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Removal of naphthalene in Brij 30-containing solution by ozonation using rotating packed bed

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Abstract

The removal efficiency of polycyclic aromatic hydrocarbons (PAHs) by ozonation using a rotating packed bed (RPB) in the surfactant-containing solution is studied. As an ozonation process starts, the ozone-containing gas is introduced and transferred into to the solution. The target PAHs in the solution would be then eliminated via both the gas stripping and ozonation decomposition. The RPB, which provides high gravitational force by adjusting the rotational speed, is employed as a novel ozone contactor. The naphthalene (NAP) and Brij 30 are chosen as the model pollutant and surfactant. Note that the experiments with different concentrations of NAP, Brij 30, and inlet gas ozone are performed for the further investigation. The residual NAP, effluent dissolved oxygen and ozone, and off-gas ozone concentrations are simultaneously monitored. As a result, the removal of NAP in the RPB is remarkably efficient compared with the convectional contactors because of its greater mass transfer coefficient. Moreover, the removal efficiency of NAP is found significantly dependent on the concentrations of NAP, Brij 30, and inlet gas ozone. It takes about several minutes to reach the steady state under the conditions of this study. In addition, it suggests employing RPBs as ozone contacting devices for the high removal efficiency of NAP. Consequently, the present study is useful for the practicable understanding of application of RPBs for the ozonation of PAHs in surfactant-containing solution.
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Keywords: Rotating packed bed; Ozonation; Ozone; Surfactant; Naphthalene; Brij 30

1. Introduction

PAHs (polycyclic aromatic hydrocarbons) have been found as the pollutants in the surface water, soil, and groundwater. The sources of PAHs are mainly from the leaking underground storage tanks, pipelines, and around railroad depots. The common PAHs in the contaminated sites include the naphthalene (NAP), phenanthrene, benzo(*a*)pyrene, fluorene, and biphenyl, etc. The properties of PAHs are semivolatile, toxicological, carcinogenic, and low solubility in water representing a long-term source of contamination. The convectional remediation technique such as the pump-and-treat method, which is proceeded via the dissolution mechanism, is not efficient for the clean up of the contaminated sites within the reasonable time period due to the low solubility of PAHs [1,2].

The combination of the surfactant-enhanced flushing and pump-and-treat treatment is considered as the practicable process to remediate PAHs-contaminated soil [3,4]. The solubility of PAHs would apparently increase with the additive of surfactant while the surfactant concentration is above its critical micelle concentration (CMC) [5,6]. At the surfactant concentration above the CMC, the surfactant molecules start to aggregate and form the micelles. The surfactants are capable of enhancing the solubility of immiscible organic solutes by the partition of organic solutes into the surfactant micelles. Thereby the time required to eliminate the PAHs of the contaminated sites can be reduced.

Ozone is one of the most effective oxidants and has been widely applied in water and wastewater treatment. Trapido et al. [7,8] showed the effective removal of PAHs by ozonation in the aqueous solution. The ozone-containing gas is usually produced by the electrical discharge into pure oxygen or oxygen-enriched gas through an ozone generator. One advantage of ozonation is the contribution of dissolved oxygen, which may be utilized

Nomenclature

C_{AGi0}	concentration of inlet gas ozone (mg/L)
C_{AGe}	concentration of off-gas ozone (mg/L)
$C_{ALb,eff}$	concentration of dissolved ozone in effluent liquid (mg/L)
$C_{BLb,eff}$	concentration of NAP in effluent liquid (mg/L)
C_{BLb0}	initial concentration of NAP in liquid (mg/L)
$C_{OLb,eff}$	concentration of dissolved oxygen in effluent liquid (mg/L)
C_S	concentration of Brij 30 (mg/L)
CMC	critical micelle concentration
CSTR	completely stirred tank reactor
g	gravitational acceleration (m/s^2)
K_{ow}	octanol–water partition coefficient
NAP	naphthalene
ORP	oxidation–reduction potential
PAHs	polycyclic aromatic hydrocarbons
RPB	rotating packed bed
t	time (min)
η_B	removal efficiency of NAP (%)

in the biological process after the decomposition of residual ozone [9,10]. The efficiency of ozonation with pollutant would be reflected by the residual concentrations of ozone and pollutant [11,12]. In addition, the rate-limiting step in many ozonation processes is attributed to the gas–liquid mass transfer. It means that the ozonation performance can be enhanced by increasing the gas–liquid mass transfer rate of ozone. Therefore, the innovation of an ozone contacting device with better mass transfer efficiency is desirable.

