



Removal of heavy metals in electroplating wastewater by powdered activated carbon (PAC) and sodium diethyldithiocarbamate-modified PAC

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ABSTRACT

We investigated simultaneous removal of heavy metals such as Cr, Ni, and Zn by adsorption onto powdered activated carbon (PAC) and PAC modified with sodium diethyldithiocarbamate (PAC-SDDC). Modification of PAC was confirmed by Fourier transform infrared spectroscopy and Scanning electron microscopy and energy dispersive X-ray spectroscopy. Both PAC and PAC-SDDC reached adsorption equilibrium within 48 h, and the adsorption kinetics followed a pseudo-second order reaction kinetics. The removal of metals was enhanced with increasing both adsorbent dosage and followed the descending order of Cr > Ni > Zn for PAC and Cr > Zn > Ni for PAC-SDDC, respectively. Adsorption kinetics followed pseudo-second order kinetics. Adsorption kinetic results were well fitted by the Freundlich isotherm except for Cr adsorption onto PAC. The optimum pH for heavy metal adsorption onto PAC was 5, whereas that for PAC-SDDC ranged from 7 to 9, indicating that modification of PAC with SDDC significantly enhanced heavy metal adsorption, especially under neutral and alkaline pH conditions. Our results imply that SDDC modified PAC can be applied to effectively remove heavy metals especially Cr in plating wastewaters without adjusting pH from alkaline to neutral.

Keywords: Activated carbon, Adsorption, Heavy metals, Isotherm, Kinetics, Sodium diethyldithiocarbamate

1. Introduction

Industrial wastewater discharge usually contains toxic and harmful substances that can have negative effects on human health and aquatic ecosystems. Every year, large amounts of wastewater containing cyanide (CN⁻) and heavy metals are discharged from alloy, automobile-manufacturing, mining-processing, and metal-plating industries [1, 2]. Metal finishing and electroplating manufacturing units are the major sources of these contaminants, which contribute considerably to the environmental risk and pollution load of the receiving water system and its surroundings [3]. Therefore, the presence of heavy metals (Cr, Ni, and Zn) in such effluents must be controlled, because these contaminants cause environmental problems due to their high toxicity even

at low concentrations [4].

For metal-plating wastewater containing heavy metals, precipitation method is the most widely used conventional treatment process [5]. Most widely used conventional method is precipitation include hydroxide precipitation, and coagulation/flocculation [6]. Among the three aforementioned methods, hydroxide precipitation is the most commonly used because it is a cost effective method for reducing heavy metal concentrations to low levels, and can be easily handled. However, this method does not completely remove metals and produce a huge amount of toxic sludge that needs additional treatment processes [7]. For these reasons, as alternatives to precipitation method, various techniques have been employed including ion-exchange resin, reverse osmosis, electrocoagulation, and adsorption [8]. Except for adsorption,



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those methods can reduce metal ion, however, these methods also have limitations due to high operational cost [6, 8, 9]. Recently, adsorption has recognized as an economical and efficient method for removing heavy metals from polluted waters [10-12].

Even though adsorption is effective in removing heavy metals, but not as effective in removing organic compounds. Hence, various modification of adsorbent has been studied to increase heavy metal removal from water [10, 12, 13]. Among the modification functional groups, dithiocarbamate derivatives can improve metal adsorption abilities by metal ligand formation [13-16]. These dithiocarbamate molecules with large hydrophobic groups strongly adsorb heavy metals to the surface and can modify the characteristics of metal adsorbent to more favorable adsorption conditions [16]. Among the dithiocarbamate derivatives, Monser and Adhoum [17] used sodium diethyldithiocarbamate (SDDC) in the fixed bed modified granular activated carbon (GAC) column. Due to the attachment of SDDC, affinities between heavy metals (Cu, Zn) and the surfaces of GAC were improved [17]. SDDC was also applied as chelating agent in multi-walled carbon nanotubes as adsorbent for solid phase extraction prior to high performance liquid chromatography (HPLC) to measure Ni, cobalt (Co) and mercury (Hg) ions in water samples [18].

Therefore, in this study, as an alternative to conventional treatment methods for removing heavy metals from metal-plating wastewater such as precipitation, we investigated the adsorption of Cr, Zn, and Ni by powdered activated carbon (PAC) and PAC modified with SDDC (PAC-SDDC). We examined the adsorption characteristics of Cr, Zn, and Ni using PAC and PAC-SDDC by determining the adsorption kinetics and isotherms of each adsorbent. We also investigate the effect of initial pH for removal of metals into each adsorbent. Finally, we proposed metal removal mechanism by PAC and PAC-SDDC based on the obtained results.

