L) of BPA and 0.05 g of activated carbon were introduced into jacketed reactors, which were capped with flexible polyethylene (LDPE) film to avoid evaporation, prevent contamination and protect the solid–liquid system inside the reactor. Adsorption equilibrium was established when the concentration of BPA did not change (for 3 days). The BPA removal efficiency and the amount of BPA adsorbed by gram of activated carbon were calculated by the Equations (1) and (2), respectively:

$$\% \text{ Removal efficiency} = \frac{(C_i - C_f)}{C_i} \times 100 \quad (1)$$

$$q = \frac{(C_i - C_f)}{m} \times V \quad (2)$$

where C_i and C_f are the initial and final concentrations of BPA in solution, respectively, mg/L, q is the amount of BPA adsorbed by gram of adsorbent, mg/g, m is the mass of adsorbent, g, and V is the volume of the solution, L.

The procedure to obtain the data of adsorption of BPA on the activated carbon in the presence of metal cations was similar to methods to obtain adsorption results of single BPA systems. The only difference was the addition of a specific content of some metal cations to each BPA solution. Tests were performed with a concentration of 20 mg/L for both BPA and each metal cation, with an activated carbon dose of 0.5 g/L, during a contact time of 48 h and at pH value of 5. When equilibrium was reached, the BPA and different metal cations concentrations were analyzed.

The determination of the content of BPA was performed by ultraviolet–visible (UV–Vis) spectrophotometry, using the spectrophotometer model Genesys 6. The BPA concentration was determined by absorbance measurements at 277.5 nm. All the samples were collected in triplicate to further ensure precision. Samples used filled three quarters of the 1 cm cell used in the UV–Vis spectrophotometer. This amounted to approximately 1.5 mL of sample. In binary tests, the determination of metal content was performed by atomic absorption spectrophotometry, using the spectrophotometer model Perkin-Elmer AAnalyst 200.

Desorption of the BPA adsorbed onto the AC-40 was studied in a batch system. First, adsorption was performed at pH 5 and an initial BPA concentration of 20 mg/L with a dose of AC-40 of 0.5 g/L. Then, desorption was performed under the following desorption conditions: use of a mixture of methanol/acetic acid of 4:1 (v/v), a dose of AC-40 of 0.5 g/L, operation time of 48 h and temperature of 25 °C, and when the operation time was finished, the supernatant was separated and analyzed by ultraviolet–visible spectrophotometry at 277.5 nm.

3. Results and Discussions

3.1. Influence of pH and Contact Time

The pH is one of the main parameters that control the removal of compounds present in aqueous solutions using solid adsorbents. According to several authors, pH can change the availability and

characteristics of the pollutants in solution and it can modify the chemical state of the functional groups that are responsible for adsorption [47–49].

Some experiments were performed to test the effect of pH on the adsorption process of BPA with activated carbon AC-40. Figure 1 shows the results.

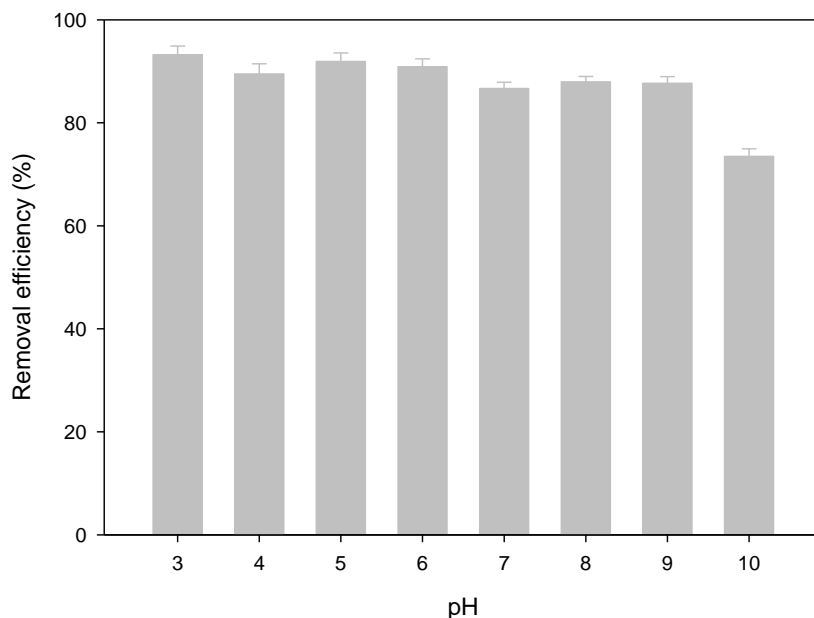


Figure 1. Bisphenol A (BPA) removal efficiency of activated carbon AC-40 according to the pH of the medium.

Data showed that pH did not significantly affect BPA adsorption. However, a slight tendency to obtain greater retention by increasing the acidity of the medium was observed. It could be because the zero point of charge was 7.5. The zero point of charge is the pH at which the surface of the adsorbent is globally neutral. Besides, it is a fundamental parameter of a material surface, governing the adsorbent–adsorbate interactions. Below this point, the surface is positively charged, otherwise it is negatively charged. Therefore, the CA surface was predominantly positive at pH values lower than 7.5. In contrast, negative charges appeared on the surface at pH values higher than 7.5 due to the dissociation of functional groups. In addition, BPA showed different states of equilibrium in an aqueous solution depending on the pH. BPA could be found in its molecular form at a pH less than 8. Deprotonation of the bisphenolate monoanion occurred at a pH of 8 [49], reaching a considerable concentration at pH 9 [50]. Therefore, electrostatic interactions between the negative charge of the surface of the activated carbon and bisphenolate anion could lead to a decrease in the overall adsorption of BPA. However, other interactions could also influence the adsorption rather than electrostatic forces, making the effect of surface charge less intense.

