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Enhanced lead(II) binding properties of heat-treated cattle-manure-compost-activated carbons

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ABSTRACT

Considerable concerns have been raised over the presence of lead(II) in water bodies. In this work, the performance of heat-treated cattle-manure-compost-activated carbons were investigated to remove lead(II) from aqueous solution. Activated carbons were prepared by one-step $ZnCl_2$ activation followed by heat treatment at different temperatures. They were characterized according to BET surface area, pore size distribution, elemental analysis, pH_{PZC} and Boehm's titration. It was found that the increase of treatment temperature resulted in a decrease in both the BET surface area as well as the concentration of phenolic groups. However, such decreases were compensated by an increase of lead(II) by more than three times to a value of 0.110 mmol/g, and this was also true for commercial F400-activated carbon. Lead(II) adsorption onto heat-treated activated carbons could be described by Langmuir isotherm with weaker interaction to active sites. Results also suggested that the increase in lead(II) uptake was due to the rich electron clouds of the $C\pi$-system, regardless of the difference in the surface area upon heat treatment.

Keywords: Activated carbon; Cattle-manure-compost; $C\pi$-system; Heat treatment; Isotherm; Lead(II)

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1. Introduction

Lead-contaminated water through the discharge of industrial effluent is one of the most serious environmental problems nowadays. Mining, electroplating, metal processing, textile, battery manufacturing, paint manufacture, pesticides, pigment manufacture, printing and photography are among the industrial activities that contribute to lead(II) contamination [1,2].

Removal of lead ions has become a subject of special concern due to its recalcitrance and persistence in the environment. Lead ions are very toxic even at low exposure, not biodegradable and easily accrue in human body, thereafter can cause acute and chronic health effects [3]. Lead can cause heart disease, brain disorder, insomnia, anemia, and impair the reproductive system [3]. Due to its health hazards, the allowable concentration of lead in drinking water as prescribed by World Health Organization is limited to 0.01 mg/L [1].

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Removing lead from water through adsorption process can be technically challenging and expensive [4,5]. In addition, the global demand for commercial activated carbon, the mainly used adsorbent in adsorption, is forecasted to increase by nearly 5–10% annually [6]. The current price of commercial activated carbon in Japan for example could reach as high as ¥3,675/kg [7]. This problem can be alleviated by exploiting abundant local carbonaceous resources into activated carbon [8,9].

The availability of potential precursors, however, may vary from one country to another. One of the identified candidates in Japan is cattle-manure-compost (CMC), which is a solid residue of temperature-phased anaerobic digestion for methane generation. For record, Japan produces nearly 9.1×10^9 tons per annum of domestic animal manure [10]. It is known that the livestock waste can cause threat to public health and environment if no proper disposal method is followed. Thus, a possible "use waste to treat waste" approach towards sustainable environment is by converting CMC into activated carbon [11].

Our present work was aimed at evaluating the lead(II) binding properties onto heat-treated CMC-activated carbons. Performance of activated carbons under different textural and surface properties was compared with the commercial activated carbon, namely Filtrasorb400 (F400). The equilibrium data were analyzed using the established Langmuir and Freundlich models, and possible mechanisms governing the adsorption were discussed.

2. Materials and methods

All analytical grade chemicals were purchased from Kanto Chemical Co., Inc. Commercial F400 activated carbon was purchased from Calgon Mitsubishi Chemical Corporation for comparative performance evaluation.

2.1. Activated carbons

CMC was obtained from JFE Corporation, Japan. The elemental composition of the precursor and F400 activated carbon is tabulated in Table 1. Two $ZnCl_2$ weight ratios of 0.5 and 1.5 were used in chemical impregnation prior to activation. One-step activation was carried out at 500°C for 1 h under a flow of 300 mL/min N_2, and followed by heat treatment at temperatures of 500, 800, and 1,000°C for 1 h under the flow of high-purity helium to produce activated carbons with different textural and surface properties. Helium was used in heat treatment to eliminate surface functional groups at higher temperatures, and this process could not be achieved by using nitrogen gas [12]. Ash and minerals were removed by soaking overnight the activated carbons in hydrochloric acid and concentrated hydrofluoric acid. The activated carbons were designated as A*x*T*y*, where A denotes CMC-activated carbon by *x* $ZnCl_2$ weight ratio, while T represents heat treatment at *y* hundred degrees Celsius.

