



The combined treatment of bisphenol A (BPA) by coagulation/flocculation (C/F) process and UV irradiation in aqueous solutions

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ABSTRACT

The coagulation characteristics of bisphenol A (BPA) with polyaluminum chloride (PACl) as a coagulant and the influence of UV irradiation as a complementary process were investigated. The influences of various coagulation parameters such as coagulant dose, pH, solution turbidity, and initial BPA concentrations were analyzed. The possible dominate mechanisms, formation, and performance of flocs over coagulation process were discussed. A coagulant dose of 17.5 mg/L was chosen as optimum dosage. Compared with other ranges of the pH, it can be seen that pH 8 was more effective. It was indicated that turbidity 16 NTU and BPA initial concentration 0.25 mg/L were as optimum conditions in the coagulation process. In this study, elimination mechanism of BPA included colloids entrapment and sweep flocs, predominantly. The results showed that the contact time of UV irradiation was an important factor in the removal efficiency of the BPA residual supernatant. The combination of C/F followed by UV irradiation showed an efficient removal method for treatment of water solutions containing BPA.

Keywords: Bisphenol A (BPA); Coagulation; Flocculation; UV irradiation; Water solution

1. Introduction

Bisphenol A (BPA) is a chemical monomer with high-volume production in industries [1]. It is used in the manufacturing of epoxy resins, phenol resins, polycarbonate plastics, flame retardants, polyesters, polyacrylates, and polysulfones [2–5]. The intermediate products made of BPA are used in making the final commercial products such as powder paints, automotive lenses, protective window glazing, baby bottles,

building materials, electronic equipment, automobiles, and medical devices [5–7]. Along with advantages and widespread application of BPA, this compound is very harmful for human health, wildlife, and ecosystem [8,9]. Since, this chemical substance placed in endocrine disrupting compounds (EDCs) group [10]. In the worldwide, EDCs are as one of the emerging groups of trace contaminants in water resources and wastewater effluents [2]. These compounds disturb the activity of the endocrine system by mimicking, blocking, and disrupting the function of natural hormones even at low concentrations. As a result, EDCs lead to decrease

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in male sperm count and fertility, and increase in testicular, prostate, ovarian, and breast cancers [8,11]. BPA, first time, was introduced as an estrogenic compound in 1938. It is also discussed to cause liver enzyme abnormalities, neurochemical and behavioral effects, cardiovascular diseases, hormonal imbalance, and type 2 diabetes [12,13].

The bioassay studies on EDCs such as BPA in birds, fish, amphibians, and insects present the critical adverse responses by reproductive deficiencies, deformities, egg and offspring development, and serum protein production, such as vitellogenin. These investigations imply that some species, particularly, are sensitive to the exposure of EDCs [14,15]. Several studies reported that BPA could be released and migrated into the environment and natural water resources from the paper mill, agricultural and industrial effluents, natural degradation of polycarbonate plastics, landfill leachates, storm water, wastewaters of industries producing BPA, etc. [1,4,11,16]. Recently, BPA has been found in diverse environments, natural and engineered, across the world such as surface and groundwater supplies, wastewater effluents, sea water, drinking water, bio solids, and sediments [1,17–20]. According to the undesirable consequences of BPA, trying to find a solution in order to eliminate it from aquatic environments seems rational. Various methods of chemical and physical treatment processes have been suggested to remove BPA from water resources involving ozonation, adsorption on activated carbon and other adsorbents [9,21], nanofiltration, reverse osmosis [22], photocatalytic decomposition, electrochemical oxidation, sonochemical [23], and biological processes [24]. However, these methods have limitations and disadvantages include long required reaction times (several days or months), high driving force, difficult operation, restrict application, and low removal efficiencies [3,9,25].

