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Enhancement of *o*-cresol removal using PAC and acclimated biomass immobilized in polyvinyl alcohol hydrogel beads

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ABSTRACT

The feasibility of using polyvinyl alcohol (PVA) hydrogel beads encapsulating powdered activated carbon (PAC) and biomass for the enhancement of *o*-cresol removal via simultaneous adsorption and biodegradation was investigated in this study. The optimum concentrations of PVA and PAC used to synthesize spherical PVA hydrogel beads with 3–4 mm diameter were found to be 7.5 and 0.5% (w/v), respectively. The results of kinetic studies showed that the rate constant of *o*-cresol removal for the initial *o*-cresol concentration range of 100–400 mg/L could be sustained when the immobilized biomass supplemented with PAC in hydrogel beads was used. However, the removal of *o*-cresol was inhibited for the initial *o*-cresol concentration above 150 mg/L when only immobilized biomass in the beads was used. Thus, the supplementation of PAC to the immobilized biomass in the beads could enhance the removal of *o*-cresol via simultaneous adsorption and biodegradation.

Keywords: Hydrogel beads; Powdered activated carbon; Immobilized biomass; Simultaneous adsorption and biodegradation; *o*-Cresol

1. Introduction

The presence of *o*-cresol in the effluents of numerous industries such as oil refineries, coal conversion plants, petrochemicals, polymeric resins, pharmaceuticals, biocides, disinfectants, insecticides, herbicides, dyes, pulp and paper, and other synthetic chemical manufacturing industries is deleterious to the environment and human health upon discharging to receiving waters [1–4]. For these reasons, a proper treatment of *o*-cresol containing wastewaters is of great importance before the final discharge. In general, various physical and chemical treatment methods such as membrane contactor, Fenton oxidation, and photochemical oxidation are applied to remove phenolic compounds, for example, cresols, from wastewaters [5–10]. However, the drawbacks of some of these methods, for instance, high treatment cost and the formation of undesirable by-products due to incomplete mineralization, have restricted their extensive application especially in treating large quantity of

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wastewaters [11–13]. Hence, biological treatment method is usually preferred as it is more economical and complete mineralization of target compounds could be achieved [14].

The conventional biological treatment method, that is, activated sludge process, normally requires large reactor volume and produces excess sludge. Occasionally, it may encounter difficulty in solid–liquid separation in the settling tank leading to the failure of the process [15]. The application of immobilized biomass via entrapment by polymeric materials, however, has eliminated the drawbacks arising from using the suspended sludge process. In addition, high concentrations of biomass could be maintained in the bioreactor without the risk of sludge washout [16]. Furthermore, the selection of microbes to be entrapped is also possible to suit the treatment of wastewaters with various characteristics [17–19]. As *o*-cresol has been recognized as an inhibitory compound which usually inhibits its own biodegradation [20], the treatment of *o*-cresol containing wastewaters by immobilized biomass could be plausibly inhibited particularly in the presence of high concentrations of *o*-cresol. In this case, improvement of immobilization is necessary in order to mitigate the inhibition effect exerted by *o*-cresol.

Activated carbon (AC) is generally an effective adsorbent which could adsorb numerous bioresistant organic compounds, including *o*-cresol, from the aqueous system [21,22]. However, to date, the potential of immobilizing biomass together with AC for use in wastewater treatment is still not thoroughly being explored. The use of AC, such as powdered activated carbon (PAC), and biomass is anticipated to enhance the removal of *o*-cresol via simultaneous adsorption and biodegradation processes. Since polyvinyl alcohol (PVA) is non-toxic to microorganisms, besides being mechanically and chemically robust and economically feasible [23], it was chosen in this study to encapsulate the PAC and biomass to form spherical hydrogel beads after cross-linking with boric acid. Hence, the objectives of this study are: (i) to determine the optimum concentrations of PVA and PAC for the synthesis of hydrogel beads and (ii) to investigate the feasibility of simultaneous adsorption and biodegradation of *o*-cresol at increasing concentration utilizing the synthesized hydrogel beads.

2. Materials and methods

2.1. Chemicals

The PVA used for the synthesis of hydrogel beads was purchased from Sigma–Aldrich and had an average MW of 89,000–98,000 with hydrolyzed percentage of more than 99%. The PAC employed was a steam-activated wood-based carbon purchased from Century Chemical Works, Malaysia with a commercial name of MALBON S4. It was dried in the oven at 103–105°C for at least 2 h and kept in the desiccators prior to use.