Table 1
Elemental composition of CMC and F400-activated carbon

Element (%)	CMC	F400
C	46.5	83.1
H	5.66	0.39
N	1.33	0.30
O	38.9	6.60
Ash	7.63	9.6
Si	5.27	–
Ca	1.52	0.84
S	0.45	3.29
Fe	0.26	3.88
K	0.13	0.51
Ti	–	0.72
V	–	0.23
Sc	–	0.12

2.2. Textural and surface characteristics

Textural characteristics were obtained at a liquid nitrogen temperature of −196°C using a Beckman Coulter SA3100 surface area analyzer (USA). The samples were out-gassed in vacuum at 300°C for 2 h prior to measurement. Surface area was estimated by the Brunauer–Emmett–Teller (BET) model assuming that the adsorbed nitrogen molecule has across-sectional area of 0.162 nm^2, while the total pore volume was determined at a relative pressure, P_s/P_0 of 0.9814. The average pore diameter can be roughly calculated from the values of BET surface area and total pore volume.

Surface chemistry of activated carbon was evaluated using Boehm's titration [13]. Different batches of 0.3 g of activated carbon were brought into contact with 15 mL solutions of $NaHCO_3$ (0.1 M), Na_2CO_3 (0.05 M), NaOH (0.1 M), and HCl (0.1 M). The mixtures were then agitated at 100 rpm and 25°C for 48 h. After which, the aliquots of each sample were back-titrated with HCl (0.1 M) and NaOH (0.1 M) for acidic and basic groups, respectively. Neutralization points were observed using pH indicators; phenolphthalein solution was used for titration of strong base with strong acid, and methyl red solution for titration of weak base with strong acid.

2.3. Removal of lead(II) ions

A stock solution of model lead(II) wastewater was prepared by dissolving the desired weight of lead(II) chloride in de-ionized water. Serial dilutions were made to obtain a range of different lead(II) concentrations. Adsorption of lead(II) was carried out at 25 ± 2˚C and 100 rpm for 48 h in a water bath orbital shaker. A fixed amount of activated carbon was added to the conical flasks containing 50 mL of lead (II) solution with known concentrations. The initial solution pH was not adjusted, and was measured as 5.3 ± 0.2. The amount of metal ions adsorbed, q_e (mmol/g) was calculated from $q_e = (C_0 - C_e) \times (V/m)$, where C_0 and C_e are, respectively, the initial and equilibrium concentrations in mmol/L, V (L) is the volume of solution and m (g) is the mass of activated carbon. Concentration of lead(II) was measured using atomic absorption spectroscopy (Rigaku novAA 300).

3. Results and discussion

3.1. Textural characteristics

Fig. 1 shows the profile of BET surface area and micropore surface area of activated carbons that have undergone heat treatment at different temperatures. In general, A1.5 possesses greater surface area and mesopore content than A0.5. A higher amount of $ZnCl_2$ used for activation has resulted in a more intense dehydrating effect to enlarge the surface area so as to increase the mesopore content [11,14]. The BET surface area for both CMC-activated carbons was found to decrease with increasing heat treatment temperatures. About 34% of the pore volume of A1.5 was diminished in the heat treatment from 500–800˚C, while the decrease in its micropore content was not significant. It is suggested that the decrease in BET surface area of the A1.5 series was due to the broadening of mesopores. However, the decrease of surface area of highly microporous A0.5 could be attributed to the widening of micropores to mesopores. Unlike the CMC-activated carbons, the increase of heat treatment temperature caused a slight increase in the surface area of F400-activated carbon.

Fig. 2 shows the N_2 adsorption-desorption profile of the A1.5 series activated carbons. The A1.5 series display concave downward isotherms with steep slopes at low P_s/P_0. According to IUPAC [15], these activated carbons can be described under type I isotherm with small type H4 hysteresis: narrow pore size distribution of a microporous material with plate-like pores. A large knee before reaching a maximum volume adsorbed as P_s/P_0 approaching unity signifies that the A1.5 series-activated carbons are rich in mesopore content. However, the degree of mesoporosity was found to decrease from 77 to 66% as the treatment temperature increases from 500 to 1,000˚C.