The rotating packed beds (RPBs) were used as gas–liquid contactors for the applications of adsorption, distillation, and stripping, etc. [13–15]. RPBs are designed to generate high acceleration of liquid owing to the centrifugal force. The target solution countercurrently encountered with gas flows through the packed material in the environment of high gravity. This novel technology is also named "Higee". According to the previous studies [13–19], RPBs have high gas–liquid mass transfer coefficients, which are important for increasing the gas–liquid mass transfer rate. Recently, RPBs have been introduced as ozonation contactors by Lin and Liu [20] and Chen et al. [21–23]. Note that the mass transfer rate of ozone per unit volume in the RPB gas–liquid contactor is remarkably higher because of its greater volumetric mass transfer coefficient and gas–liquid concentration driving force as compared to the convectional gas–liquid contactors like the completely stirred tank reactor (CSTR). It is feasible to reduce the volume of an ozone gas–liquid contactor by using a RPB, while the proper design of the dimensions and operating conditions of a RPB system is required. However, the study about the ozonation processes for the removal of PAHs in RPBs is still not available.

Referring to the previous studies about the ozonation in a RPB [20–23], the contribution of this study is to investigate the ozonation processes with the PAH in a RPB. The NAP as one of

the main PAHs is chosen as the model pollutant while Brij 30 is used as the surfactant. Based on the study, the dynamic variations of the ozone, NAP, and oxygen concentrations are monitored. Furthermore, the effects of Brij 30, NAP, inlet gas ozone concentrations on the removal efficiency of NAP are examined. As a result, the applicability of ozonation for the removal of NAP in the RPB is demonstrated by the good performance from the experimental data. Consequently, the present study can provide useful information about the ozonation with PAHs in a RPB.

2. Materials and methods

2.1. Chemicals

The NAP as the target PAH was prepared with different concentrations in the Brij 30-containing solution. The NAP obtained from Merck with chemical formula as $C_{10}H_8$ has the molecular weight of 128.2, Henry's constant of 0.0197 (dimensionless ratio of gas to liquid concentration at equilibrium), water solubility of 32 mg/L, and octanol–water partition coefficient (K_{ow}) of 2291 at 25 °C [24]. Brij 30 as a non-ionic surfactant is purchased from Acros with the molecular weight and CMC of 363 g/mol and 9.7 mg/L, respectively [25]. The Brij 30-containing solution was prepared by deionized water. The volatilization of Brij 30 during the experiments can be neglected according to the blank test.

2.2. Instrumentation

The RPB consists of a packed-bed rotator and a stationary case, where the schematic diagram of the RPB may be found elsewhere [19]. The packed bed stuck to the rotor shaft in the case has the dimensions of inner (0.023 m) and outer (0.059 m) radii with the axial height of 0.02 m. The 304 stainless steel wire with the weight of 0.0686 kg wrapped in shape of annular rings is stacked in the packed bed. The density and diameter of the wire are 8478 kg/m^3 and 2.2×10^{-4} m, respectively. Accordingly, the specific area of packing and voidage per unit volume are 793 m^2/m^3 and 0.956 m^3/m^3, respectively. The rotator is connected to a rotor shaft on two bearings, which are in turn mounted on a steel structure. The shaft is connected to a motor, which is controlled by a speed regulator. The rotational speed is controlled as 900 rpm, which provides gravitational force of 35.4 g based on the arithmetic mean radius of the RPB. Liquid enters the RPB through six holes in the liquid distributor. These six holes are arranged in two vertical groups of three per group and the groups are spaced 180° apart. The liquid is sprayed on the inside edge of RPB and thrown outwards by the centrifugal force. The gas is introduced from the outside and flows counter currently with respect to the liquid in the RPB.