2.2. Culturing of *o*-cresol acclimated biomass

A 5 L beaker with a working volume of 4 L was used as a sequencing batch reactor (SBR) with the operating periods of FILL, 0.5 h; REACT, 8 h; SETTLE, 1 h; DRAW, 0.5 h; and IDLE, 14 h for every cycle. The SBR was initially inoculated with the activated sludge collected from a local municipal sewage treatment plant at Batu Ferringhi, Penang, Malaysia and fed with the feed solution containing (in mg/L): peptone (32), sucrose (109), KH_2PO_4 (32), K_2HPO_4 (180), $(NH_4)_2SO_4$ (212), $NaHCO_3$ (425), $MgSO_4$ (49), $FeCl_3.H_2O$ (18.8), and $CaCl_2$ (40). During each cycle, 3 L of feed solution was introduced into the SBR throughout the FILL period. Subsequently, the REACT period began with the activation of aeration pump to maintain the dissolved oxygen (DO) concentration above 4.0 mg/L in the mixed liquor and the ejector to agitate the mixed liquor. Prior to the termination of the REACT period, an appropriate amount of mixed liquor was wasted to maintain the solids retention time at 20 days in the reactor. The sludge was allowed to settle in the following period to enable the supernatant to be drawn out from the reactor during the DRAW period, with the residual volume of 1 L throughout the IDLE period in preparation for the next cycle. Once the steady state was attained, *o*-cresol was added into the feed in increasing concentration up to 500 mg/L to replace the peptone and sucrose as the sole substrate. The pseudo-steady state was presumably attained once complete removal of *o*-cresol in the SBR was achieved within 5% of the react time. The mixed liquor suspended solids (MLSS) concentration was found to be about 2,100 ± 30 mg/L at the pseudo-steady state. This wasted mixed liquor was used to blend with polymeric matrix forming spherical immobilized biomass hydrogel beads as described in the following section.

2.3. Determination of optimum concentrations of PVA and PAC for the synthesis of hydrogel beads

Three samples of PVA powder of 2.5, 7.5, and 9.5 g in weights were initially prepared and each of which was mixed with 2.0 g of sodium alginate and 50.0 mL of distilled water. These mixtures containing different weights of PVA were stirred and heated until the solids were completely dissolved and the solutions were cooled to 30–40˚C. To each of the polymeric solutions was added 0.5 g of PAC and 50 mL of distilled water and the resulting solution was homogenized by gentle stirring. At this point, the three polymeric matrices contained PVA concentrations of 2.5, 7.5, and 9.5% (w/v), respectively, each with 0.5% (w/v) of PAC. Each of the polymeric matrices was extruded using a peristaltic pump through a thin borosilicate glass dropper into 200 mL of gelating solution consisting of saturated boric acid and 1.0% (w/v) of $CaCl_2$, yielding spherical PVA hydrogel beads of 3–4 mm (average of 3.71 mm) in diameter which was measured using a light microscope. The presence of 2.0% (w/v) of sodium alginate in the final volume of each polymeric matrix was to prevent the agglomeration of beads in the gelating solution. The beads were then kept gently stirred in each of the gelating solutions for 2 h for solidification and subsequently washed with tap water under aerated condition using an aeration pump until the foaming produced by unpolymerized PVA is stopped.

Three 2 L beakers were individually filled with 900 mL of feed solution containing 200 mg/L of *o*-cresol. These solutions were then separately added with 100 mL of each type of beads developed as described above. The circulation of beads in each of the batch reactors was provided by aeration using aeration pump with the sampling of *o*-cresol being conducted at the same time until its concentration reached a constant value, which represented the attainment of the adsorption equilibrium. These experiments were conducted in triplicate for the estimation of standard error. The selection of the optimum concentration of PVA was based on the maximum adsorption capacity of the hydrogel beads for *o*-cresol.

Using the optimum PVA concentration, the hydrogel beads with different PAC concentrations, namely 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.8, and 1.0% (w/v) were synthesized. Again, the optimum PAC concentration was chosen based on the maximum adsorption capacity of the synthesized hydrogel beads for *o*-cresol.