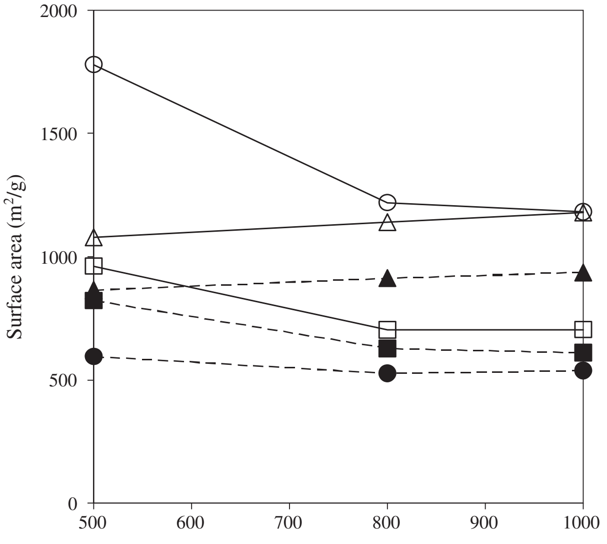

Fig. 1. Effect of treatment temperature on surface area of activated carbons. Symbols of A0.5 (□), A1.5 (○) and F400 (Δ), depict the activated carbons. Open and closed symbols represent the BET surface area and micropore surface area, respectively.

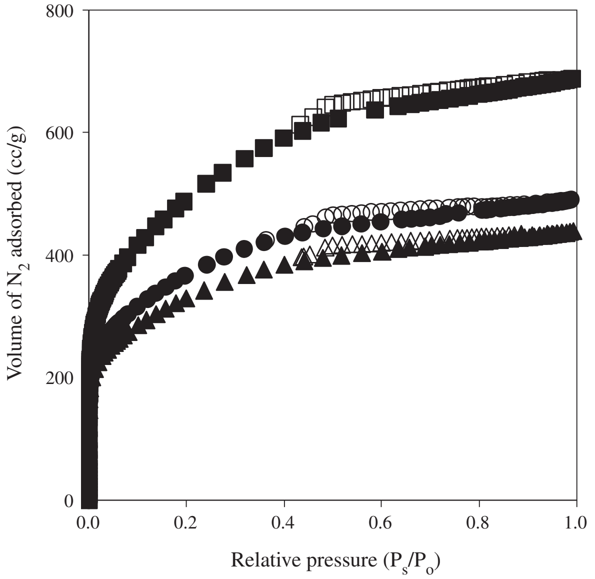

Fig. 2. Effect of treatment temperature on N2 adsorption (closed symbol)-desorption (open symbol) profile of A1.5 series activated carbons. Symbols of A1.5T5 (□), A1.5T8 (○), and A.5T10 (Δ), depict the activated carbons.

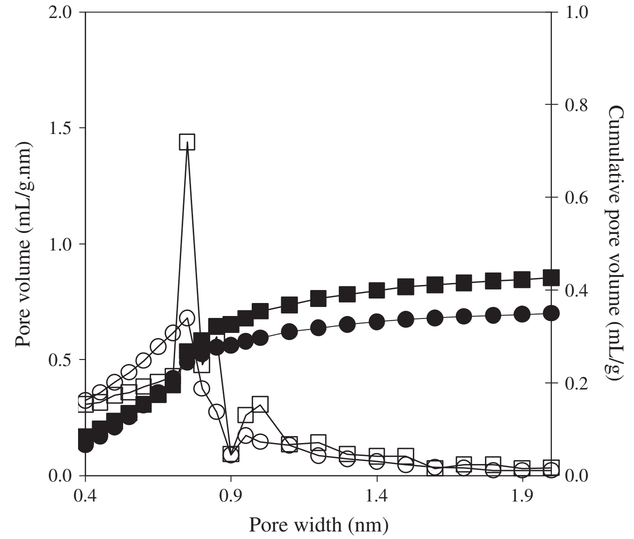

Fig. 3. Effect of treatment temperature on pore size distribution of heat-treated activated carbons. Symbols of A0.5T5 (□) and A0.5T8 (○), depict the activated carbons. Open and closed symbols represent differential pore volume and cumulative pore volume, respectively.

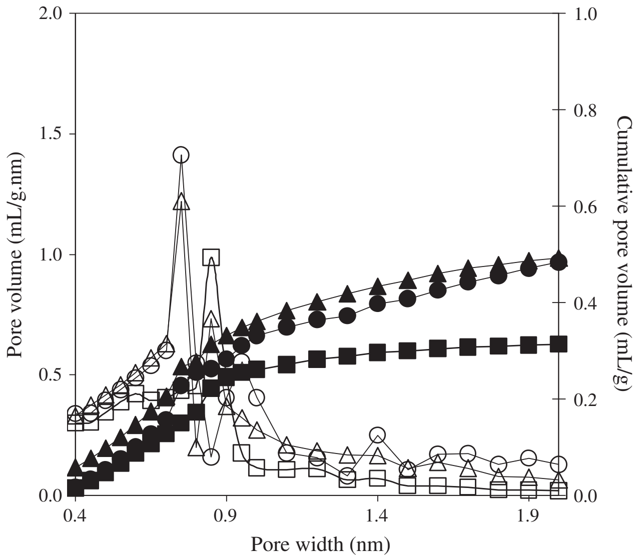

Fig. 4. Pore size distribution of 1,000℃-heat-treated activated carbons. Symbols of A0.5T10 (□), A1.5T10 (○), and F400T10 (Δ), depict the activated carbons. Open and closed symbols represent differential pore volume and cumulative pore volume, respectively.