The meter of dissolved oxygen (model Oxi 340, Wissenschaftlich-Technische Werkstätten GmbH & Co. KG (WTW), Weinheim, Germany) with the sensor of model CellOx 325 (WTW, Weinheim, Germany) is used to measure the effluent dissolved oxygen concentration ($C_{OLb,eff}$) continuously. The variation of $C_{OLb,eff}$ can represent the gas–liquid mass transfer rate because the dissolved oxygen is considered inert with
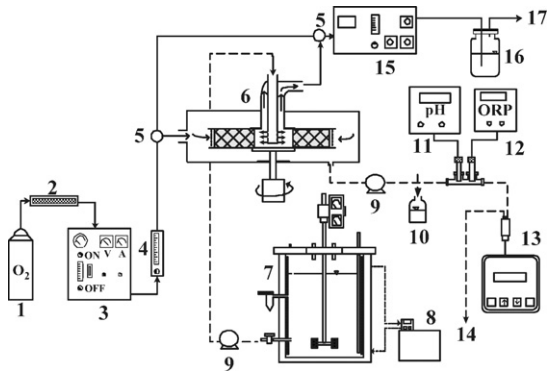
Fig. 1. Experimental apparatus sketch. (—, – –, - - -) ozone gas stream, experimental solution, isothermal water. Components: (1) oxygen cylinder; (2) drying tube; (3) ozone generator; (4) flow meter; (5); three-way valves; (6) rotating packed bed gas–liquid contactor; (7) liquid-storage tank; (8) thermostat; (9) pumps; (10) sampling port; (11) pH meter; (12) ORP meter; (13) dissolved ozone detector; (14) water exit; (15) gaseous ozone detector; (16) KI solution; (17) vent to hood.

the slight effect of chemical reactions. The effluent dissolved ozone concentration ($C_{ALb,eff}$) is analyzed by the liquid ozone monitor (model 3600, Orbisphere Lab., Neuchâtel, Switzerland) with a sensor of membrane-containing cathode. The pH (model 300T, Suntex, Taipei, Taiwan) and oxidation–reduction potential (ORP) meters (model 900C, Apogee, Taipei, Taiwan) with sensors are used to measure the values of pH and ORP of solution, respectively. The effluent residual NAP concentration ($C_{BLb,eff}$) is analyzed using high performance liquid chromatography (HPLC) system with 250×4.6 mm model 516 C18 (5 μm) column (SUPELCO, Bellefonte, PA, USA), and UV/visible detector (model 500, Viscotek, Houston, TX, USA) at 254 nm. The HPLC effluent with flow rate of 1.5 mL/min has the composition with water:CH$_3$CN of 50:50. The injection volume of the analytic solution is 20 μL. The experimental apparatus employed in this work is shown in Fig. 1.

2.3. Experimental procedures

Before starting the ozonation experiments, the ozone-containing gas is bypassed to a photometric analyzer (model SOZ-6004, Seki, Tokyo, Japan) to assure the stability and determine the inlet gas ozone concentration (C_{AGi0}). The liquid-storage tank is equipped with a thermostat to maintain a constant temperature of solution at 25 °C in all experiments. The employed Brij 30 concentration (C_S) of 100–1000 mg/L represents for 10.3 to 103 CMC of Brij 30 with the corresponding enhanced-solubility of NAP of 42–143 mg/L in the solution. The initial value of pH of the experimental solutions is about 5.3–5.4. The liquid flow rate entering the RPB is controlled as 0.3 L/min in the ozonation processes. The gas stream at the preset flow rate of 1.94 L/min is directed into the RPB at initial time ($t = 0$). The ozonation experiments of NAP are carried out for about 10 min to ensure reaching the steady state in this study. The effluent liquid is sampled at desired intervals to analyze the variations of $C_{BLb,eff}$, $C_{ALb,eff}$, $C_{OLb,eff}$, and ORP in the course of experiments. In addition, the concentration of the off-gas ozone (C_{AGe}) is measured simultaneously.