These results were in good agreement with those reported by other researchers. Thus, Wang and Xiao [51] indicated that the adsorption of BPA with four different activated carbons decreased at values of pH higher than 9. Chang et al. [52] found that the pH had an important role in the retention process of BPA with an activated carbon prepared from rice straw. Soni and Padmaja [53] also studied the adsorption of BPA on activated carbon prepared from palm shell. They found that at pH values below 9, the adsorption of BPA was not influenced by the pH of the medium. Bohdziewicz and Liszczyk [54] found a decrease in the adsorption of BPA by commercial activated carbon at pH values higher than or equal to 10. However, at pH values below 10, the adsorption capacity was not affected.

Figure 2 shows the BPA removal efficiency of activated carbon with different contact times. The removal efficiency increased with increasing contact time. At pH 5, the amount adsorbed increased quickly in the first 0.5 h of contact. Then, at 22 h, an adsorption percentage of 80% was reached. If

the process continued until 48 h, the adsorbed percentage would increase approximately up to 91%. Finally, at 72 h of operating time, the removal of BPA reached a value of 92% (a very low change with respect to 48 h). At other pH values, as pH 7 or pH 8, the adsorption was performed in a more progressive way. However, the change between 48 and 72 h was also low.

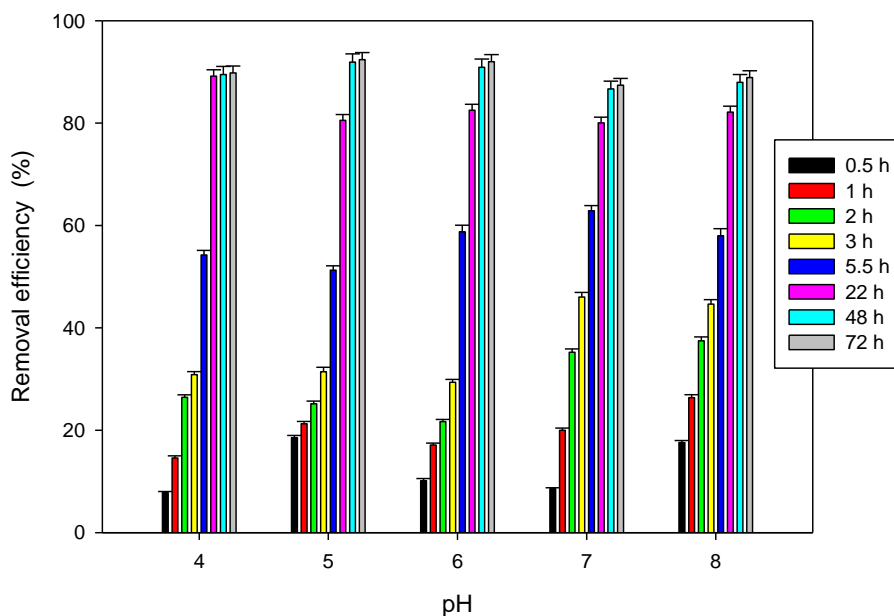


Figure 2. BPA removal efficiency of activated carbon AC-40 depending on pH at different contact times.

Considering the results obtained, from the removal efficiency point of view, a contact time of 48 h and a pH of 5 were selected for the rest of the experiments. There were not important differences in the pH range of 4 and 8. An adjustment by addition of acidic/basic chemicals could represent an important part of the cost of the wastewater treatment process and for that reason, an economic evaluation could indicate other pH values as a better option.

3.2. Influence of Adsorbent Dosage

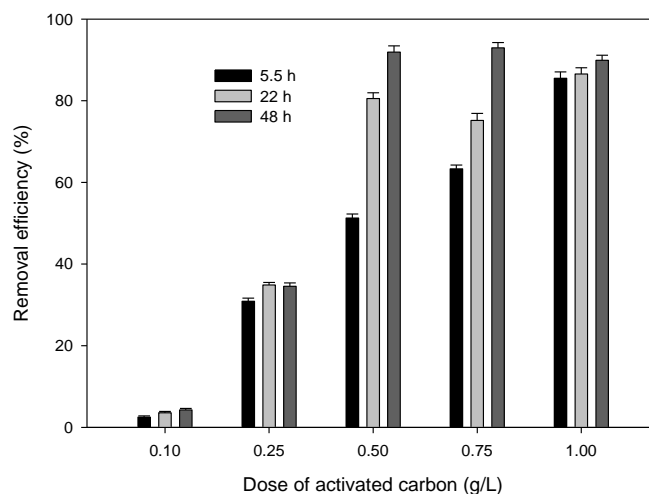
Figure 3 shows the results of tests performed to determine the minimum dose of activated carbon necessary to achieve maximum removal of BPA.

As the dosage of activated carbon increased, the BPA removal efficiency from solutions increased for all the contact times. However, differences between 0.5, 0.75 and 1 g/L were very slight for long contact times. For example, BPA removals (%) of 92%, 93% and 90% were achieved with adsorbent dosages of 0.5, 0.75 and 1 g/L, respectively, for a contact time of 48 h. However, great differences were observed at very short times. For example, very high adsorption attained at 5.5 h when using 1 g of adsorbent per L. With regard to the amount of BPA adsorbed by gram of activated carbon, in general, it was higher for a dose of 0.5 g/L. Above this adsorbent dose, a decrease in the uptake of the BPA was observed. In conclusion, if time was not considered, the use of an adsorbent dose of 0.5 g/L was recommended since at 22 h of operating time, very good BPA removals were achieved.