2. Materials and Methods

2.1. Materials

Commercially available PAC (Shinki Chemical, South Korea) was used as an adsorbent for PAC and PAC-SDDC. The characteristics of PAC are shown in Table 1. SDDC was used (Sigma-Aldrich, USA) to create the modified PAC; Table S1 in Supplementary Materials shows the structure and physicochemical properties of SDDC. A multi-element calibration

Table 1. Characteristics of PAC and SDDC Modified PAC (PAC-SDDC)

Adsorbent	PAC	PAC-SDDC
Zeta potential (mV)	-26.5	-35.8
Surface area ($\text{m}^2 \text{g}^{-1}$)	945	844
Pore volume ($\text{cm}^3 \text{g}^{-1}$)	0.52	0.47
Pore size (\AA)	21.9	22.2
Bulk density	2.28	1.87
pH_{pzc}	10.70	8.53

PAC: powdered activated carbon; SDDC: sodium diethyldithiocarbamate

standard solution dissolved in 5% nitric acid was purchased from Perkin Elmer (USA), and $\text{ZnSO}_4 \cdot \text{H}_2\text{O}$, $\text{K}_2\text{Cr}_2\text{O}_7$, and $\text{NiSO}_4 \cdot 7\text{H}_2\text{O}$ were used for the experiments (Sigma-Aldrich, USA). Trace amounts of 1 N NaOH or H_2SO_4 (Sigma-Aldrich, USA) were used to alter the initial pH of the artificial wastewater. Each solution was prepared with deionized water (Millipore, USA).

2.2. PAC Modification with SDDC

A 12-g sample of dried PAC was mixed with 250 mL of 5.30 mmol/L SDDC solution in a water bath at 50°C at a stirring speed of 200 rpm for 3 d [17, 19]. After this treatment, the mixture was filtered through a $0.45\text{-}\mu\text{m}$ membrane filter (A045A047A, ADVANTEC, Japan) to separate the sorbent, then washed with distilled water. The SDDC-modified PAC was then oven dried at 100°C for 12 h. Both adsorbents were dried at 100°C for 8 h before use.

2.3. Characterization of PAC and Modified PAC (PAC-SDDC)

Scanning electron microscopy (SEM; JSM-7610F, JEOL, USA) and energy dispersive X-ray spectroscopy (EDS; X-MAX 50, Oxford, UK) were used to check the chemical properties and morphology of the adsorbents. To measure the specific surface area of the adsorbents, a surface area analyzer (ASAP 2420, Micrometrics, USA) was used with gaseous nitrogen for surface probing via the Brunauer–Emmett–Teller (BET) method. Zeta analyzer (ELSZ-1000, Photal Otsuka Electronics, Japan) were used to investigate the surface charges of the adsorbents. The functional groups on the surfaces of the adsorbents were analyzed by Fourier transform infrared spectroscopy (FT-IR; VERTEX80v, Bruker, USA). The pH_{pzc} values (point of zero charge) were obtained using the pH drift method [20]. The specific physicochemical properties of each adsorbent are listed in Table 1.

2.4. Adsorption Experiment

To examine the effect of time and initial pH on heavy metal adsorption, bench-scale and batch adsorption experiments were conducted. The initial concentration of each metal (Cr, Ni, and Zn) was set to 50 mg/L in the distilled water samples by adding stock solution. PAC and PAC-SDDC containing 30 g/L at pH 5 to 11 were injected into the artificial heavy-metal wastewater samples. After injection, the samples were agitated at a constant speed of 50 rpm during the reaction. At the final step of the experiment, the samples were collected from the flasks and analyzed. The conditions for the adsorption experiments are summarized in Table S2.

The adsorption capacity q_e (mg/g) at equilibrium was calculated as follows;

$$q_e = \frac{(c_o - c_e)V}{M} \quad (1)$$

where q_e is the adsorption capacities of heavy metal after reached adsorption equilibrium (mg adsorbate/g adsorbent), V is the water volume (mL), C_o is the initial concentration

of heavy metals (mg/L), and C_e is the residual concentrations of 3 metals (mg/L).