2.4. Immobilization of biomass in the hydrogel beads with PAC for the simultaneous adsorption and biodegradation of o-cresol

The mixed liquor containing biomass wasted from the SBR (Section 2.2) was centrifuged and the supernatant was discarded. The concentrated biomass was then blended with the polymeric matrix with the optimum PVA and PAC concentrations before being extruded into the gelating solution. The maximum biomass concentration that could be immobilized to yield the spherical PVA hydrogel beads with diameter of 3–4 mm was found to be 20 g/L. Next, the hydrogel beads with the maximum biomass concentration were used in the batch reactors containing feed solutions with concentrations of *o*-cresol ranging from 100 to 400 mg/L. The concentrations of *o*-cresol and COD in the batch reactors were determined at frequent intervals once aeration was started until *o*-cresol was completely removed in each batch reactor. These experiments were conducted in triplicate for the estimation of standard error.

For comparison with the removal of *o*-cresol solely via biodegradation, the hydrogel beads were synthesized without the addition of PAC. The degradation of *o*-cresol at the concentration range of 100–400 mg/L in the batch reactors was then studied.

2.5. Analytical methods

The *o*-cresol concentration was analyzed according to the 4-aminoantipyrine method as described in the Standard Methods [24] by using a Vis spectrophotometer (Dynamica model HALO VIS-10) at λ_{max} of 500 nm. The concentration of COD was determined via the closed reflux titrimetric method in which the digested sample with standard potassium dichromate solution was titrated with standard ferrous ammonium sulfate titrant [24]. The MLSS concentration was measured by filtering the mixed liquor and the residue was dried to a constant weight at 103–105˚C [24]. The DO concentration was measured using a DO meter (YSI 550A).

3. Results and discussion

3.1. Optimum concentrations of PVA and PAC for the synthesis of hydrogel beads

The optimum concentrations of PVA and PAC to develop the hydrogel beads were determined based on the maximum adsorption capacity of the beads for *o*-cresol in the batch reactors. The results of the batch experiments showed that the amounts of *o*-cresol adsorbed at the PVA concentrations of 2.5, 7.5 and 9.5% (w/v) were 26.5 ± 0.5, 33.7 ± 0.0, and 25.2 ± 0.6 mg, respectively. As the maximum adsorption capacity for *o*-cresol was obtained with PVA concentration of 7.5% (w/v), this concentration of PVA was regarded as the optimum concentration for the synthesis of hydrogel beads in the subsequent studies. It was observed that the beads containing PVA concentration of 2.5% (w/v) totally disintegrated after 4 h of aeration in the batch reactor. The presence of higher concentration of PVA would generally promote extensive bonding among the polymer strands which leads to the formation of more compact lattice structure of beads and causes diffusion limitation [25]. This was demonstrated by lower adsorption capacity for the beads containing 9.5% (w/v) PVA for *o*-cresol in comparison with beads containing 7.5% (w/v) PVA.

The amounts of *o*-cresol adsorbed at different concentrations of PAC in the hydrogel beads are shown in Fig. 1. It is evident that the maximum adsorption capacity for *o*-cresol was achieved using hydrogel beads with 0.5% (w/v) PAC. The reason for lower adsorption capacity of beads with 1.0% PAC as compared to beads with 0.5% PAC was attributed to the polymeric matrix containing 1.0% (w/v) PAC being too viscous, thus forming beads of larger size (4–5 mm diameter). Conventionally, the bead diameter is inversely correlated to the surface area/volume ratio, the smallest beads corresponding to the highest surface area/volume ratio value and vice versa. Nunes et al. [23] had revealed that the beads with larger diameter would encounter more diffusion restrictions due to lower surface area/volume ratio value, which explained lower amount of *o*-cresol being adsorbed by PAC despite higher percentage was used.

3.2. Removal of o-cresol

When hydrogel beads with immobilized biomass and PAC were used in the removal of *o*-cresol, enhancement of *o*-cresol removal via simultaneous adsorption and biodegradation processes was possible. The time courses of *o*-cresol concentration at the initial concentration range of 100–400 mg/L in the batch reactors using hydrogel beads with immobilized PAC and biomass and using beads with immobilized biomass only are shown in Fig. 2(a) and (b), respectively. For all the cases, these time courses data were found to be well fitted to the zeroth-order formulation ($R^2 > 0.97$). Yao et al. [26] had also described the removal of cresol using the zeroth-order kinetics. The pseudo-zeroth-order rate constants of *o*-cresol removal ($k_{o\text{-cresol}}$) calculated for both batch reactors at various initial *o*-cresol concentrations are shown in Fig. 3. It was observed that the batch reactor using hydrogel

