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A proposed activated carbon for the removal of silver nanoparticles from water environment

Suhartono Suhartono [1], Achmad Syafiuddin [2,*]

[1]Department of Statistics, Institut Teknologi Sepuluh Nopember, 60111 Surabaya, East Java, Indonesia
[2]Department of Water and Environmental Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia
*corresponding author e-mail address: ***ac.syafi@gmail.com*** | Scopus ID 56682128000

ABSTRACT

This work aims to propose an activated carbon derived from natural material for the removal of silver nanoparticles (AgNPs) from water environment. The activated carbon was derived from coconut shells via chemical approach. For the description of kinetic mechanism, several kinetic adsorption models were employed. They are the pseudo-first-order, pseudo-second-order, Power, Avrami, mixed 1,2-order, fractal-like pseudo-first-order, and fractal-like pseudo-second-order. This investigation was carried out using a batch experiment. Evaluation of the models was carried out using six statistical indicators. This investigation exhibited that the proposed activated carbon had capability to remove AgNPs from water environment. Kinetic adsorption behavior can be described well using the mixed 1,2-order compared to other employed models. In general, this study successfully proposed activated carbon derived from natural resource for the removal of AgNPs from the environment. This study is useful for the development of natural adsorbent materials for various pollutants removal.

Keywords: *Activated carbon; silver nanoparticles; pollutant removal.*

1. INTRODUCTION

The occurrence of nanoparticles pollutant in environment because of industrialization and urbanization has become a major concern [1-4]. Silver nanoparticles (AgNPs) have been intensively explored for various applications [5-9]. However, AgNPs have also been toxic to aquatic biota. It is well-known that the occurrence of AgNPs in the environment was released from industrial products [10,11]. Therefore, their removal from the water environment is urgently needed.

Since adsorption has been ratified to have capability for removal of heavy metals from aqueous system, it recently becomes attractive research subject to be done [12]. For instance, glass beads were employed as an adsorbent for removal of AgNPs [13]. It was found that AgNPs can be removed up to 75% by their proposed adsorbent. Alternatively, the aged iron oxide magnetic particles were also proposed for the removal of AgNPs from aqueous solution [14]. Interestingly, almost 100% AgNPs were removed using the proposed adsorbent. It is also noticed that a high adsorption capacity of 909.1 mg/g was achieved when the nitrogen rich core-shell magnetic mesoporous silica was used as an adsorbent to remove AgNPs [15]. Moreover, a commercial activated carbon, Norit® CA1, was used as an efficient adsorbent for the adsorption of AgNPs [16]. It was also found from their experiment that the adsorption efficiency ranging from 30% to 80% can be achieved using the employed activated carbon.

Several mathematical models have been intensively used for the understanding of AgNPs adsorption mechanism. For instance, four kinetic models have been investigated to understand the kinetic mechanism of AgNPs adsorption [17]. Their study reported that the Pseudo-second-order showed the best performance for the describing adsorption process compared to other employed mathematical models.

A similar finding was also obtained from previous work when several models were used for explaining the behaviors of AgNPs adsorption by following the pseudo-second-order [14]. It is noted that the pseudo-second-order performed well for various data. However, inconsistency can be seen when *Aeromonas punctata* was used as the adsorbent to remove AgNPs from aqueous solution [18]. It can be concluded that AgNPs adsorption experimental data can be well demonstrated when the Pseudo-first-order was employed compared to that of the Pseudo-second-order.

Although the aforementioned common models have been examined, their suitability for various data is still questionable. It is noticed that the mathematical kinetic models are commonly derived and performed for a particular case depending on the experimental conditions and adsorbents. In addition, performance of current established kinetic models depends highly on nature properties of pollutants Therefore, an effort to provide a general mathematical model to describe kinetic adsorption of AgNPs is crucially needed.