The influence of treatment temperature on pore size distribution of A0.5 series activated carbons is shown in Fig. 3. Both A0.5T5 and A0.5T8 demonstrate bimodal distribution which centered at 0.75 nm, i.e. the micropore region. The increase of treatment temperature to 800℃ caused the decline in peak intensity and pore volume. Fig. 4 shows the pore size distribution of activated carbons treated at 1,000℃. A0.5T10 displayed a unimodal distribution with pores centered at 0.85 nm, which is 0.10 nm greater than those discussed in Fig. 3. This result is consistent with the support that the decrease of surface area of the A0.5 series was due to the enlargement of micropores. A1.5T10 and F400T10 showed a sharp distribution at 0.75 nm, and an identical amount of pore volume. However, A1.5T10 exhibited a broad multimodal distribution which corresponds to a highly mesoporous carbon. The average pore widths for all activated carbons studied were within the lower edge of mesopore, ranging from 2.1 to 2.5 nm.

3.2. Surface characteristics

Surface functional groups and elemental properties of activated carbons are tabulated in Table 2. Generally, the composition of hydrogen, nitrogen and oxygen on the surface of activated carbons decreased as the temperature increases to 800℃, because of the release of volatiles and surface functional groups. This has resulted in an increase of the carbon content. At 1,000℃, however, the carbon content was slightly decreased with a small rise in the composition of hydrogen and oxygen. The liberation of electron withdrawing functional groups may enrich electron density on the carbon surface, to which moisture could be readily adsorbed at room temperature [12,16]. About 66% of nitrogen content was removed in heat treatment up to 1,000℃. This could consist of nitrogen functional groups such as amines and amides, while the remaining nitrogen on aromatic carbon skeletons is expected to include quaternary nitrogen, pyridinic and pyrrolic [17].

Yield of activated carbons was slightly decreased with increasing temperature. It suggests the effectiveness of helium to impede further burn-off of carbon at higher temperature, and selectively remove the volatiles and surface functionalities [12]. From Table 2, the increase in treatment temperature has resulted in the increase of pH_{PZC} [18] and the equilibrium pH of lead(II) removal. This could be attributed to the basic surface of activated carbons as the treatment temperature increases because of the release of acidic functional groups [12,19]. For all activated carbons studied, the values of pH_{PZC} were always greater than those of equilibrium pH which implies a positively charged surface in adsorption.

Table 2 also demonstrates the influence of treatment temperature on surface functional groups of activated carbons [18]. It is clear that carboxylic and lactonic groups were readily released at 500℃, thus leaving behind only phenolic groups that contribute

Table 2
Surface functional groups and elemental properties of activated carbons

Activated carbons	Yield (%)	pH_{PZC}	pH_e	Functional groups (mmol/g)				Elemental composition (daf, %)			
				Carboxylic	Lactonic	Phenolic	Basic	Carbon	Hydrogen	Nitrogen	Oxygen*
A0.5D5	39.2	7.7	5.5 ± 0.1	0	0.02	0.69	0.47	85.9	1.33	2.03	10.7
A1.5D5	39.7	8.5	5.8 ± 0.2	0	0	0.78	0.47	90.4	1.26	1.82	6.55
F400D5	85.6	9.6	5.8 ± 0.2	0	0	0.42	0.41	94.2	0.05	0.48	5.31
A0.5D8	36.2	10.2	6.0 ± 0.2	0	0	0.49	0.59	92.8	0.09	1.59	5.52
A1.5D8	36.6	10.3	6.0 ± 0.2	0	0	0.52	0.60	94.6	0.13	1.38	3.88
F400D8	83.9	10.9	6.0 ± 0.1	0	0	0.44	0.56	96.0	0.03	0.62	3.37
A0.5D10	35.6	10.4	6.2 ± 0.1	0	0	0.29	0.74	91.2	0.45	0.65	7.75
A1.5D10	36.4	10.4	6.2 ± 0.1	0	0	0.28	0.67	93.5	0.25	0.70	5.55
F400D10	83.9	10.9	6.2 ± 0.1	0	0	0.27	0.58	91.2	0.29	0.34	8.18

Notes: pH_{PZC}: pH of point zero charge, pH_e: equilibrium pH, daf: dry, ash free.
*calculated by difference.

to the surface acidity of activated carbon. The trend shows, with the increase of temperature, that the concentration of phenolic groups decreased to an identical amount, while the concentration of basic groups increased. It was well tallied with the values of pH_{PZC}. The basic groups on activated carbon could be generally attributed to the electron-donating character and the delocalized π-electrons on the graphitic structure that could behave as Lewis bases [19].