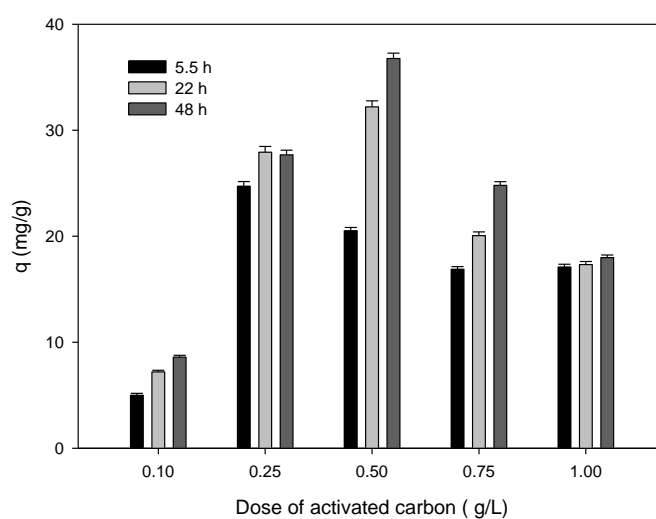
These results agreed with those obtained by numerous authors. Thus, Bautista-Toledo et al. [37] found an increase in the retention of BPA when 0.05 g of activated carbon was used for a period of 24 h in a BPA solution of 175 mg/L. Liu et al. [11] reported an increase in retention of BPA with two carbons activated with nitric acid that were selectively modified. They obtained maximum adsorption of BPA (432 mg/g) when changing the amount of activated carbon used. Tsai et al. [55] detected an increase in retention of BPA when the dose of adsorbent increased. This occurred because the number of adsorption sites increased in parallel with the increase in the dose of the adsorbent, when the

concentration of BPA was small (<20 mg/L). If the concentration was too high, the adsorbent would reach saturation quickly. As a result, it would stop contaminant retention.

According to the results, a concentration of 0.5 g/L of activated carbon was selected, since at this concentration the minimum amount of adsorbent is used with good adsorption removal percentages (if enough contact time is maintained).



(a)

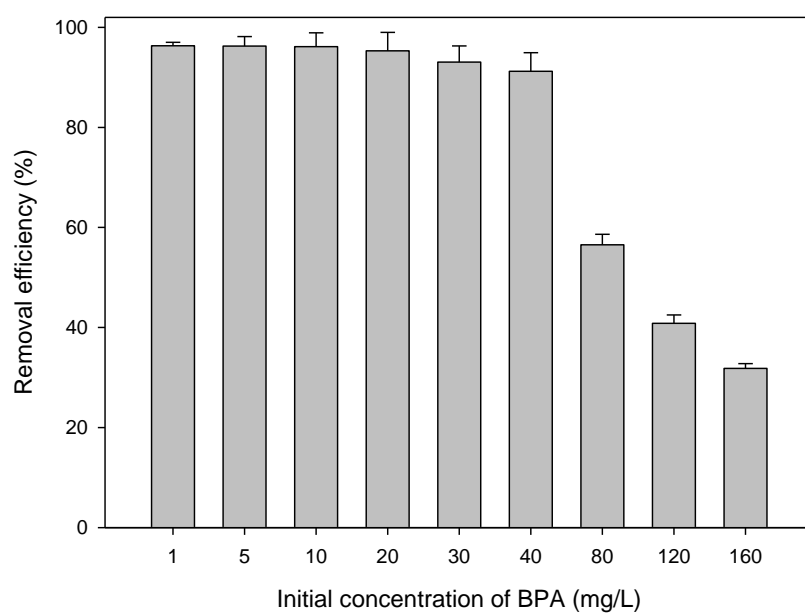


(b)

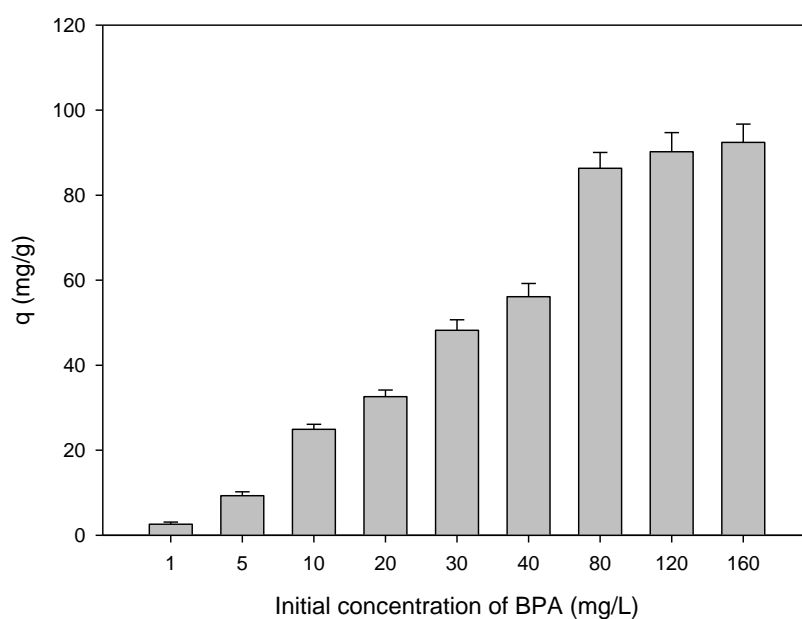
Figure 3. (a) BPA removal efficiency and (b) amount of BPA adsorbed by gram of activated carbon depending on adsorbent dosage at different contact times.

3.3. Influence of the Initial Concentration of BPA

Figure 4 shows the results of the influence of the initial concentration of BPA on its removal from the aqueous solution by AC-40.



(a)



(b)

Figure 4. (a) BPA removal efficiency and (b) amount of BPA adsorbed by gram of activated carbon depending on initial concentration of BPA.