To investigate the effect of adsorbent dosage on heavy metal adsorption of PAC and PAC-SDDC and to determine the isotherms, adsorption experiments were conducted individually, with pH dosage adjustment. The total volume of each test sample was 40 mL along with 5, 10, 15, 20, 25, 30, and 35 g/L of PAC and PAC-SDDC. At the first step of the experiments, concentration of 3 metals was set at 50 mg/L, and the initial pH was set to 7.0. All samples collected from each experiment were filtered with membrane filter (0.45 μm , A045A047A, ADVANTEC, Japan), then were diluted before analysis.

2.5. Isotherm

The Freundlich adsorption isotherm applies to adsorption on non-specific and heterogeneous sites on solid surfaces and reportedly well fits heavy metal adsorption results, compared with the Langmuir isotherm [21]. Hence, the isotherm results were fitted to the Freundlich isotherm (Eq. (2)). The relevant equation for the adsorption model is as follows:

$$q_e = K_f C_e^{\frac{1}{n}} \quad (2)$$

where C_e is the equilibrated concentration (mg/L), $1/n$ is the Freundlich intensity parameter, and K_f is the Freundlich adsorption coefficient ((mg/g) (mg/L)^{-1/n}), respectively.

2.6. Analysis of Heavy Metal

Inductively coupled plasma-mass spectrometry (ICP-MS, NEXION 350 D, Perkin Elmer, USA) was used to analyze the concentration of Cr, Ni, and Zn in diluted samples. Table S3 shows the QA/QC data.

3. Results and Discussion

3.1. Characterization of PAC and PAC-SDDC

First, the physicochemical characteristics of each adsorbent were examined. Fig. 1 shows SEM images and EDS patterns of the prepared adsorbents. While the surface of the PAC-only adsorbent was solely composed of carbon, whereas PAC-SDDC consisted of carbon and various inorganic elements (Fig. 1). After modification with SDDC, sulfur in SDDC compound was present on the PAC-SDDC surface. This result implies that SDDC was successfully adsorbed to the PAC surface.

Fig. 2 shows the FT-IR spectrum of each adsorbent; the main FT-IR spectra peak positions of PAC-SDDC showed a C=S band in the range of 950-1,040 cm^{-1} and a C=N band in the range of 1,460-1,550 cm^{-1} , indicating that the modification of PAC with SDDC was successful. The FT-IR spectra of the adsorbents were in agreement with previous researches [16, 22, 23].

The physicochemical properties of each adsorbent are shown in Table 1. The surface area of PAC-SDDC measured using a