3. Results and discussion

3.1. Ozonation of NAP in RPB

The variations of $C_{BLb,eff}$, $C_{ALb,eff}$, C_{AGe}, $C_{OLb,eff}$, ORP, and pH during the ozonation of NAP with time in the condition of C_S, initial NAP concentration (C_{BLb0}), and C_{AGi0} of 100, 10, and 30 mg/L, respectively, are shown in Fig. 2. There are two characteristic times of concern in the course of the ozonation of NAP using a RPB. One is the time for reaching the constant $C_{BLb,eff}$; the other one is that for arriving the steady state of $C_{ALb,eff}$. After the $C_{BLb,eff}$ decreases to the constant value, the $C_{ALb,eff}$ apparently starts to increase with ozonation time. It is because that the reaction rate between the NAP and ozone is considerably fast [26,27] so that most ozone transferred from the gas phase into the solution would be consumed immediately in the initial stage. As the $C_{ALb,eff}$ becoming a constant implies that the steady state of the whole ozonation system in the RPB has been achieved.

The initial attacks of ozone molecules on NAP are proceeded via the dipolar cycloaddition or electrophilic substitution of ozone on the unsaturated bonds of benzene rings [26]. The decomposition of NAP would accompany with the generation of the hydrophilic by-products including oxalic, formic, phthalic, and phthalaldehydic acids, etc. These by-products are considered less hazardous and harmful to the environment. Meanwhile, the surfactant-containing solution is regenerated, and then can be reused in the remediation process to enhance the dissolution of PAHs. One may address that the small surface tension of the Brij 30-containing solution (of about 26 dyn/cm, while that of pure water is 72 dyn/cm at 25 °C) as the index of the surfactant activity shows the negligible change after the ozonation process. The phenomenon can be elucidated by the previous study of Chu
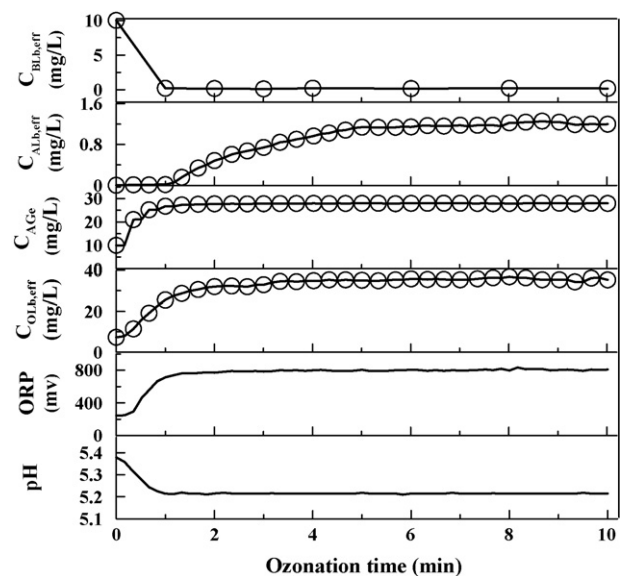

Fig. 2. Profiles of naphthalene, dissolved ozone, effluent gas ozone, dissolved oxygen ($C_{BLb,eff}$; $C_{ALb,eff}$; C_{AGe}; $C_{OLb,eff}$) concentrations, ORP, and pH when ozonation of naphthalene in RPB. $C_S = 100$ mg/L, $C_{BLb0} = 10$ mg/L, $C_{AGi0} = 30$ mg/L. Symbols and line: experiments.

et al. [28], which observed the reactivity of ozone with the Brij35 is quite low.

On the other hand, the C_{AGe} and $C_{OLb,eff}$ rapidly increase in the early period and approach to constant within the shorter time compared with $C_{ALb,eff}$. The utilization efficiency of inlet gas ozone can be estimated as 6.79% from the steady state value of C_{AGe}. The low utilization efficiency of ozone is resulted from the relatively high applied ozone dosage and small contactor volume (0.185 L) of the present RPB system. Furthermore, the $C_{OLb,eff}$ of 35.3 mg/L at steady state is employed to estimate the gas–liquid mass transfer coefficient of oxygen as $0.114\,s^{-1}$ based on the saturation concentration of dissolved oxygen in water at 25 °C (40.08 mg/L) [19]. In comparison with the gas–liquid mass transfer coefficient of 0.00588–$0.0171\,s^{-1}$ in the bubble column reactors [29] and 0.02–$0.04\,s^{-1}$ in the stirred reactor [30], the value of gas–liquid mass transfer coefficient in the RPB is remarkably greater.