In closing the research gap, the present study aims to propose and evaluate an activated carbon derived from natural resource for the removal of AgNPs from water environment. In addition, seven kinetic models, which are the pseudo-first-order, pseudo-second-order, Power, Avrami, mixed 1,2-order, fractal-like pseudo-first-order, and fractal-like pseudo-second-order for describing the kinetic adsorption were also comprehensively evaluated using six statistical indicators. Outcomes from this study are beneficial for a complete understanding of the adsorption mechanisms that are important for improvement of adsorption capacity and other related properties for future application.

2. MATERIALS AND METHODS

2.1. Materials.

In this work, coconut shells were collected from a location in Johor Bahru, Malaysia. Silver nitrate ($AgNO_3$) as a silver salt was supplied by QReC, Auckland, New Zealand. *Eleusin indica* was used as reducing as well as stabilizing agents to produce AgNPs. *Eleusin indica* can be categorized as the species of grass in the Poaceae family and commonly used for animals food. Zinc chloride ($ZnCl_2$) obtained from QReC, Auckland, New Zealand was used as an activation agent to produce activated carbon material. Solutions were prepared using the ultrapure water with resistivity 18.2 M cm supplied by the Arium Ultrapure Water System, Sartorius Malaysia Sdn Bhd, Kuala Lumpur, Malaysia.

2.2. AgNPs synthesis.

To remove any impurities, 18 g of fresh *Eleusin indica* was washed using tap water and following by the ultrapure water three times each. The extraction was carried out in a 500 mL Erlenmeyer flask containing 200 mL ultrapure water and 18 g of fresh *Eleusin indica*. The mixture was then boiled for 30 min. After cooled at room temperature, the extracts were filtered using a nylon membrane filter of 0.45. AgNPs production was prepared by mixing $AgNO_3$ solution with a concentration of 0.15 M with *Eleusin indica* extract with a ratio of 1:1 (volume of $AgNO_3$ solution: volume of *Eleusin indica* extract) and then stirred for 24 h to complete the reduction of the silver ions into the nanoparticle.

2.3. Activated carbon production.

Coconut shells were firstly washed using tap water and following by the ultrapure water several times to completely remove any impurities before ground and sieved to obtain pieces of irregular shapes with the sizes of <3 mm It was then dried in an oven at a temperature of 70 °C for 24 h. The shells were mixed with $ZnCl_2$ at a weight ratio of 1:1 in the ultrapure water for 24 h. Next, it is dried in an oven at a temperature of 110 °C for 24 h. The activation for activated carbon production was carried out by heating the treated activated carbon at a temperature of 500 °C for 1 h. The produced activated carbon was washed using the ultrapure water three times and then dried in an oven at a temperature of 110 °C for 24 h.

2.4. Adsorption experiment.

AgNPs solution (200 mg/L) was prepared by series dilution procedure using the ultrapure water. The adsorption was carried out by mixing adding 1 g of the activated carbon into 10 mL AgNPs solution and continued by magnetically stirring the mixture for 30 to 120 min. Next, adsorption capacity was estimated using the following formula:

$$q(t) = \frac{\left(C_i - C(t)\right) \square V}{W} \qquad (1)$$

where $q(t)$ is the capacity of adsorption in the unit of mg/g), C_i is the initial concentration in the unit mg/L, $C(t)$ is the final concentration at time t in the unit of mg/L, V is the volume of solution in the unit of L, and W is weight of the activated carbon in the unit of g used for the removal of AgNPs.

2.5. Characterizations.

AgNPs concentration was estimated using the inductively coupled plasma optical emission spectroscopy (ICPOES). The digestion was carried out using the Milestone Start D model (No.134263) with 12 digestion vessels. Surface morphologies were characterized using the field emission scanning electron microscopy (FESEM ZEISS Supra 35VP) operated at an accelerating of 5 kV.

2.6. Kinetics models.

The pseudo-first-order model was initially proposed as a first-order rate model. The model can be expressed mathematically as:

$$q_t = q_e\left[1-\exp\left(-k_{p1}t\right)\right] \qquad (2)$$

where q_e is the adsorption capacity at equilibrium (mg/g), and k_{p1} is the constant of the model (min^{-1}).