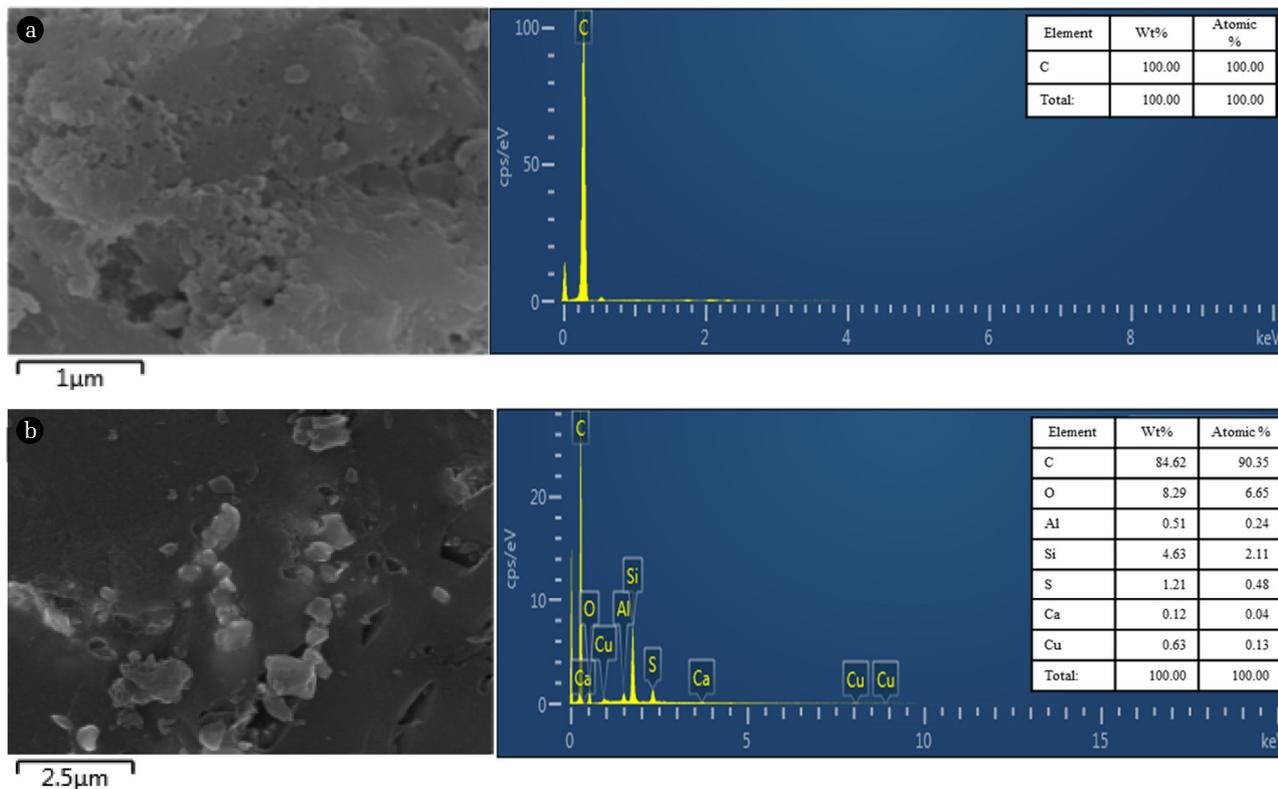


Fig. 1. SEM images and EDS patterns of (a) PAC and (b) PAC-SDDC.

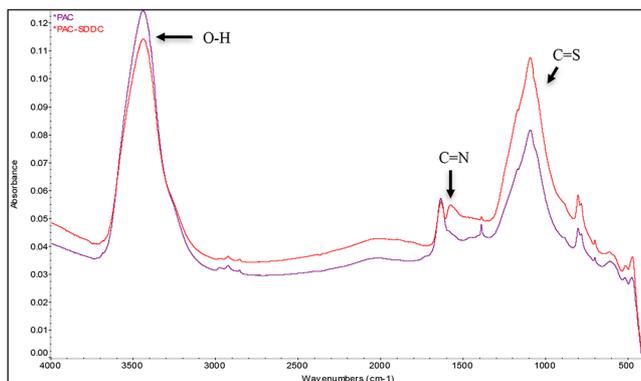


Fig. 2. FT-IR spectrum of PAC and PAC-SDDC.

surface analyzer decreased slightly from 945 to 844 m² g⁻¹ after modification of PAC; the bulk density also decreased from 2.28 to 1.87. However, the pore volume and pore size did not change significantly. Interestingly, the pH_{pzc} value, the pH where the electrical charge density of the surface is zero, decreased from 10.7 for PAC to 8.53 for PAC-SDDC. It was reported that the decrease in pH_{pzc} value can be achieved by the increase in the acidic functional group [24]. In fact, the pK_a of dithiol group in SDDC is known as 2.0 [25], thus the decrease in pH_{pzc} values in PAC-SDDC imply that the acidic moiety increased on the PAC surface such as sodium dithiocarbamate group present in SDDC.

The zeta potential of PAC-SDDC also decreased compared to that of PAC (Table 1). It has been reported that the lower the carboxylic group, the lower the zeta potential value [24]. In addition, FT-IR measurement also indicates that reduction of OH functional group in carboxylic group (-COOH) of PAC due to SDDC attachment on adsorbent.

3.2. Adsorption Kinetics

The effect of time during competitive metal removal kinetics by PAC and PAC-SDDC was measured. Fig. 3 shows the effect