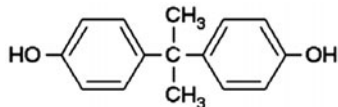
Among the water treatment processes, coagulation/flocculation (C/F) process is a simple technique along with low installation costs [26]. It is reported that BPA could be absorbed onto suspended solids and colloidal particles and therefore sediment in water [8]. While C/F process has been commonly applied to reduce the pollution load of water in treatment plants, some studies have shown that C/F individually is not very effective means for removing EDCs such as BPA [2,4,11]. In this regard, the incomplete elimination of BPA creates a challenge, especially at the trace concentrations [10]. Since the traditional treatment technologies have not been separately successful in treating these compounds, so, in many cases, combined water treatment processes have been considered [3,11]. UV irradiation is a developing technology in treatment sites because it is a cost-benefit way and without the formation of disinfection byproducts [27]. Therefore, efficiency assessment of UV in combination with other conventional treatment procedures with removal of EDCs especially BPA is important [28]. Consequently, the aim of this study was to determine the C/F performance in removing BPA by polyaluminum chloride (PACl) as a coagulant along with using photo-degradation by UV irradiation as a complementary treatment technique. In the C/F process, the effects of operating parameters including pH, the coagulant dose, aqueous solution turbidity, and BPA concentrations were surveyed. The removal efficiency (RE) of high-pressure UV lamp also was evaluated in different contact times, as well.

2. Materials and methods

2.1. Materials

The physicochemical properties of the BPA (99% pure) that was supplied from Sigma-Aldrich (St. Louis, MO, USA) are presented in Table 1. Methanol

Table 1
The characteristic of BPA

Common name (Abbreviation)	Bisphenol A (BPA)
Molecular weight (g/mol)	228.1
log K_{OW}	3.3
pK_a	9.6–10.2
Chemical structure	
Water solubility	120–300 mg/L
Vapor pressure (25°C)	5.3×10^{-9}
Chemical formula	2,2-(4,4-Dihydroxydiphenyl) propane
Made by	Combining acetone and phenol

(GC grade), H_2SO_4 , NaOH, and other chemical solutions and reagents had analytical grade and were obtained from Merck Co., (Germany). PACl was commercially provided from Behin Ab Co. (Iran). The clay (montmorillonite) and solid-phase extraction (SPE) cartridges (GmbH, 3 mL/500 mg) were purchased from Laviosa Co., (Italy) and Macherey-Nagel Co., (Germany), respectively.

2.2. Experiments

2.2.1. Coagulation/flocculation tests

A stock solution was prepared by adding 10 mg BPA into a mixture of Milli-Q water (900 mL) and methanol (100 mL) and was kept in a refrigerator. The other diluted solutions were prepared from this stock solution. A jar test apparatus (Phipps & Bird Stirrer Model 7790-402) was applied in bench-scale experiments. In this study, investigation of the affective variables in C/F process was carried out as follows:

Coagulant doses of 5–18 mg/L, pH in the range of 4–9, turbidities from 3 to 30 NTU and BPA concentrations in the range of 0.25–1.5 mg/L. The mixing speed and retention time for coagulation and flocculation processes were 200 rpm at 5 min and 40 rpm at 20 min, respectively. Sedimentation was conducted for 30 min with no turbulence after C/F processes. Turbidity of the samples was adjusted by montmorillonite and was measured using a Eutech instrument TN-100 turbidimeter. In addition, H_2SO_4 and NaOH were applied as pH values adjustor.

2.2.2. Photo-degradation experiments

Photo-degradation experiments were performed using a high-pressure mercury UV lamp (Arda 150 W.ca, with peak wavelength at $\lambda = 254$ nm, irradiation intensity = 7.1 W/m^{-2} , covered with quartz). The UV exposures were done in a 1-L beaker (bench scale) that UV lamp was vertically fixed in the center of this beaker and sample supernatant of optimal C/F conditions was introduced into this container for the removal of BPA residual. A magnetic stirrer was used for complete samples mixing. To avoidance of light energy lost, the beaker was carefully covered with aluminum foil. In photo-degradation tests, it was noted that the important studied factor was contact times in the range of (0–50 min).

2.3. Analytical methods

RE (%) of BPA by C/F processes and UV irradiation was calculated by following equation:

$$RE \% = \frac{C_i - C_a}{C_i} \times 100 \quad (1)$$

where C_i and C_a (mg/L) are initial and the final BPA concentrations, respectively.