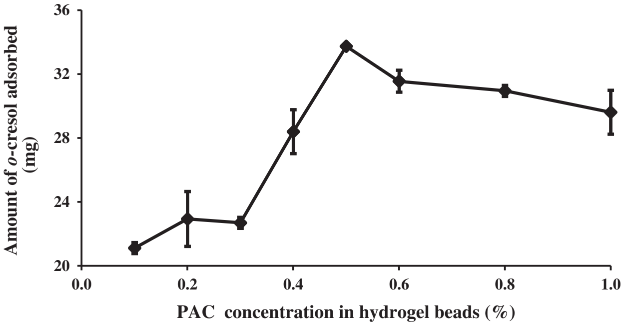

Fig. 1. Amount of *o*-cresol adsorbed by hydrogel beads of varying PAC concentrations. Error bars indicate standard deviations.

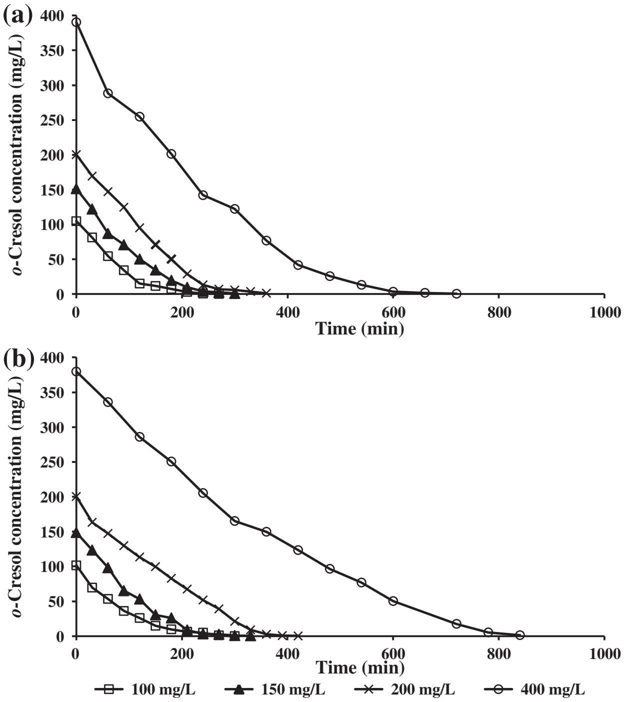

Fig. 2. Time courses of *o*-cresol removal; (a) batch reactors containing hydrogel beads with PAC and biomass, (b) batch reactors containing hydrogel beads with biomass.

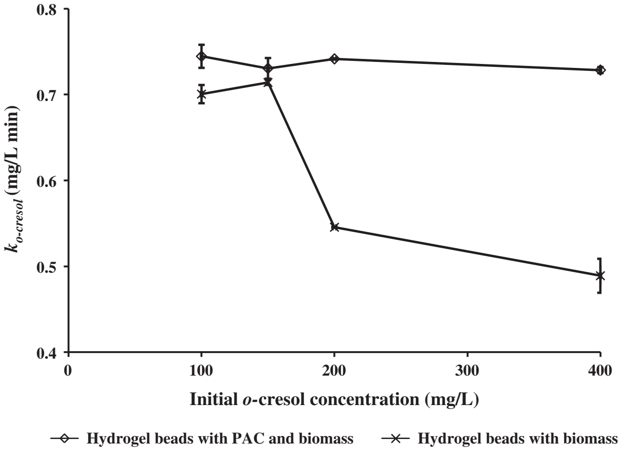

Fig. 3. Values of rate constant for *o*-cresol removal in batch reactors containing hydrogel beads with PAC and biomass and biomass only at various initial concentrations of *o*-cresol. Error bars indicate standard deviations.

beads with PAC and biomass could sustain the rate constant of *o*-cresol removal at above 0.7 mg/L min for the initial concentration of *o*-cresol up to 400 mg/L. In contrast, the rate constant of *o*-cresol removal in the batch reactor using hydrogel beads with biomass only decreased drastically from 0.71 to 0.55 mg/L min when the initial concentration of *o*-cresol was increased from 150 to 200 mg/L. The value of rate constant decreased further to 0.49 mg/L min when the initial concentration of *o*-cresol was 400 mg/L. These results highlight the superiority of hydrogel beads encapsulating both PAC and biomass as the *o*-cresol removal rate could be sustained at higher concentrations of *o*-cresol as compared with the beads immobilized with only biomass. This was due to the initial adsorption of *o*-cresol by PAC which could partially offset the inhibition effect exerted by *o*-cresol on biomass particularly at higher concentrations, thus enhancing the removal of *o*-cresol in the batch reactor using hydrogel beads with PAC and biomass.