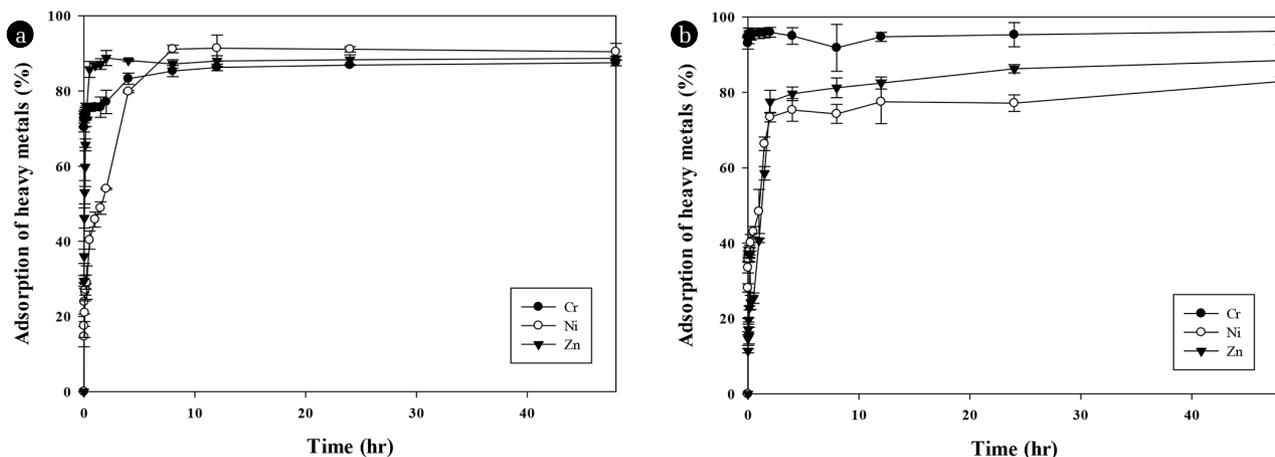


Fig. 3. Effect of time on the adsorption removal of heavy metals using (a) PAC and (b) PAC-SDDC ([C₀] = 50 mg/L; pH = 7.0 ; temperature = 25°C; time = 0-48 h; [PAC] and [PAC-SDDC] = 30 g/L; n = 3).

of time on heavy metal adsorption using PAC and PAC-SDDC. The optimal time for heavy metal adsorption on PAC was 8 h and for PAC-SDDC was increased to approximately 12 h (Fig. 3). In addition, Zn showed the highest adsorption rate on PAC followed by Cr, then Ni. In contrast, Cr showed the highest adsorption rate on PAC-SDDC followed by Zn, then Ni (Fig. 3). Gomes *et al.* [25] reported that heavy-metal adsorption competition sequences were variable, but statistically the most common sequence was Cr, followed by Zn, then Ni. In this study, PAC did not follow this rule while PAC-SDDC did. There was no clear rationale for this phenomenon and further study is needed.

The adsorption kinetics of activated carbon can be fitted to a pseudo-first order equation (Eq. (3) and (4)) and a pseudo-second order equation (Eq. (4) and (6)) [26, 27]. The pseudo-first-order equation (Eq. (3)) describes the adsorption of activated carbon based on the metal concentration and the adsorption quantity of the activated carbon:

$$\frac{dq}{dt} = K_1 (q_e - q_t) \quad (3)$$

$$\ln(q_e - q_t) = \ln q_e - K_1 t \quad (4)$$

where q_e and q_t are the adsorption capacities (mg/g) at equilibrium and at time t , respectively. The pseudo-first-order rate constant (min⁻¹) can be obtained by plotting a graph of $\ln(q_e - q_t)$ versus time.

In contrast, the rate of the pseudo-second-order reaction depends on the amount of heavy metal adsorbed on the adsorbent. The pseudo-second-order model is described by Eq. (5) and (6);

$$\frac{dq}{dt} = K_2 (q_e - q_t)^2 \quad (5)$$

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad (6)$$

where, K_2 is the pseudo-second-order rate constant (g/mg/min)

Table 2. Adsorption Kinetics Rate Constants and R^2 Values of 3 Metals on PAC and PAC-SDDC ($[C_0] = 50$ mg/L; pH = 7.0; temperature = 25°C; time = 0.5-120 min; [PAC] and [PAC-SDDC] = 30 g/L)

Adsorbent	Metal	Pseudo first-order kinetics		Pseudo second-order kinetics	
		K_1 (min^{-1})	R^2	K_2 ($\text{g}/(\text{mg}\cdot\text{min})$)	R^2
PAC	Cr	0.035	0.502	0.789	1.000
	Ni	0.023	0.933	1.119	0.988
	Zn	0.044	0.822	0.669	0.999
PAC-SDDC	Cr	0.024	0.332	0.626	1.000
	Ni	0.015	0.351	0.825	0.974
	Zn	0.012	0.847	0.813	0.961

and can be obtained by plotting a graph of t/q_e versus t .

The adsorption kinetic results are shown in Table 2 and Fig. S1; both PAC and PAC-SDDC adsorbents were well fitted into the pseudo-second order equation. Several researches reported that fitting to the pseudo-second order equation imply that adsorption processes are governed by chemisorption [28, 29].

3.3. Effect of Adsorbent Dosage

The role of adsorbent dosage in heavy metal removal was also investigated. Adsorbent dosage plays a major role in the adsorption of contaminants, given that an increase in the adsorbent dosage creates more reaction sites for adsorption [30]. However, excessive use of adsorbent can worsen the cost-effectiveness during the water treatment process [31]. For this reason, the optimization of the adsorbent dosage of PAC and PAC-SDDC is needed.