The BPA residual in the studied samples was extracted by SPE method. The SPE cartridges were initially conditioned with 5 mL methanol and 10 mL Milli-Q water. Then, the samples were passed through these conditioned cartridges with a flow rate of 20 mL/min. To release BPA into experimental tubes, 2 mL methanol was passed through the cartridge and then was dried by N_2 gas.

The BPA concentrations in the samples were determined by a gas chromatograph equipped with a flame ionization detector (GC-FID) (Agilent technology GC, 7890A, USA). The injection sample volume was 2 μ L; the carrier gas was helium (purity 99.995%) with a flow rate of 1 mL/min. H_2 was used as fuel gas (flow rate = 30 mL/min); the used column was Cp-sil 5 Cb (25 m \times 320 μ m \times 1.2 μ m); and the detector temperature was held at 250°C. The temperature of the oven was set at 100°C for 1 min and then increased up to 280°C at a rate of 30°C/min for 6 min. Total run time was 12.4 min.

3. Results and discussion

3.1. Determination of optimum coagulant dosage

In order to determination of optimal coagulant dose, different doses ranging from 5 to 18 mg/L were used. In this section, the initial pH value and the concentration of BPA were 7 and 1 mg/L, respectively (Fig. 1(a)). As can be seen, a PACl dosage of 17.5 mg/L had maximum efficiency for BPA removal (38%). Since destabilization of particles occurs usually under conditions of zeta potential values close to zero (–4 to +3 mV) [29,30]. Therefore, 17.5 mg/L is probably appropriate dose provider zeta potential near zero for BPA coagulation that lead to the large flocs formation and significant settleability. At lower doses of coagulant, and at pH 7, charge neutralization was probably the dominate removal mechanism that increases the agglomeration of the smaller and lighter flocs [31–33]. On the other hand, the coagulant dose increasing would entrap the most of the colloidal particles in the aluminum hydroxide precipitates (sweep coagulation) [33,34]. Xiaoying et al. [4] reported that the achieved optimum dose of PACl- Al_{13} and zeta potential for the removal of BPA were 13 mg/L and 0.36 mV, respectively [4]. Therefore, in this study, an

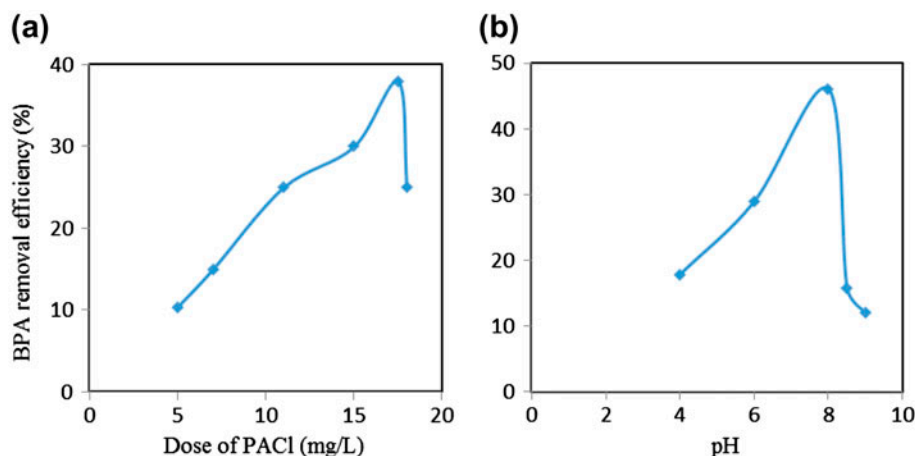


Fig. 1. BPA RE: (a) the influence of PACl various doses (BPA concentration = 1 mg/L and pH 7) and (b) the effect of pH values (BPA concentration = 1 mg/L and PACl dose = 17.5 mg/L).

optimum dose of 17.5 mg/L of PACl was selected and used for the next experiments.