It should be noted that BPA concentration did not influence BPA removal (%) for concentration values below 40 mg/L because the active sites on the adsorbent surface are not saturated. However, when the concentration was higher than 40 mg/L, a significant decrease was observed in the adsorption percentage. In this sense, at concentrations of BPA below 80 mg/L, the retention was higher because the pores of the carbon surface were occupied almost entirely. However, when the concentration of BPA was too high, the carbon surface underwent a supersaturation of its pores. As a consequence, a fraction of BPA was not retained by the adsorbent. These results were in agreement with those obtained by numerous authors. Thus, Bautista-Toledo et al. [37] obtained the same results in several studies with different concentrations of BPA and activated carbon. With regard to the amount of BPA removed expressed in mg/g of adsorbent (Figure 4b), it increased from 2.6 to 92.4 mg/g with an increase in the

initial concentration from 1 to 160 mg/L. The main reason for this important increase is the increase in the mass transfer driving force [56].

Based on the results obtained, an initial concentration of 20 mg/L was selected as the initial concentration of BPA for the rest of the experiments.

3.4. Kinetic Study

A kinetics study in adsorption allows the determination of the BPA removal rate from the aqueous medium. This study provides a basis for understanding the mechanism that controls the process and it is essential to select the optimum operating conditions in the system designed for the effluent treatment with this type of pollutant [57,58].

Experiments were performed with a total contact time of 48 h, pH = 5, a BPA initial concentration of 20 mg/L, an adsorbent concentration of 0.5 g/L and a temperature of 298 K. Figure 5 shows the values of q (mg retained/g sorbent) versus time.

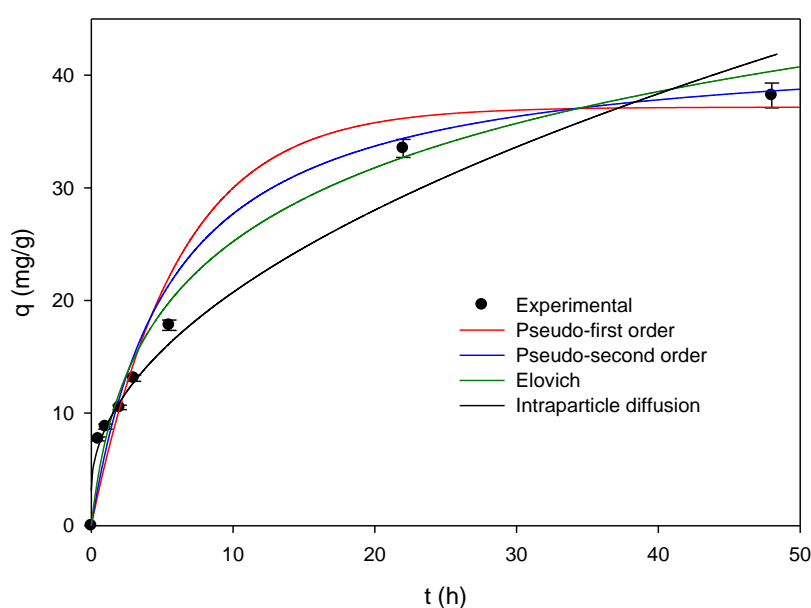


Figure 5. Variation in the adsorption capacity of BPA by activated carbon depending on the contact time and adjustment results of the four kinetic models.

These experimental results were fitted by a nonlinear regression to four kinetic models: pseudo-first-order, pseudo-second-order, Elovich and intraparticle diffusion models. Table 2 shows the representative equations of the models and their parameters. Likewise, Table 3 shows the results obtained when experimental data were fitted to the four selected kinetics models.

The pseudo-first- and pseudo-second-order models reproduced very well the experimental results. However, the intraparticle diffusion model showed a low R^2 value. Specially, the good correlation coefficients (R^2) of the pseudo-second-order model (it presented the highest R^2 value) and the similarity of the experimental and predicted adsorption capacity of this model indicated that the adsorption of BPA by activated carbon could be chemical adsorption, which is consistent with the results of Section 3.1. previously presented. In addition, the retention process was faster at first, but the equilibrium was not reached until 48 h of contact time. This indicates that, although adsorption happened quickly, the process subsequently proceeded more slowly until equilibrium was reached.

Other authors found similar results in their studies about BPA removal by activated carbons. Chang et al. [52] used an activated carbon obtained from rice straw agricultural waste. They found that the pseudo-second-order model fitted the experimental adsorption data better. Wang and Xiao [51] studied the adsorption of BPA by activated carbon reporting that the adsorption process followed the

first-order kinetics. Soni and Padmaja [53] reported that the kinetic data of adsorption of BPA onto palm shell activated carbon could be fitted well by a pseudo-second-order kinetic model. Tang et al. [59] showed that the kinetics of BPA removal onto activated carbon-alginate beads with cetyltrimethyl ammonium bromide fitted good to the pseudo-first-order, pseudo-second-order and Elovich models.

Table 2. Equations and parameters representative of the four selected kinetic models.

Model	Equation	Parameters
Pseudo-first-order	$q = q_e (1 - e^{-k_1 t})$	q_e adsorption capacity, mg/g k_1 kinetic constant of the pseudo-first-order, h^{-1}
Pseudo-second-order	$q = \frac{t}{\frac{1}{k_2 \times q_e^2} + \frac{t}{q_e}}$	q_e adsorption capacity, mg/g k_2 kinetic constant of the pseudo-second-order, g/mg·h
Elovich	$q = \frac{1}{b} \ln(a \times b) + \frac{1}{b} \ln(t)$	a velocity of initial adsorption, mg/g·h b area of the occupied surface, g/mg
Intraparticle	$q = k_p \times t^{1/2} + C$	k_p the intraparticle diffusion rate constant (mg/(g·min ^{1/2})) C constant of the intraparticle diffusion model (mg/g)

Table 3. Adjustment results of the experimental data to the four selected kinetic models.