Note that the time required for the ozonation system to reach the steady state in a RPB system is comparatively short because of the plug flow hydrodynamics and greater mass transfer coefficient [20,31]. Therefore, it is advantageous for the practical application of ozonation with PAHs using a RPB in situ. Moreover, the ORP with the initial value of 246 mV remarkably increases to the steady state value of about 756 mV in the initial stage. The ORP mainly depends on the elimination of $C_{BLb,eff}$ that may be applied as a supplementary index for indicating the stability of the removal efficiency of NAP in a RPB system. The pH value of the solution decreasing from 5.38–5.21 reveals the formation of the related organic acids.

3.2. Effects of system variables on removal of NAP

The ozonation performance in the respect of the removal of NAP in the Brij 30-containing solution using the RPB is evaluated with various conditions of system variables. The removal efficiency of NAP (η_B) defined by Eq. (1) at steady state in the various conditions are shown in Table 1. The system variables include 6 levels of C_S, 4 levels of C_{BLb0}, and 5 levels of C_{AGi0}. As indicated in Table 1, the η_B value ranging from 4.66–100% shows the significant difference. Certainly, the effects of system variables on the η_B need to be considered and investigated.

$$\eta_B = \frac{(C_{BLb0} - C_{BLb,eff})}{C_{BLb0}} \tag{1}$$

It shows that the η_B definitely decreases with the increase of C_S as illustrated in Fig. 3. Especially in the cases of sole gas stripping ($C_{AGi0} = 0$), the addition of Brij 30 into the solution dramatically reduces the value of η_B. It is caused by the smaller Henry's constant in the condition of higher C_S to lead the lower driving force for the volatilization of NAP. For the ozonation processes, the lower removal performance is also resulted from the slower gas–liquid mass transfer rate of ozone because of the increasing gas–liquid interfacial resistance with the presence of surfactant [32,33]. Further, the individual contributions of gas stripping and ozonation to the η_B value are estimated based on Table 1. It is found that the removal of NAP via the gas stripping is less than 20% in the ozonation cases when $C_S \geq 100$ mg/L.

Table 1
Summary of η_B under different concentrations of Brij 30, naphthalene, and inlet gas ozone in RPB Contactor

Run no.	C_S (mg/L)	C_{BLb0} (mg/L)	C_{AGi0} (mg/L)	η_B (%)
1	0	10	0	76.8
2	0	10	30	100
3	100	10	0	12.9
4	100	10	6	57.3
5	100	10	12	80.4
6	100	10	30	98.2
7	100	10	40	98.3
8	250	25	30	83.7
9	300	50	0	8.93
10	300	50	30	69.5
11	500	10	30	86.7
12	500	50	30	62.4
13	1000	10	0	8.49
14	1000	10	30	82.3
15	1000	50	0	7.35
16	1000	50	30	44.2
17	1000	100	30	36.3

Therefore, the NAP is mainly eliminated through the ozonation mechanism in the Brij 30-containing solution while the volatilized amount of NAP in the exhaust gas would be reduced as well.

The effect of C_{BLb0} on the η_B for the gas stripping and ozonation of NAP in the RPB is depicted in Fig. 4. One may note that the η_B value of gas stripping carried out in a CSTR is independent on the C_{BLb0} value [34]. On the contrary, the flow conditions of both gas and liquid phases in a RPB are close to the plug flow [19,20]. As shown in Fig. 4 and Table 1, the η_B value of the gas stripping process in a RPB decreases with the increase of C_{BLb0}. For the ozonation cases, the η_B value apparently decreasing with the higher C_{BLb0} suggests that the greater pollutant loading in a RPB treated by the same ozone dosage would have the lower removal efficiency.