3.3. Removal of lead(II) ions

Fig. 5 shows the maximum removal of lead(II) ions onto activated carbons treated at different temperatures. In general, all activated carbons studied show an increasing trend of lead(II) uptake with increasing treatment temperatures. At 500℃, A1.5T5 and F400T5 showed a greater lead(II) removal than A0.5T5, which could be explained by mesopore-rich and high surface area of the former activated carbons [14]. The lead(II) uptake onto activated carbons was found to increase significantly after the heat treatment at 800℃. For example, A0.5T8 showed three times greater uptake of lead(II) when compared to A0.5T5. This was also true for A1.5T8 and F400T8, where the removal was about 2.5 times higher than their 500℃ counterparts. At this point, the surface area and mesopore content are not only the contributors for the increment in lead(II) uptake. Despite the decrease in surface area (as depicted in Fig. 1), it is suggested that the increasing uptake of lead(II) was attributed to Cπ-cation interaction, i.e. the attraction of metal ions to the delocalized π-electrons [20].

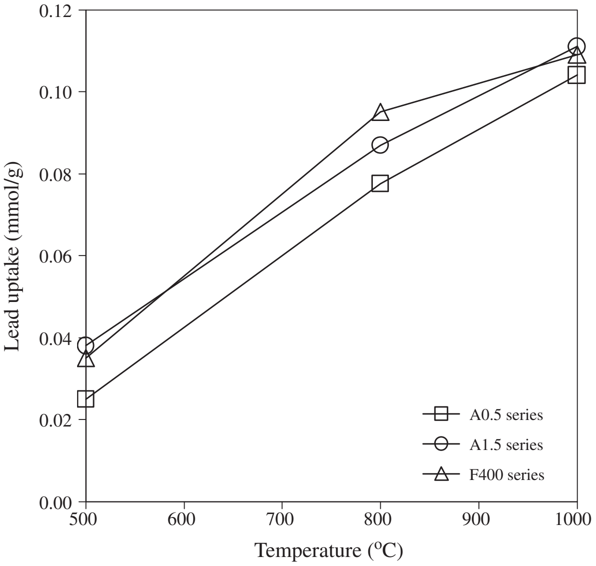

Fig. 5. Effect of heat treatment on the maximum uptake of lead(II) by activated carbons.

Cπ-cation interaction occurs when the phenolic groups, i.e. the electron withdrawers, are released in heat treatment at higher temperatures causing the aromatic skeletons to become rich in electron density which thereafter is viable for lead(II) removal. The effect was more pronounced for activated carbons treated at 1,000˚C, where lead(II) uptake was similar at about 0.110 mmol/g. Because of a small alteration in the surface area (Fig. 1), it could be adequately inferred that the electron density was deficient in an 800˚C-heat treatment as compared to that of 1,000˚C. This was tied-up with the profile of phenolic groups as shown in Table 2. For all activated carbons studied, it was observed that the influence of phenolic groups was likely to offer a trivial effect towards lead(II) removal even though some researchers demonstrated that the acidic groups are strong enough to bind divalent metal cations [21].

From Table 2 and it is obvious that the increase in equilibrium pH during adsorption was a result of protons attraction onto the electron-rich π-system. The amount of protons adsorbed was found to increase, i.e. from 0.0079 to 0.0095 mmol/g, for activated carbons treated at 500 and 1,000˚C, respectively. On the molecular basis, however, the amount of surface protonation was considerably small when compared to the amount of lead(II) adsorbed, due to the difference in attractive force towards different charge of cations.