Model	Parameters		
Pseudo-first-order	$k_1 = 0.164$	$q_e = 37.17$	$r^2 = 0.995$
Pseudo-second-order	$k_2 = 0.004$	$q_e = 43.06$	$r^2 = 1.000$
Elovich	$a = 11.325$	$B = 0.099$	$r^2 = 0.992$
Intraparticle diffusion	$k_p = 5.577$	$C = 3.082$	$r^2 = 0.964$

3.5. Equilibrium Study

Figure 6 shows the retention capacity of BPA, q_e (mg retained/g of sorbent when equilibrium is reached), versus the equilibrium concentration of BPA in the liquid phase, C_e .

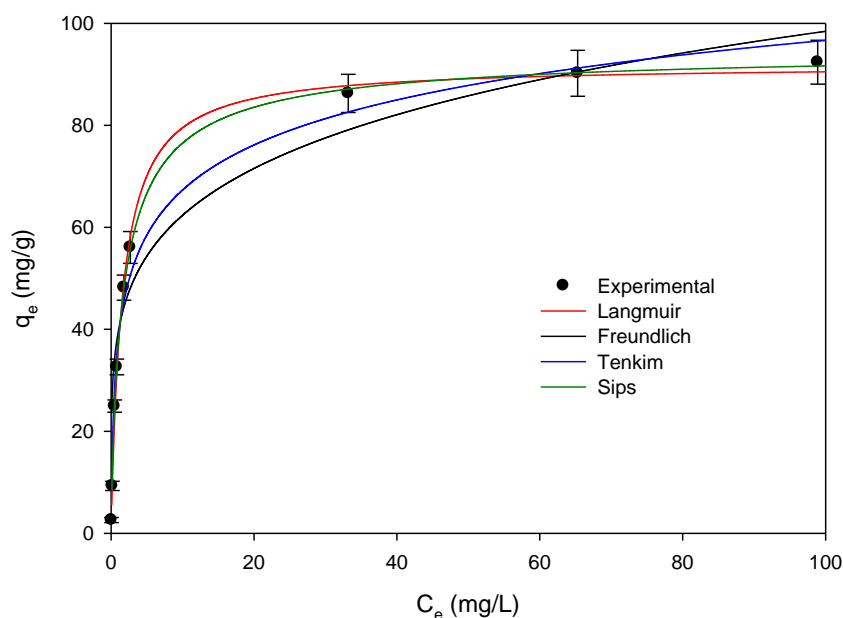


Figure 6. Equilibrium adsorption of BPA with activated carbon. Experimental results and adjustment to the selected models.

Different types of adsorption isotherms were found in the literature [51,60,61]. The isotherm obtained in this work was type I. The type I isotherm is monolayer adsorption easily explained using a Langmuir isotherm. Examples of type I adsorption isotherms are given by microporous solids such

as activated carbon (material used in this study). It is also observed that the maximum adsorption capacity was close to 92 mg/g, and it was reached when the initial concentration of BPA was 160 mg/L.

In order to study the equilibrium of the process, these results have been adjusted to different models which are shown in Table 4. Further, Table 5 shows the fitting results and the parameters of the models obtained. Figure 7 represents the isotherm obtained with the models used.

The Langmuir and Sips models were the best in reproducing the experimental results, as the value of the parameter n of the Sips isotherm was equal to the unit, which indicated that this model tended to be a Langmuir isotherm. This result indicates that adsorption is monolayer on the active sites of the adsorbent that has a homogeneous surface and molecules do not interact with each other. Likewise, the value of the maximum adsorption obtained with the Langmuir and Sips models was very close to the result obtained experimentally. These findings were similar to those obtained by other authors [52–54]. However, maximum capacity values of adsorption of AC-40 activated carbon were higher than those reported in the literature [62–65].

Finally, although adsorption processes performed in batch systems applied to the decontamination of wastewater are more frequent to determine the maximum adsorption capacity as the maximum amount of chemical adsorbed onto the adsorbent by mass of the adsorbent, in this work, the partition coefficients at the studied concentrations were also determined as the ratio of the concentration of BPA in solid phase (AC-40) to the concentration in liquid phase when the two concentrations are at equilibrium. Table 6 shows the partition coefficient as a function of the initial concentration of BPA. Data showed that the degree of partitioning varied with concentration. The linear partition coefficient was about 50–52 L/g (the amount adsorbed versus its solution concentration) at initial BPA concentrations lower or equal than 10 mg/L. This can be observed in Figure 6 since the first experimental points were plotted in a linear trend. However, as the initial concentration of BPA increased, the linear partition coefficient decreased until approximately 1 L/g at 160 mg/L. It also can be observed in the change of the slope of the isotherm (Figure 6) as the BPA concentration was increased. In adsorption studies, when an L-type plot (isotherm) is obtained, the q_m coefficient is a convenient and reliable parameter to be applied for adsorption capacities assessment better than the partition coefficient.

Table 4. Representative equations and parameters of the four selected models for the study of the equilibrium process.

Model	Equation	Parameters
Langmuir	$q_e = \frac{bq_m C_e}{1+b C_e}$	q_m maximum adsorption capacity, mg/g b constant related to the affinity of the adsorbent for the adsorbate
Freundlich	$q_e = K_F \times C_e^{\frac{1}{n}}$	K_F equilibrium constant, (mg/g)·(L/mg) ^{1/n} n constant related to the affinity between the adsorbent and the adsorbate
Sips	$q_e = \frac{q_m b C_e^{1/n}}{1+b C_e^{1/n}}$	q_m maximum adsorption capacity, mg/g b constant related to the affinity of the adsorbent for the adsorbate n parameter characterizing the system's heterogeneity
Temkin	$q_e = \frac{RT}{b} \ln(A_T C_e)$	A_T constant of union of the equilibrium (L/g), b Temkin constant, B constant related to the heat of adsorption (J/mol), $B = \frac{RT}{b}$

Table 5. Adjustment results of the experimental data to the four selected isotherm models.