The time courses of COD removal via simultaneous adsorption and biodegradation and via biodegradation only are presented in Fig. 4(a) and (b), respectively. It was observed that the trends of COD removal in both batch reactors followed closely the trends of *o*-cresol removal as shown in Fig. 2. This was also observed in the study conducted by Lim et al. [27] which showed that the decrease of COD concentration was in parallel with the removal of 4-chlorophenol from the mixed liquor. In addition, for all the *o*-cresol concentrations studied, near complete removal of COD was also achieved in the same duration required for the complete removal of *o*-cresol. This indicated that complete degradation of *o*-cresol could have been achieved during the biodegradation of *o*-cresol in both batch reactors. In general, the biological removal of toxic organic compounds such as phenolic compounds would lead to the accumulation of intermediate compounds which are capable of exerting COD before being biodegraded further when the concentrations of the initial toxic organic compounds were exhausted [28–30]. Therefore, the use of hydrogel beads to bioremediate *o*-cresol containing wastewaters can enhance the overall treatment process since complete biodegradation of *o*-cresol is achievable.

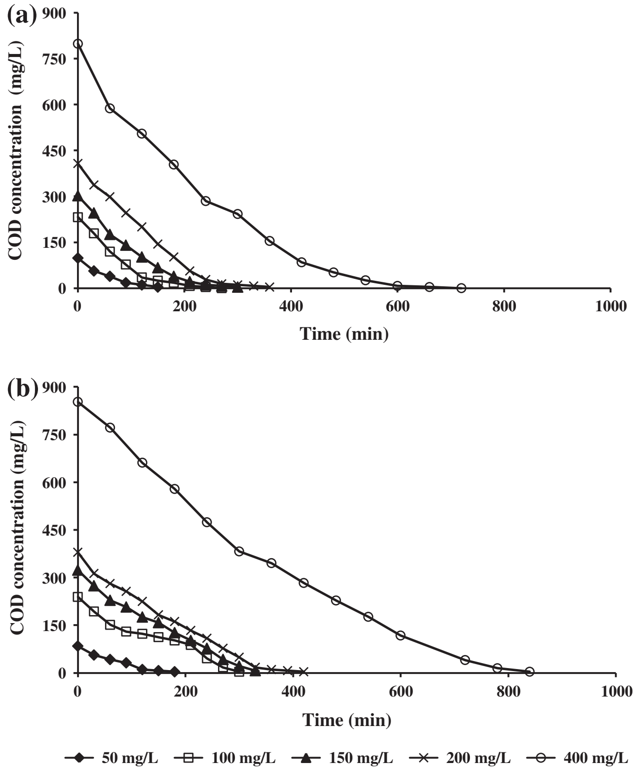

Fig. 4. Time courses of COD removal; (a) batch reactors containing hydrogel beads with PAC and biomass, (b) batch reactors containing hydrogel beads with biomass.

The reusability of PVA-biomass beads was studied earlier and it was reported that at least three cycles of use were possible if the initial *o*-cresol concentration was 100 mg/L [31]. However, the number of cycles of use was reduced to two and to only one when the initial *o*-cresol concentration was increased to 400 and 800 mg/L, respectively. The disintegration of beads was caused by the excessive growth of biomass within the beads due to the surplus supply of *o*-cresol [31]. It is believed that the reusability of PVA–PAC-biomass hydrogel beads would be affected by the initial *o*-cresol concentration in the same manner.

4. Conclusions

The optimum concentrations of PVA and PAC for the synthesis of spherical PVA hydrogel beads with 3–4 mm diameter based on the maximum adsorption capacity for *o*-cresol were found to be 7.5 and 0.5% (w/v), respectively. From the kinetic studies, when the hydrogel beads with PAC and biomass was used, a fairly constant rate of *o*-cresol removal could be sustained for the initial *o*-cresol concentration varying from 100 to 400 mg/L. In comparison, the removal of *o*-cresol exhibited inhibition for the initial *o*-cresol above 150 mg/L when hydrogel beads with only biomass were used. These results indicate the importance of PAC supplementation to immobilized biomass in hydrogel beads for the enhancement of *o*-cresol removal via simultaneous adsorption and biodegradation processes.

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