Fig. 5 is employed to illustrate the interactive effects of C_S and C_{BLb0} on the η_B for the ozonation processes of NAP in the
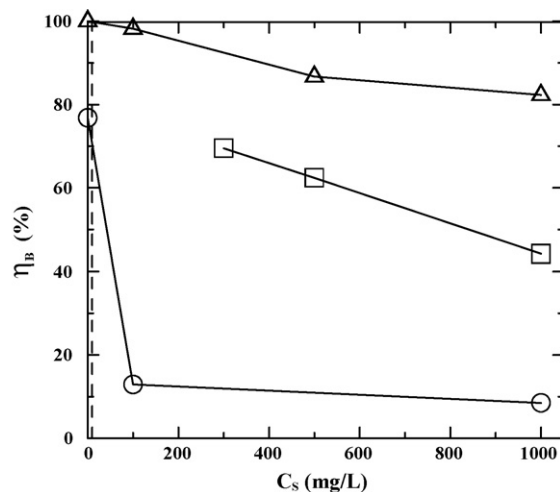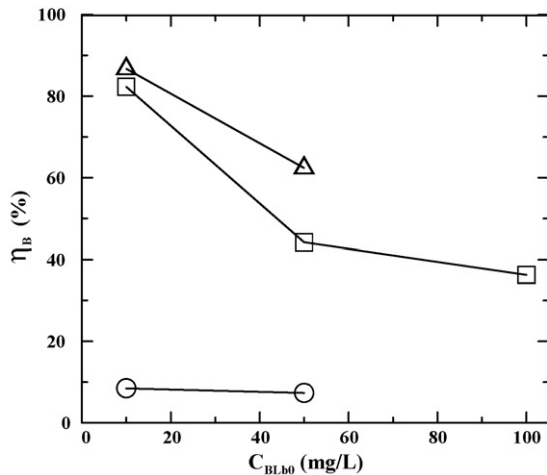

Fig. 3. Variations of η_B with C_S for gas stripping and ozonation of NAP in RPB. Symbols: experiments (○) $C_{BLb0} = 10$ mg/L and $C_{AGi0} = 0$ mg/L. (△) and (□) $C_{BLb0} = 10$ and 50 mg/L with $C_{AGi0} = 30$ mg/L. (- - -) CMC of Brij 30.

Fig. 4. Variations of η_B with C_{BLb0} for gas stripping and ozonation of NAP in RPB. Symbols: experiments ($\bigcirc$) $C_S = 1000$ mg/L and $C_{AGi0} = 0$ mg/L. ($\triangle$) and ($\square$) $C_S = 500$ and 1000 mg/L with $C_{AGi0} = 30$ mg/L.

Fig. 6. Variations of η_B with C_{AGi0} for ozonation of NAP in RPB. Symbols: experiments ($\bigcirc$) $C_S = 0$ mg/L and $C_{BLb0} = 10$ mg/L. ($\triangle$) $C_S = 100$ mg/L and $C_{BLb0} = 10$ mg/L. ($\square$) $C_S = 300$ mg/L and $C_{BLb0} = 50$ mg/L. ($\bullet$) $C_S = 1000$ mg/L and $C_{BLb0} = 10$ mg/L. ($\blacktriangle$) $C_S = 1000$ mg/L and $C_{BLb0} = 50$ mg/L.

RPB. The experimental conditions are with the ratio of C_{BLb0} to C_S as a constant of 0.1 and the C_{AGi0} of 30 mg/L. The results show that the η_B value is significantly decreased with the concurrent increases of C_S and C_{BLb0}. It should be addressed that the trend of the η_B in Fig. 5 shows the more remarkable decrease than those with the sole increase of C_S or C_{BLb0} as depicted in Figs. 3 and 4, respectively. That indicates the accumulative effect of C_S and C_{BLb0} on the decrease of the η_B. It can be explained that the higher C_S would decrease the η_B value due to the lower gas–liquid mass transfer rates of both ozone and NAP while the Brij 30 is considered of low reactivity with ozone. Furthermore, the case of the higher C_{BLb0} needs the greater transferred ozone dosage to reach the same η_B value. When both the C_S and C_{BLb0} simultaneously increase, the related phenomena mentioned above would take place to result in the lower η_B.