Model	Parameters		
Langmuir	$q_m = 91.90$	$b = 0.64$	$r^2 = 0.996$
Freundlich	$K_F = 39.49$	$n = 5.04$	$r^2 = 0.941$
Sips	$q_m = 95.06$	$b = 0.632$ $n = 1.23$	$r^2 = 1.000$
Temkin	$b = 194.47$	$A_T = 19.65$	$r^2 = 0.981$

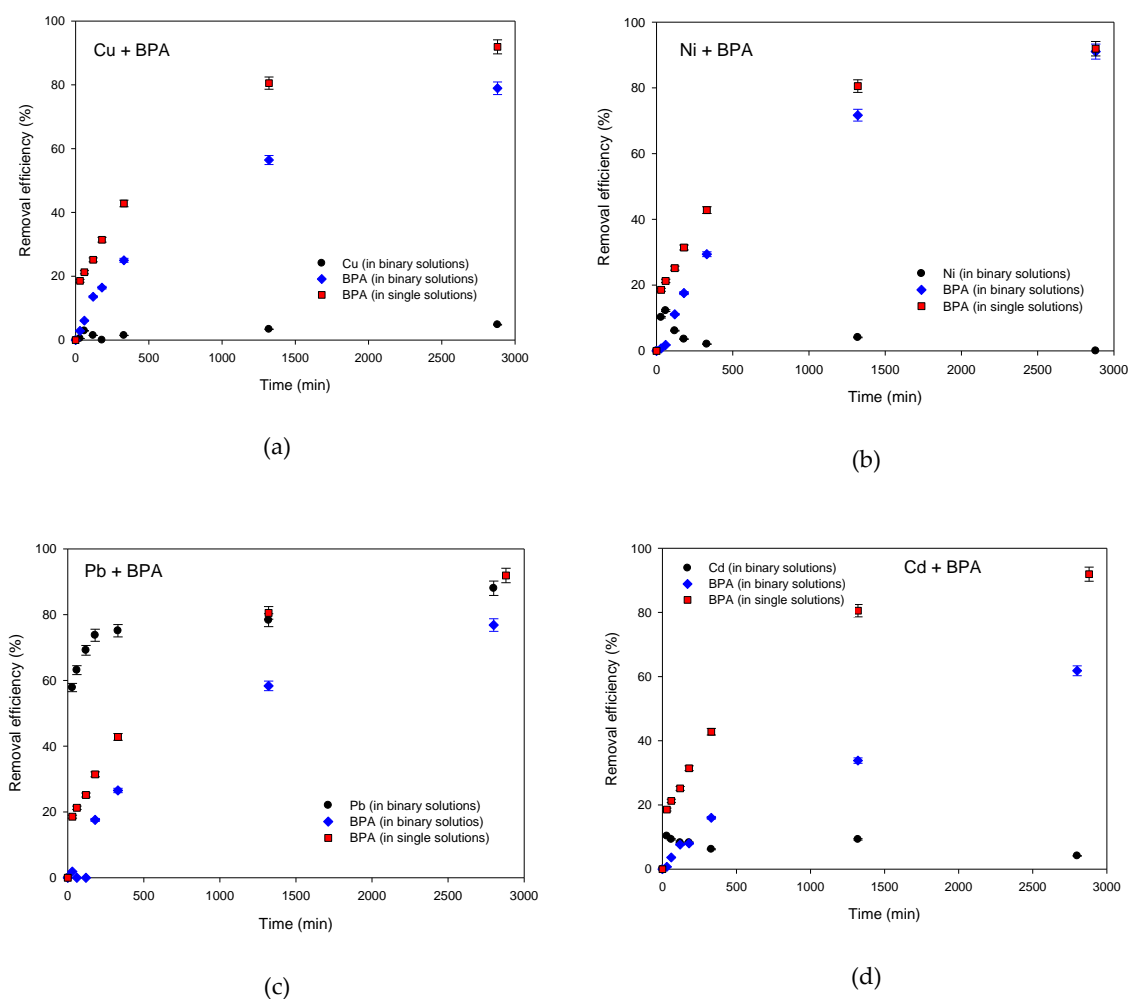


Figure 7. BPA removal (%) in single solutions (red marker) and in binary solutions with (a) copper, (b) nickel, (c) lead and (d) cadmium (black marker for metal and blue one for BPA) using activated carbon AC-40 as the adsorbent.

Table 6. Partition coefficient in function of concentration.

Initial Concentration of BPA, mg/L	Partition Coefficients, L/g
1	52.00
5	51.40
10	49.80
20	40.75
30	26.78
40	20.78
80	2.60
120	1.38
160	0.93

3.6. Adsorption of BPA-Metals Cations Mixtures

Different contaminants such as BPA and a wide variety of metal ions could be present in wastewater. The presence of these ions in solution can lead to competition between metals and the contaminant for the union with the adsorptive sites but also could enhance the BPA adsorption. For example, Han et al. [39] analyzed the influence of copper (II) and lead (II) ions on the adsorption of BPA onto lignin. These researchers found that the presence of the metal cations modified the surface of lignin,

converting it on a less negatively charged surface. It was an advantage for the adsorption of anion species of BPA. Further, Liu et al. [38] studied the simultaneous adsorption of BPA and cadmium (II) ions on activated montmorillonite and found that the maximum adsorption capacity of BPA changed from 74.55 to 80.77 mg/g when cadmium was presented in the solution. Similarly, Bautista-Toledo et al. [37] reported that the presence of chromium (III) enhanced the adsorption of BPA on different activated carbons. The authors attributed the improvement in BPA adsorption to the in situ formation of complex compounds formed by Cr (III) and BPA (Cr (III)-BPA coordination compounds).