Fig. 4 shows the improved removal of heavy metals at higher amounts of PAC at pH 7. While with increasing amounts of PAC from 5 to 30 g/L, increased Ni removal from 74.3 to 91.2%, of Zn from 82.9 to 90.1% and of Cr 89.0 to 89.5% were observed, the same increased PAC-SDDC amounts improved the removal from 65.6 to 81.7% for Ni, from 72.7 to 85.0% for Zn, and from 92.1 to 96.9% for Cr, respectively. The result indicates that increasing the adsorbent dosage led to an enhancement in removal efficiency due to a greater surface area of adsorbent. Our result

indicates that the optimization between adsorbent dosage and metal removal is needed for application to metal plating wastewaters. However, the less removal of metal was achieved when PAC-SDDC was used than PAC (Fig. 4). This result indicates that PAC is more effective for adsorption of Cr, Ni, and Zn compared to PAC-SDDC at least in neutral pH conditions, and might be due to that some of the reaction sites for metal adsorption in PAC are replaced with SDDC in PAC-SDDC adsorbent.

3.4. Adsorption Isotherm

Isotherms for heavy metal adsorption results were fitted independently to the Freundlich model, and fitting parameters are shown in Table 3 and Fig. S2. The goodness-of-fit (R^2) shows that the Freundlich isotherm was appropriate for adsorption of each heavy metal on PAC and PAC-SDDC except for Cr on PAC. When PAC was used as an adsorbent, Ni showed the highest K_f and $1/n$ values, and Cr showed the lowest K_f and $1/n$ values among three metals. However, using PAC-SDDC as an adsorbent, Cr showed the highest K_f and $1/n$ values, and Ni showed the lowest values. It was reported that the adsorption process is governed mainly by intra-particle diffusion, when the Freundlich intensity parameter ($1/n$) is 0.5 [32, 33]. Thus, our result indicates that this phenomenon played a key role especially during the adsorption of Cr ($1/n = 0.50$) and Zn ($1/n = 0.49$) into PAC-SDDC.

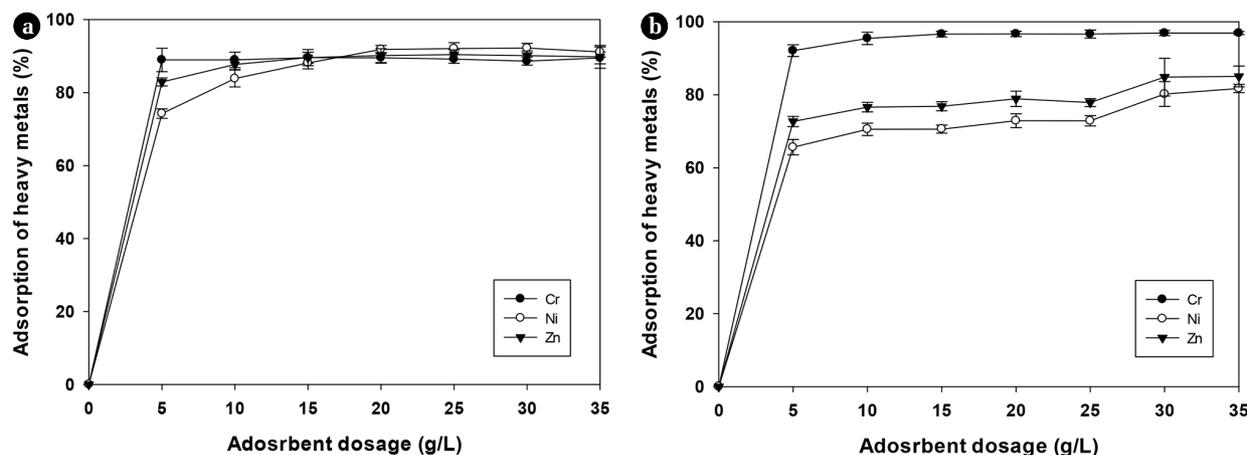
**Fig. 4.** Effect of adsorbent dosage on the adsorption of each metal using (a) PAC and (b) PAC-SDDC ($[C_0] = 50$ mg/L; pH = 7.0 ; temperature = 25°C; time = 48 h; [PAC] and [PAC-SDDC] = 5 to 35 g/L; $n = 3$).