3.2. Effect of pH on BPA removal

The pH of the samples is an important factor in the C/F process [33]. Therefore, in this section, the pH impact on the BPA removal from the aqueous solutions was evaluated (Fig. 1(b)). The pH ranges of the samples were adjusted from 4 to 9 by using 1 N sodium hydroxide and sulfuric acid. Fig. 1(b) shows the BPA RE at pH 8 (optimum pH) was only 46% and it was increased with growing the pH up to 8 and then decreased, although pH continues to enhancement. The behavior and interaction of colloids in the solutions are strongly influenced by their electrical charge [33]. At low pH values, small flocs were created and the low formation speed of destabilization of colloids was achieved. This was probably due to excess absorption of H^+ and highly charged particles, which may result in disruption of electrical interaction [34]. However, at pH 8.0, BPA molecules have not charged and probably there is no charge interaction existing either [35]. Besides, the pK_a values of BPA are in the range of 9.6–10.5. Therefore, it is implying that the ionization of BPA occurring at around pH 8–10 to form the phenolate and bisphenolate anions [36].

In the study conducted by Badruddoza et al. [36], it was shown that the adsorption of BPA remained roughly unchanged at pH below about 7.0. While the BPA adsorption increased in pH above about 7.0 and gradually decreased [36]. Anyway, the removal mechanism might be included initially the BPA adsorption onto destabilized colloids and subsequently entraps into a voluminous agglomeration of

precipitate [4]. Chen et al. [37] and Xiaoying et al. [4] indicated efficient coagulation of BPA occurred at alkaline pH [4,37].

3.3. Turbidity influence

Each colloidal particle carries an electrical charge, which naturally is negative charge [33]. In this study, montmorillonite was added to BPA solutions as turbidity producing agent, to simulate the conditions of natural waters (ranged from 3 to 30 NTU) and to evaluate the turbidity influence on the BPA RE. Fig. 2(a) illustrates the changes of BPA RE along with turbidity variations. The raising of turbidity led to more removal of the BPA (>55%) for turbidity 16 NTU, but then, the additional growth of turbidity possessed reverse effects. These results can be attributed to two probable mechanisms that discussed here:

- (1) The presence of more electrostatic attraction (charge neutralization) between cationic hydrolysis products of PACl and negative charges of montmorillonite surface due to the turbidity enhancement. On the other hand, it can be also derived that relatively high turbidity evinced generation of large flocs that cause overwhelming of these huge flocs and loses of the effect of entrapment and adsorption mechanisms. Xiaoying et al. also obtained similar results, in appropriate turbidity of 16.5 NTU [4].
- (2) Hydrophobic compounds such as the BPA have $\log K_{ow} > 2.5$. They can attach to the particles and thus can be effectively removed by the coagulation/sedimentation process

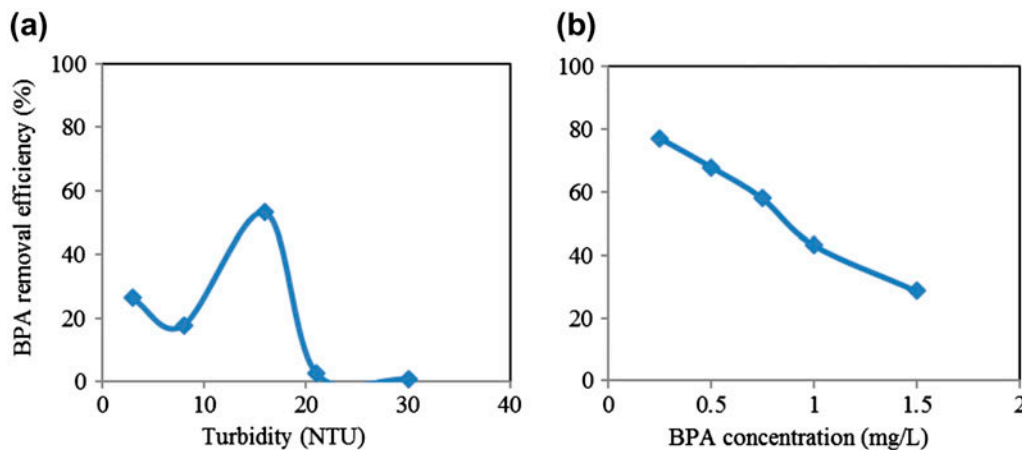


Fig. 2. (a) The effect of turbidity (NTU) on BPA RE (BPA concentration = 1 mg/L, pH 8 and PACl dose = 17.5 mg/L) and (b) determination of optimum BPA concentration (turbidity = 16 NTU, pH 8 and PACl dose 17.5 mg/L).