In this section, the interaction of some metal cations present in water with BPA was analyzed. The aim was to observe the possible interactions between the metal and the contaminant and/or metal and adsorbent, especially if they had synergistic properties. In all cases, the experiments of mixtures of metals and BPA have been compared with the initial experiment using only BPA. Cadmium, copper, lead and nickel were selected as the representatives of heavy metals because they are very common in wastewater. Figures 7 and 8 show the results.

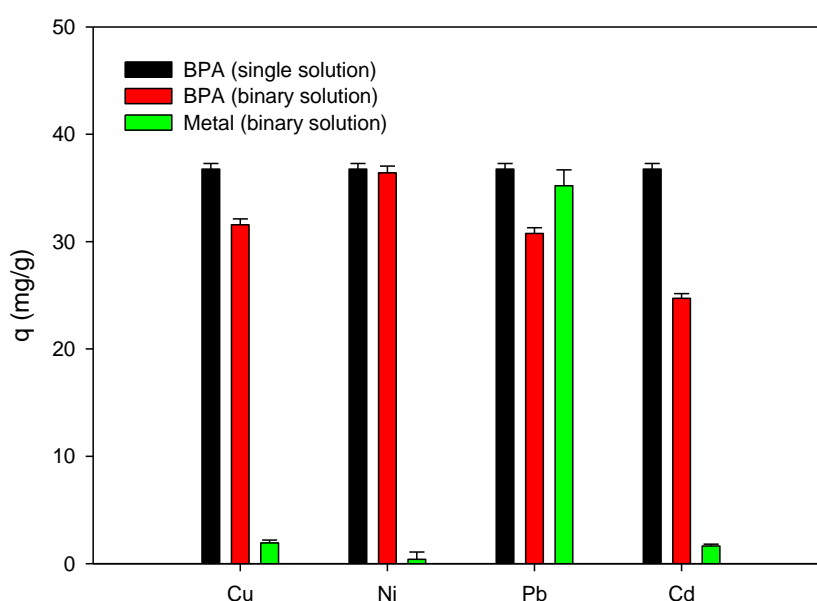


Figure 8. BPA adsorption capacity in single solutions (black color) and in binary solutions with different heavy metals (green marker for metal and red one for BPA) using activated carbon AC-40 as the adsorbent.

Contrary to the referenced studies of Han et al. [39], Lui et al. [38] and Bautista-Toledo et al. [37], the results showed that none of the metal cations improved the efficiency or adsorption capacity of BPA, suggesting a competition with BPA to occupy the porous surface of carbon and consequently decreasing its effectiveness. This is consistent with investigations carried out by Schiewer and Volesky [66], who systematically studied the effect of ionic strength on the adsorption of cations such as Zn^{2+} , Cd^{2+} , Cu^{2+} and Na^{+} . They reported that an increase in ionic strength led to a decrease in the adsorption achieved due to the increase in the electrostatic charge. Niu and Volesky [67] studied the removal of anionic metal complexes ($Au(CN)^{2-}$, CrO_4^{2-} , SeO_4^{2-} and VO_4^{3-}), concluding that an increase in ionic strength decreases the removal.

In the case of the mixture of Cd (II) and BPA, retention of BPA on activated carbon decreased. The difference in adsorption of BPA in the mixture solution (metal-BPA) and in the BPA solution was significant. BPA adsorption capacity changed from 36.8 (single solution) to 24.7 mg/g (binary solution). However, Cd was not greatly adsorbed (adsorption capacity of 1.6 mg/g). This also occurred in mixtures of Cu (II) and BPA, although to a lesser extent. If the predominant binding force of BPA is electrostatic attraction, a decrease in the positively charged sites on the carbon surface could negatively influence the BPA adsorption. A change in the charge of the carbon surface and pH of the solution

during the process due to the adsorption of metals could explain the decrease in the BPA adsorption. Earlier studies reported that the ionic strength of the solution notably influenced the adsorption of different species [37,67,68]. Further, Meng et al. [69] studied the adsorption of phenol and cadmium onto soil and found that the adsorption of mixed pollutants (Cd^{2+} and phenol) showed an antagonistic effect on adsorption. Chemical adsorption was the main mechanism of adsorption of cadmium ions while physical adsorption was the primary mechanism for phenol adsorption (very influenced by the physico-chemical properties of the carbon surface). This indication was consistent with the pH evolution of the system (Table 7) and the point of zero charge reported in Table 1. The net charge of activated carbon decreases with the increase in pH values. Yang et al. [70] noted that the electrostatic surface charge was smaller with the pH of the adsorption system closer to the pH_{pzc} of the activated carbon. In addition, BPA exists in its molecular form under $\text{pH} < 8$ but begins to deprotonate to a negatively charged form at around $\text{pH} 8$ [13,51,70]. Thus, when pH was increased above 7.5, a repulsive electrostatic interaction may be established between the negatively charged surface of the activated carbon and bisphenolate anion. In the mixture of Ni (II) and BPA, the metal was not adsorbed and BPA removal in the mixture solution was very similar to the BPA removal in the single BPA solution. This means that the presence of Ni (II) did not affect the adsorption of BPA. Finally, in the case of the mixture of Pb (II) and BPA, the results suggested that both Pb (II) and BPA were notably adsorbed by activated carbon, probably because they formed a complex in situ with activated carbon, adsorbing and synergistically removing both contaminants present in the solution, or perhaps they distributed the available active sites of the adsorbent for Pb and BPA adsorption.