Table 3. Freundlich Isotherm Fitting Parameters for Adsorption of 3 Metals on PAC and PAC-SDDC ($[C_0] = 50$ mg/L; pH = 7.0 ; temperature = 25°C; time = 48 h; [PAC] and [PAC-SDDC] = 5-35 g/L)

Adsorbent	Metal	Freundlich Coefficient		
		K_f (mg g ⁻¹)(mg /L) ^{-(1/n)}	1/n	R ²
PAC	Cr	NA	NA	NA
PAC	Ni	0.32×10^{-3}	0.72	0.986
PAC	Zn	0.80×10^{-3}	0.41	0.956
PAC-SDDC	Cr	4.13×10^{-3}	0.50	0.947
PAC-SDDC	Ni	1.35×10^{-6}	0.38	0.802
PAC-SDDC	Zn	0.20×10^{-3}	0.49	0.806

3.5. Effect of Initial pH on Adsorption

To examine the effect of initial pH on adsorption, adsorption experiments were conducted under different pH conditions. Fig. 5 shows the effect of initial pH on heavy metal adsorption. As the pH increased, the adsorption removals of Ni and Zn were enhanced slightly whereas the Cr removal rate decreased significantly when PAC was used (Fig. 5(a)). Mishra and Patel [34] reported that the higher concentration and mobility of H⁺ ions at acidic pH condition resulted in decreased adsorption of Ni and Zn due to competitive adsorption between metal and H⁺ ions. The surface of PAC can be mainly covered by H⁺ ions when the pH is lower than the p*H*_{pzc} value (10.7) of PAC. For this reason, the adsorption of metal cations (Ni and Zn) can decrease at lower pH value due to limited available binding sites on the PAC surface. However, when the pH increases, the competition between metals and H⁺ ions can decrease, thus adsorption of Ni and Zn can increase.

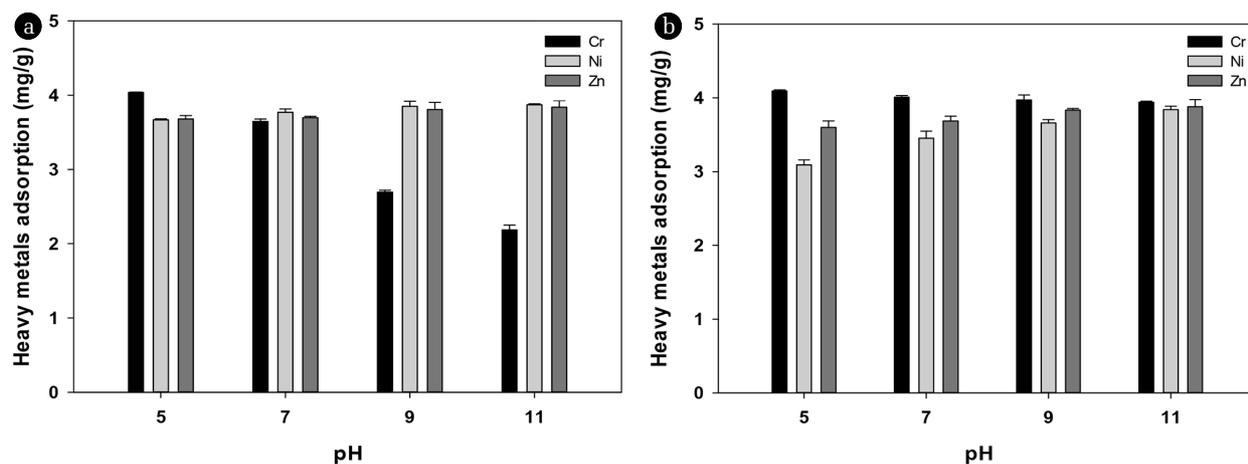
The effect of pH on the adsorption can also be explained by surface complex formation theory (SCF), in which it is assumed that adsorption of metal ions and anions is mainly rely on pH and on the release of OH⁻ ions and H⁺ [35]. Moreover, the increased pH can lead to binding of metals with OH⁻ ions, which results in enhancement of hydrophobicity of metals. This has a positive

effect on metal adsorption because hydrophobic contaminants are removed more readily than hydrophilic contaminants [36].

However, Cr adsorption in PAC decreased sharply by increasing pH (Fig. 5(a)); this result can be attributed to surface charge and shift in the ionic species in the solution [37]. In aqueous solution, Cr can exist in the form of H₂CrO₄, HCrO₄⁻ and Cr₂O₇²⁻ ions under acidic pH conditions, but shifts to bivalent CrO₄²⁻ as pH increases over 6.0 [38]. This bivalent ion (CrO₄²⁻) becomes dominant in alkaline pH condition, and therefore twice the number of sites of univalent charge is needed to neutralize [39]. As a result, the adsorption efficiency of Cr into adsorbent significantly decreases in general.