Fig. 6 shows the calculated distribution of lead species in 0.24 mmol/L solution at varying pH. The values of equilibrium constant at 25˚C were obtained from [22]. From Table 2, the highest equilibrium pH of 6.3 was recorded by activated carbons treated at 1,000˚C. At this solution pH, approximately 96.1% of lead species was Pb^{2+}, while the remaining 3.9% was $Pb(OH)^{+}$. It has been reported that little metal adsorption was obtained in acidic solution due to excessive protonation that repulses away metal ions from the carbon surface [14,23]. On the contrary, increasing equilibrium pH in adsorption increases the propensity of precipitate complexes to evolve, thus nullify the quantified amount of lead(II) adsorbed onto activated carbon. From Fig. 6, stable precipitate namely Pb(OH) only appears in solution at pH 6.6, which is slightly higher than the equilibrium pH recorded from this work. It is believed, however, that the formation of solid precipitate during adsorption (hybrid process), to some extent, could expedite the removal of lead species from contaminated water [24].

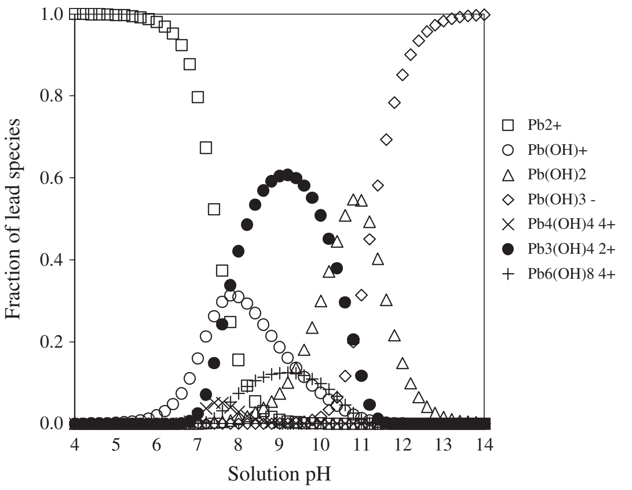

Fig. 6. Fraction of lead species in aqueous solution at different pH.

3.4. Isotherm studies

A consistent equilibrium pH for every activated carbon as shown in Table 2 indicates uniform amount of protons adsorbed regardless the increase of lead(II) concentration in adsorption. Therefore, it is adequate to utilize single component isotherm models to represent the adsorption data. In general, information gained from the isotherm studies is important for designing a good and practical adsorption process. Two generally used isotherm models, namely Langmuir [25,26] and Freundlich [27] were employed in this work to analyze and characterize the adsorption data. The Langmuir isotherm assumes uniform monolayer adsorption on a homogeneous surface, and is given by,

$$q_e = \frac{C_e \times Q \times b}{1 + b \cdot C_e} \tag{1}$$

where Q represents maximum uptake and b is adsorption affinity. The Freundlich isotherm describes multilayer adsorption on a heterogeneous surface, and is given by,

$$q_e = K_F \times C_e^{\frac{1}{n}} \tag{2}$$

where K_F is the Freundlich constant and n is the empirical parameter describing the energetic heterogeneity of the sites. These two models were solved and compared by means of linear and nonlinear approaches for optimum correlation of determination (r^2). For linear least-squares method, the constants of Langmuir and Freundlich were predicted by plotting C_e/q_e vs. C_e and log q_e vs. log C_e, respectively. Isotherm constants under nonlinear approach were determined by *Solver* add-in of Microsoft Excel by maximizing the r^2.

Fig. 7 shows the equilibrium uptake of lead(II) onto 1,000˚C-heat-treated activated carbons. It is apparent that these activated carbons demonstrate a concave downward curve that indicates a preferable adsorption of lead(II). The respective isotherm constants for these activated carbons are tabulated in Table 3. The Langmuir constants varied for different approaches of linear and nonlinear, with the linear approach offered a significantly lower r^2. This arises from the difference of error structures and mathematical complexities associated with the linear approach. In linear least-squares method, the scatter vertical points (equilibrium uptake) across the line assume a Gaussian distribution, to which the error distribution is consistent for all equilibrium concentration (C_e). The error distribution is likely to get altered after transforming into a linearized form, thus deviate the true values of constants. Similar trend was also observed under Freundlich isotherm. It suggests the applicability of nonlinear method over linear method in determining the isotherm constants [28]. It is also obvious from Table 3 that the Langmuir isotherm (nonlinear approach) was more appropriate to represent the adsorption data based on the r^2 values which are close to unity. In addition, the values of maximum uptake, *Q* and adsorption affinity, *b* differ only slightly from one another, and in accordance with the plots shown in Fig. 7.