The Pseudo-second-order model is known as a kinetic model and was used for the adsorption of divalent metal ions [19]. This model is expressed as:

$$q_t = \frac{k_{p2}q_e^{\,2}t}{1+q_e k_{p2}t} \qquad (3)$$

where k_{p2} is the constant of the model (min^{-1}).

The nonlinear power is given as [20]:

$$q_t = k_p t^{v_p} \qquad (4)$$

where k_p and v_p are the constants of the model.

Avrami kinetic model is widely used to describe kinetic mechanism of several sorbate such as methylene blue or Hg(II) [21,22]. It is expressed as:

$$q_t = q_e\left[1-\exp\left(-k_{av}t\right)^{n_{av}}\right] \qquad (5)$$

where k_{av} and n_{av} are the constant of the model (min^{-1}) and form of the fractional reaction order of the model (-), respectively.

The mixed 1,2-order model is mathematically expressed as [23]:

$$q_t = q_e\frac{1-\exp(-kt)}{1-f_2\exp(-kt)} \qquad (6)$$

where f_2 and k are the constants of the model.

A modification of the pseudo-first-order model by introducing a fractal concept was also carried out to propose a new model called fractal-like pseudo-first-order [24]. The model is expressed as:

$$q_t = q_e\left[1-\exp\left(-k_{flfo}t^{\alpha}\right)\right] \qquad (7)$$

where k_{flfo} is the coefficient of the model and α is the constant of the model.

A modification of the pseudo-second-order model by introducing a fractal concept was also proposed [24]. The model can be described mathematically as:

$$q_t = \frac{k_{flso}q_e^{\,2}t^{\alpha}}{\left[1+q_e k_{flso}t^{\alpha}\right]} \qquad (8)$$

where k_{flso} is the fractal-like pseudo-second-order coefficient and α is the constant of the model.

2.7. Parameter estimation.

For optimizing parameter of all employed models, the MATLAB Optimization Toolbox curve fitting function, ''lsqcurvefit'' was used. Optimization procedure was carried out by applying the Trust Region Reflective Newton algorithm. Statistical indicators, namely, the coefficient of determination (R^2), root mean squared error (RMSE), percentage of error in maximum estimated value (E_{max}), percentage of error in minimum

estimated value (E_{min}), mean absolute percent error (MAPE), and mean absolute deviation (MAD) were applied for evaluation purpose. The best performance was indicated based on the minimum arithmetic rank. For a comprehensive overview, all statistical indicators are presented as follows:

$$R^2 = 1 - \sqrt{\frac{\sum (x_{obs,i} - x_{model,i})^2}{\sum (x_{obs,i} - \bar{x}_{obs})^2}} \quad (9)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n} (x_{obs,i} - x_{model,i})^2}{n}} \quad (10)$$

$$E_{max} = \left| \frac{x_{max\,model} - x_{max\,obs}}{x_{max\,obs}} \right| \times 100\% \quad (11)$$

$$E_{min} = \left| \frac{x_{min\,model} - x_{min\,obs}}{x_{min\,obs}} \right| \times 100\% \quad (12)$$

$$MAPE = \left(\frac{1}{n} \sum \frac{|x_{obs,i} - x_{model,i}|}{|x_{obs,i}|} \right) \times 100\% \quad (13)$$

$$MAD = \frac{1}{n} \sum |x_{obs,i} - x_{model,i}| \quad (14)$$

where $x_{obs,i}$ is the observed data. In addition, $x_{model,i}$ is the predicted data and n is the number of data used for the analysis.

3. RESULTS

3.1. Removal of AgNPs using activated carbon.

Removal of AgNPs from water environment becomes a major concern for policy makers since they are associated with their toxicity not only for microorganism but also for public health. United States Environmental Protection Agency (USEPA) has recommended the allowable limit for AgNPs in drinking water by 0.1 mg/L. Therefore, a suitable method for the removal of AgNPs until achieving the allowable limit is critical [25-27]. In this study, concentration of AgNPs removed by the proposed activated carbon is presented in Figure 1.