In contrast to PAC, PAC-SDDC showed a significantly different adsorption tendency for heavy metals. The adsorption removal of Ni and Zn were enhanced as the pH increased, but consistently lower than compared with PAC in all pH ranges, especially under acidic and neutral pH conditions (Fig. 5(b)).

The FT-IR spectrum in Fig. 2 shows that there was a decrease in the negatively charged hydroxyl functional group (O-H band at 3,400 cm⁻¹) after SDDC modification, which is mainly assigned to the carboxylic group O-H stretching in PAC [40, 41]. This functional group in PAC is known to have an effect on heavy metal removal (Eq. (7) and (8)) [42].

**Fig. 5.** Effect of initial pH on the adsorption removal of Cr, Ni, and Zn using (a) PAC and (b) PAC-SDDC ($[C_0] = 50$ mg/L; pH = 5 to 11; temperature = 25°C; time = 48 h; [PAC] and [PAC-SDDC] = 30 g/L; n = 3).



Therefore, consistently lower adsorption of Ni and Zn on PAC-SDDC compared to PAC indirectly showed that SDDC was successfully adsorbed to the PAC surface, as the ligand can change OH groups in adsorbent to form ligand surface complexes [35].

However, the adsorption removal of Cr was affected only slightly by pH change, and increased significantly over the entire pH ranges in PAC-SDDC compared to PAC (Fig. 5(b)). This result can be explained by the complex formation between dithiocarbamate group in SDDC and Cr ion [16, 43]. It is reported that SDDC has versatile binding abilities and forms complexes with most transition metals, especially Cr species [44]. Through this process, negatively charged Cr can approach the adsorbent surface and form complexes with SDDC such as Cr(DDC)₃ and Cr(DDC)₂(ODDC). In fact, due to this phenomenon, SDDC was applied for Cr species analysis including CrO₄²⁻ and Cr₂O₇²⁻ [45, 46]. Setiyanto et al. [47] also reported that Cr(DDC)₂(ODDC) is a majority product of reaction resulted from its higher dissociation energy (Cr(DDC)₃: -3.24 eV; Cr(DDC)₂(ODDC): -10.7 eV) and chemical stability. Monser and Adhoum [17] also reported enhancement of Cr removal by modified GAC (granular activated carbon) using SDDC.

Our results imply that PAC-SDDC can remove Cr more effectively than regular PAC especially at alkaline pHs. Since metal plating wastewater usually have alkaline pH, PAC-SDDC can be effectively used to treat metal plating wastewater containing especially Cr without decreasing pH.

4. Conclusions

In this study, the removal of Cr, Ni, and Zn metals using PAC and SDDC-modified PAC (PAC-SDDC) was investigated by examining the effects of time, adsorbent dosage, and initial pH on the adsorption kinetics of each adsorbent. PAC modification was confirmed by FT-IR, SEM, EDS, and physicochemical measurements. Increasing the time enhanced the adsorption of heavy metals. Both adsorption reactions followed a pseudo-second order reaction. The removal efficiency increased with adsorbent dosage. The removal of heavy metals using PAC followed the descending order of Cr > Ni > Zn; for PAC-SDDC, the removal results were in the order Cr > Zn > Ni. Adsorption removal results were in good agreement with the Freundlich isotherm, except for Cr adsorption on PAC. While PAC showed efficient removal of Cr, Ni and Zn at acidic and neutral pHs, PAC-SDDC was effective especially Cr at alkaline pH conditions. Our results indicate that SDDC-PAC can be used to remove these alkaline metal plating wastewaters, especially Cr without decreasing pH.

Further investigations of adsorption process optimization to apply PAC-SDDC to remove heavy metals from metal-plating wastewaters. The effects of pre-oxidation of cyanide using the alkaline chlorination or AOP process such as UV-H₂O₂ process on adsorption are required.

Supplementary Materials

Table S1 shows the structure of SDDC and its physicochemical property. Experimental conditions of adsorption experiment are summarized in Table S2, and Table S3 shows the QA/QC results during heavy metal measurement using ICP-MS. Fig. S1 shows the pseudo-second order plot obtained by the linear regression method for the adsorption of each metal onto PAC and PAC-SDDC. Finally, Freundlich isotherm plot obtained by the linear regression for the adsorption of 3 metals onto PAC and PAC-SDDC is provided in Fig. S2.

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