Essential characteristic of Langmuir isotherm can be expressed in terms of a dimensionless separation factor, R_L as [29],

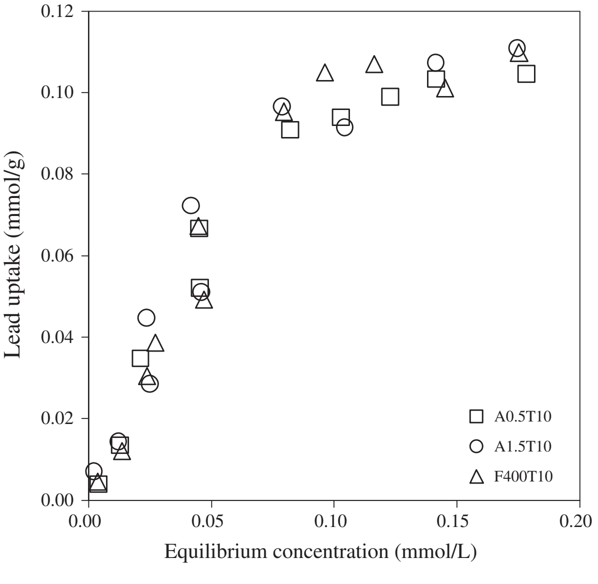

Fig. 7. Equilibrium uptake of lead by 1,000˚C-heat-treated activated carbons.

Table 3
Isotherm constants of lead(II) adsorption onto heat-treated activated carbons by linear and nonlinear approaches

Activated carbon	Langmuir model					
	Nonlinear $q_e = QbC_e/(1 + C_e b)$			Linear; C_e/q_e vs. C_e $(C_e/q_e) = (1/bQ)+(C_e/Q)$		
	Q (mmol/g)	b (L/mmol)	R^2	Q (mmol/g)	b (L/mmol)	R^2
A0.5T10	0.157	13.6	0.978	0.213	6.82	0.717
A1.5T10	0.164	13.3	0.944	0.170	11.7	0.806
F400T10	0.178	11.1	0.950	0.240	6.00	0.628
	Freundlich model					
	Nonlinear $q_e = K_F C_e^n$			Linear; log q_e vs. log C_e log q_e=$(1/n)$ log C_e + logK_F		
	K_F	$1/n$	R^2	K_F	$1/n$	R^2
A0.5T10	0.304	0.548	0.938	0.668	0.867	0.940
A1.5T10	0.311	0.544	0.915	0.432	0.677	0.934
F400T10	0.345	0.586	0.906	0.702	0.870	0.947

$$R_L = \frac{1}{1+bC_0} \tag{3}$$

where b is the adsorption affinity (Langmuir constant), and C_0 is the initial concentration of lead(II). The R_L value indicates the type of isotherm and the nature of adsorption irrespective the shape of isotherm. It suggests whether the adsorption of lead(II) is unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$) or irreversible ($R_L = 0$). Fig. 8 shows the R_L profile of 1,000°C-heat-treated activated carbons.

As can be seen, these activated carbons exhibit the R_L values ranging from 0.25 to 0.93, which are conformed to the favorable region of adsorption at all initial concentrations studied. However, the lowest R_L value recorded in this work was found to be considerably greater than those reported in the literature [30–32], which implies a lower affinity of lead(II) removal onto heat-treated activated carbons.

Fig. 9 shows the equilibrium profile of lead(II) adsorption onto A0.5 series activated carbons. The calculated affinity of A0.5T5 based on Langmuir isotherm was 39.9 L/mmol, which is three times higher than that of A0.5T10. Although the uptake of lead(II) increased with heat treatment, the affinity of removal was found to be reduced. Thus, it is suggested that the electron-dense π-system could adequately attract more lead(II) ions, but the interaction is undeniably weaker. Some researchers have reported that the carboxylic groups, i.e. one of the acidic functional groups, could promote stronger interaction (affinity) with divalent metal ions so as enhancing the adsorption [33,34]. Yet, it should be noted that such functional groups are rare to be readily available on the surface of activated carbon upon activation, in which further (and costly) oxidation process would be required for that purpose.

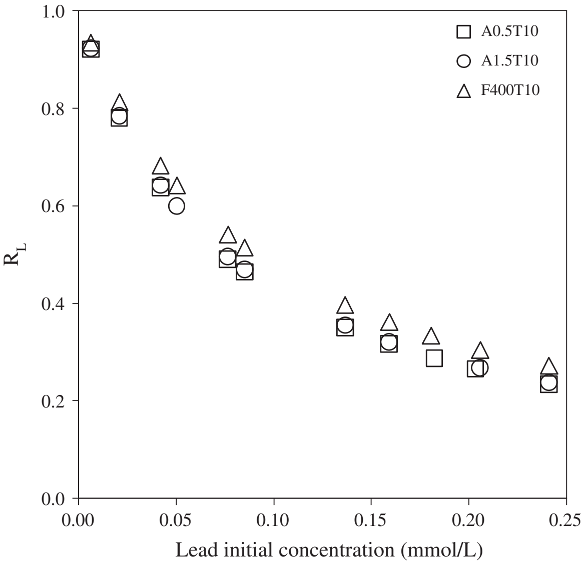

Fig. 8. Langmuir separation factor of 1,000°C-heat-treated activated carbons.