Table 7. pH evolution on simultaneous adsorption experiments.

Time, min	Cd (II) + BPA	Cu (II) + BPA	Ni (II) + BPA	Pb (II) + BPA
0	5	5	5	5
30	6.9	6.3	6.2	6.4
60	7.5	6.7	6.5	6.8
120	7.8	6.8	6.6	7.1
240	8.1	7.2	7.0	7.2
480	8.0	7.5	7.0	7.3
1440	8.0	7.4	7.3	7.3
2880	7.9	7.6	7.5	7.5

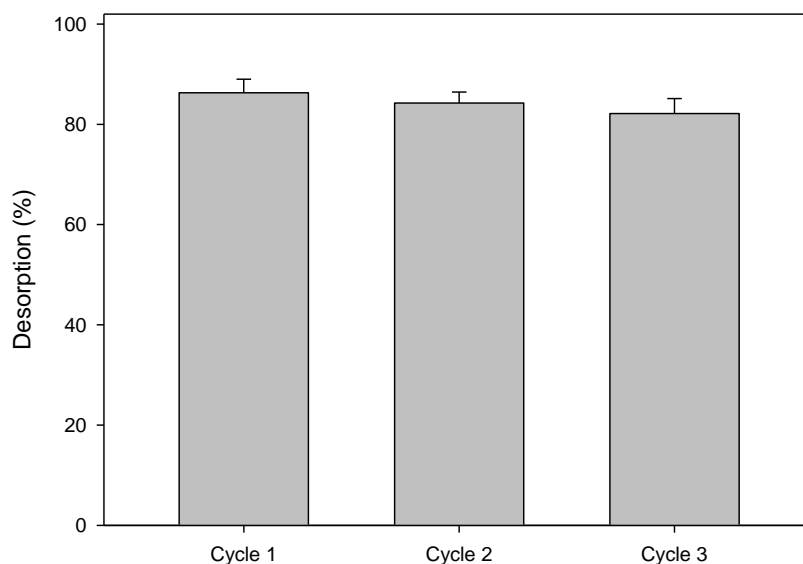
In conclusion, the metal ions may affect the sorption of BPA on AC-40 from different aspects: (1) the metal ions can reduce the BPA adsorption due to the decrease in sorption sites on the AC surface; (2) the metal ions can change the ionic strength of the solution and influence the adsorption of the different species; (3) the metal ions can promote the hydrophobic interaction, which may enhance the BPA adsorption on AC; and (4) the metal ions can promote the formation of complex compounds (metal–BPA coordination compounds) which increase the BPA adsorption.

3.7. Cycles of BPA Adsorption/Desorption

The reuse of AC-40 was investigated in three consecutive cycles of adsorption/desorption, without significant loss in the adsorption capacity. The adsorption capacity in the three consecutive adsorption–desorption cycles is reported in the Table 8. Figure 9 shows the desorption percentage of BPA. The removal efficiency was higher than 90% in all cases, indicating that the activated carbon has good reusability. Further, desorption of BPA from the AC-40 was about 86.3% in the first desorption cycle and only decreased slightly after three adsorption–desorption cycles. These results indicated that the interaction of the BPA molecule to the binding sites on the adsorbent surface can be disrupted without any effect on the properties of the adsorbent. Similar results were found by other authors in their studies of the desorption of BPA by mixtures of methanol-acetic acid [71–73].

Table 8. Removal efficiency and adsorption capacity of BPA adsorption in three consecutive cycles.

Cycle	% Removal	q, mg/g
1	92.5	37.00
2	91.4	36.56
3	90.2	36.08

**Figure 9.** BPA desorption (%) in each adsorption–desorption cycle.

4. Conclusions

The aim of this work was to investigate the application of a commercial activated carbon (AC-40) to remove BPA in single and simultaneous adsorption systems. The results showed that pH did not significantly affect the BPA adsorption at low pH values. However, when the acidity of the medium was increased, a slight improvement in the removal efficiencies was observed. Further, as the dose of activated carbon increased, the BPA removal efficiency increased at all contact times. Additionally, the concentration of BPA did not influence the adsorption, for the values of initial BPA concentration analyzed in this work (similar to those than can be found in some industrial wastewaters).

The isotherm obtained for BPA adsorption onto AC-40 was type I and the maximum adsorption capacity was close to 90 mg/g. The Langmuir and Sips models were the best to reproduce the experimental results.

The experiments with mixtures of metals and BPA showed that none of the metal cations analyzed improved the efficiency of the adsorption process. In the case of the mixtures of Pb (II) and BPA, the results seemed to indicate that both Pb (II) and BPA can be efficiently removed from wastewater using AC-40.

Desorption tests showed good desorption efficiency (higher than 80%) and also BPA adsorption capacities were very similar after three adsorption–desorption cycles, indicating that the activated carbon has good reusability. In conclusion, results of the work showed good adsorptive behavior of AC-40 for BPA removal in batch systems in single- and binary-solute adsorption. However, from a practical point of view and as a potential research line, complete experimental research on packed-bed columns including desorption studies should be carried out.

Author Contributions: Conceptualization, M.C.; formal analysis, M.A.M.-L. and C.E.; investigation, M.A.M.-L. and C.E.; methodology, M.A.M.-L., A.R. and M.C.; software, I.I.-R. and C.E.; supervision, M.A.M.-L.; validation, C.E.; resources, M.C.; writing—original draft, M.A.M.-L., A.R., I.I.-R., and C.E.; writing—review and editing, M.A.M.-L. and M.C. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Acknowledgments: Authors thank M.I. Bautista-Toledo for the activated carbon donation.

Conflicts of Interest: The authors declare no conflict of interest.

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