In addition, the effect of the applied ozone dosage expressed by C_{AGi0} on the η_B is shown in Fig. 6. Obviously, the difference of the η_B between the processes of sole gas stripping and ozonation is distinct. The greater η_B value can be attained by

introducing the ozone-containing gas with higher C_{AGi0} in the RPB system. One may address that the transferred ozone dosage depends on both C_{AGi0} and gas–liquid mass transfer coefficient of ozone. As for a usual ozonation reactor, the C_{AGi0} or reactor volume has to be enhanced for the same removal efficiency owing to its smaller gas–liquid mass transfer coefficient. As a result, all the findings obtained in this study are useful for the proper operation of the ozonation processes with PAHs in the surfactant-containing solution using a RPB.

4. Conclusions

Ozonation combined with the rotating packed bed is employed as an efficient way for the removal of naphthalene (NAP) in the Brij 30-containing solution. The elimination of NAP in the ozonation processes using a RPB includes two mechanisms of gas stripping and ozonolysis simultaneously. The presence of Brij 30 as the surfactant decreases the gas–liquid mass transfer of both NAP and ozone, resulting in the lower removal efficiency of NAP. The following conclusions may be drawn:

1. After the introduction of ozone gas was started, the effluent gas ozone and dissolved oxygen concentrations increase with ozonation time rapidly, while the effluent residual NAP concentration decreases concurrently. The effluent dissolved ozone concentration would remarkably start to increase after the residual NAP concentration reaches the constant.
2. Note that the gas–liquid mass transfer coefficient in the RPB, which may decrease with the presence of the surfactant, is still remarkably greater compared with those of the convectional contactors. The RPB system is allowed to reach the steady state quickly within several minutes. The oxidation–reduction potential (ORP) varying with the residual NAP concentration can be used as a supplementary index in the course of the ozonation process.
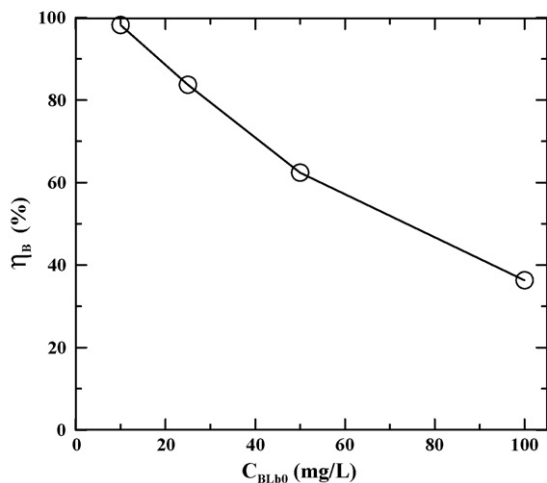

Fig. 5. Variations of η_B with C_{BLb0} for ozonation of NAP in RPB. Symbols: experiments $C_{AGi0} = 30$ mg/L. $C_{BLb0}/C_S = 0.1$ mg/L/(mg/L).
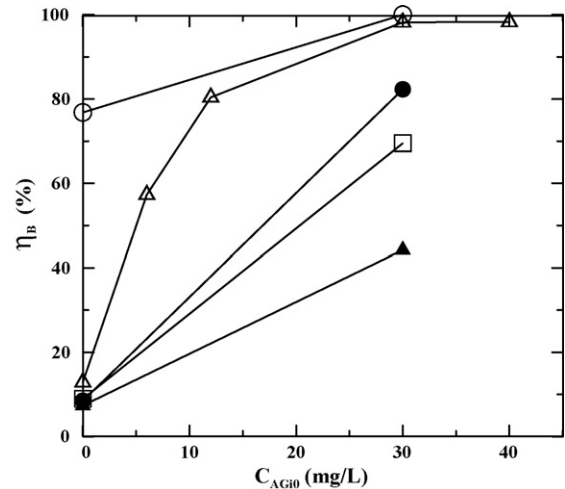
3. The ozonation system in a RPB with lower Brij 30 and NAP concentrations as well as higher inlet gas ozone concentration is favorable for the removal efficiency of NAP. The presence of Brij 30 would significantly reduce the volatilization of NAP from the solution. When the Brij 30 and NAP concentrations increase together, the accumulative effect appears to lead the more remarkable decrease for the removal efficiency of NAP.

Acknowledgement

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