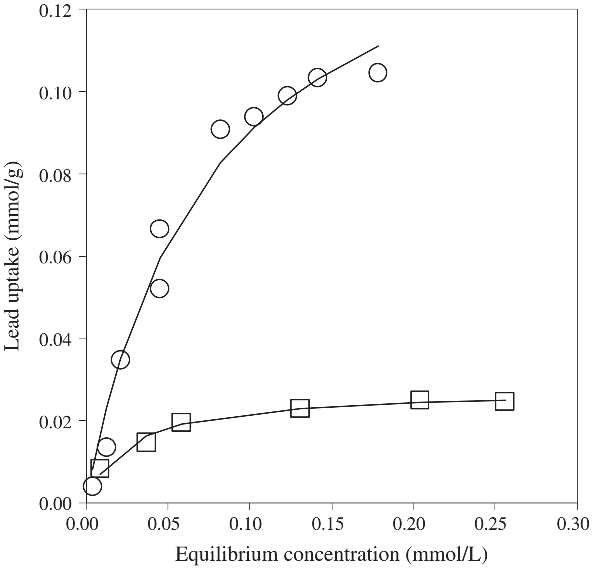

Fig. 9. Experimental data of lead(II) removal by A0.5T5 (□) and A0.5T10 (○). Lines were predicted from Langmuir isotherm.

3.5. Removal mechanisms

Lead(II) adsorption onto heat-treated activated carbons was attained through Cπ-cation interaction. The mechanisms can be visualized in Fig. 10. It is noted that the acidic functional groups pull out the electron density from the π-system, thus making it less nucleophilic. The release of such groups in heat

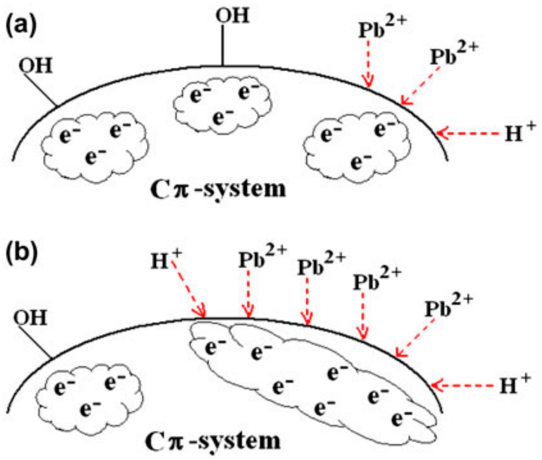

Fig. 10. Schematic representation of lead(II) removal onto (a) 500°C- and (b) 1,000°C-heat-treated activated carbons.

treatment thus eliminates the electron withdrawing effect so as enhancing the electron clouds within the graphitic structure. As can be referred from Table 2, the concentration of phenolic groups of 1,000˚C-heat-treated activated carbons was identical at about 0.28 mmol/g, even though their surface area varies fairly widely (Fig. 1). With that amount of phenolic groups, the density of electron clouds per gram of each activated carbon is expected to be the same. In that sense, it is speculated that more lead(II) ions would be attracted to the π-system as the density of electron clouds increases, to which its influence prevails the contribution of surface area alone.

4. Conclusions

The present study demonstrated the removal of lead(II) ions onto heat-treated activated carbons. Two CMC-derived activated carbons and a commercial F400 were treated under helium at different temperatures to promote different surface and textural characteristics. The BET surface area and the concentration of phenolic groups on the surface of activated carbons were decreased with an increasing treatment temperature. With such deceasing trends, the increase of lead (II) removal was thought to be attributed to an electron-rich Cπ-system. This was due to the diminishing electron withdrawing effect irrespective of the dissimilarity of surface area. The highest lead(II) uptake was similar at 0.110 mmol/g for 1,000˚C-heat-treated activated carbons. In this work, lead(II) adsorption could be reasonably described as monolayer adsorption onto homogenous surface with lower affinity. With the inevitable rise in the price of commercial activated carbon, heat-treated CMC-activated carbon is foreseen as a promising candidate to abate lead(II) contamination in aqueous streams.

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