[38,39]. In this study, the RE of a hydrophobic compound, BPA ($\log K_{ow} = 3.3$), was also higher than 55% in the coagulation phase and with the solution turbidity of 16 NTU. Nam et al. [39] assumed a classification for hydrophobic compounds. Using this classification, it was found that the removal efficiencies of the hydrophobic micropollutants such as BPA generally increased as the turbidity of the water increased [39].

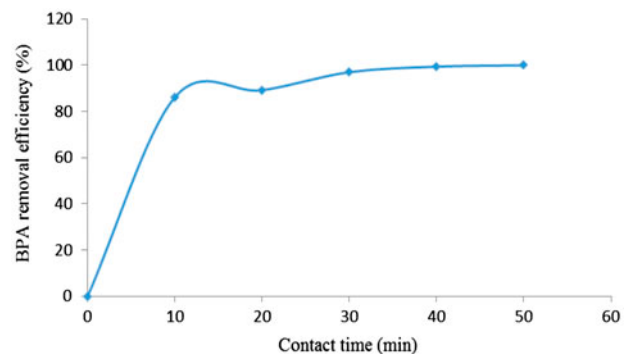


Fig. 3. RE (%) of BPA by UV irradiation over the time.

3.4. Determination of optimum concentration of BPA

The influence of initial BPA concentration on its RE was investigated under optimum specified conditions. Fig. 2(b) includes information about RE at different initial BPA concentrations (0.25–1.5 mg/L). It can be observed that with increment in the initial BPA concentration, its RE was decreased from about 80% in the initial concentration of 0.25 mg/L to approximately 30% in the BPA initial concentration of 1.5 mg/L. This may stem from a constant dose of coagulant along with increasing gradually of pollutant that leads to no well entrapment and adsorption of BPA in flocs structure.

3.5. Photo-degradation of BPA

In these series of the experiments, the degradation efficiency of residual BPA of C/F process was studied by UV irradiation. Fig. 3 shows degradation efficiency of the residual BPA in C/F process supernatant by photo-oxidation (UV light) respect to contact times in

the ranges of 0–50 min. As can be shown in this figure, the RE of BPA was increased sharply in the range of 0–10 min UV-irradiation time. However, a little increase was occurring in removal trend of BPA in the range of 10–50 min contact time. In the other word, the UV-irradiation time in the second contact time series (10–50 min) had the same performance for BPA removal. A probable mechanism for BPA decline with UV application is as follows:

The generation of hydroxyl free radicals and their assault on the aromatic BPA structure [10]. Thus, because of the increasing of the hydroxyl radicals over the time, the rate of BPA destruction had subsequently improved (Fig. 3). Wang et al. [23] reported that during the determination of the optimum pH in the photo-catalytic process by the low-pressure UV lamp, the BPA removal rate was increased with time [23]. In this study, the UV-irradiation time of 10 min was selected as the optimum contact time for residual PBA removal. This time also is appropriate regarding to the economic aspects.

4. Conclusions

Bench-scale experiments were carried out by determining of appropriate conditions for BPA removal with C/F processes followed by photo-degradation. During C/F step of this combined treatment, it was found that the optimum dose of PACl was 17.5 mg/L and the entrapment of colloids in the aluminum hydroxide precipitates was the dominant removal mechanism. Optimum pH, turbidity, and BPA concentration for efficient removal of BPA was 8, 16 NTU, and 0.25 mg/L, respectively. It was also obtained that adsorption, charge neutralization, entrapment, and hydrophobicity were responsible mechanisms for BPA elimination, jointly. The supernatant of C/F process was surveyed to assess of photo-oxidation method in BPA destruction. The photo-degradation study showed that contact time was an effective factor in BPA elimination. The combination of C/F followed by UV irradiation showed an efficient removal method for treatment of water solutions containing BPA.

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