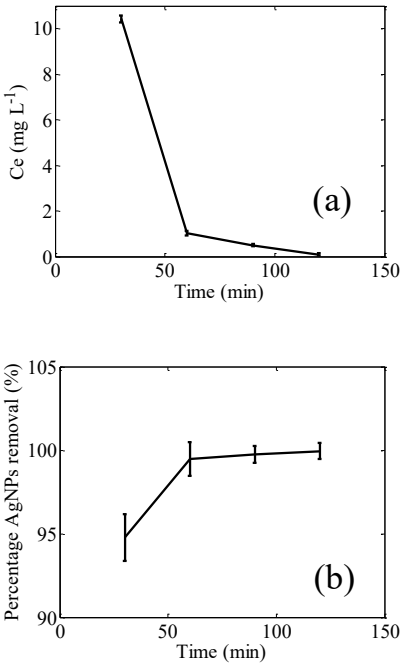

Figure 1. (a) Concentration of AgNPs in the solution and (b) percentage of AgNPs removal.

This study found that AgNPs can be significantly removed from the water for 30 min. The decrease in concentration is consistent until 120 min. The activated carbon can achieve the allowable limit regulated by the USEPA at 120 min. in general, the proposed activated carbon had the capability to remove AgNPs up to 99.9% as shown in Figure 1(b). The proposed activated carbon has the potential for removal of other pollutants [28-31].

Deposition of AgNPs via the presently proposed activated carbon can be observed in Figures 2(a)-2(b). The surface morphology revealed that almost all activated carbon surface is enveloped by AgNPs as shown in Figure 2(b). In general, deposition of AgNPs onto activated carbons depends highly on the physio-chemical properties of both activated carbon and nanoparticles. In addition, other parameters such as environmental condition and procedure for removal are also critical.

Figure 2(a) shows the surface morphology of activated carbon after the adsorption. The distribution of AgNPs on the activated carbon is depicted in Figure 2(b). By considering the surface morphology characterization, possible rules of the deposition of AgNPs on the activated carbon surface can be described as follows. AgNPs can be completely deposited on the carbon surface. Alternatively, it is also possible that AgNPs can enter the activated carbon pores. Moreover, AgNPs can agglomerate before continuing the attachment of AgNPs on the surface of activated carbon.

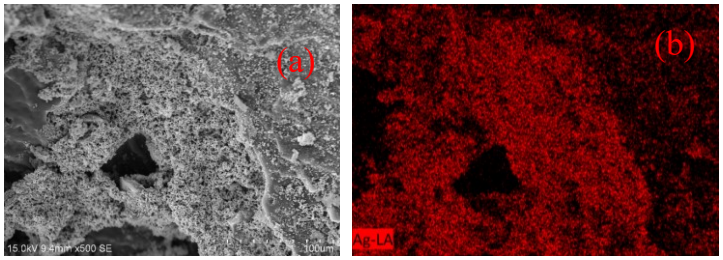

Figure 2. (a) Surface morphology of activated carbon after the adsorption and (b) distribution of AgNPs on the activated carbon.

3.2. Kinetic adsorption behaviors.

Figure 3 shows the experimental adsorption data and optimization using the kinetic models. Table 1 lists the optimized parameters for all employed models. All models containing q_e parameter including pseudo-first-order, pseudo-second-order, Avrami, mixed 1,2-order, fractal-like pseudo-first order, and fractal-like pseudo-second order exhibited good performance with the data obtained from the batch experiment. Specifically, the corresponding q_e values are 1.99 mg g^{-1}, 0.21 mg g^{-1}, 1.99 mg g^{-1}, 1.99 mg g^{-1}, 1.99 mg g^{-1}, and 2.00 mg g^{-1}, respectively. Moreover, the Power model performed in less accurate prediction as shown in Figure 3(a).

3.3. Performance of adsorption kinetic models.

The performance of all employed kinetic models is presented in Figures 3(a)-(g). It is noticed that the pseudo- first-order, Avrami, mixed 1,2-order, fractal-like pseudo-first order, and fractal-like pseudo-second order exhibited good performance.

Alternatively, the pseudo second-order and Power kinetic models show less precise for the data.

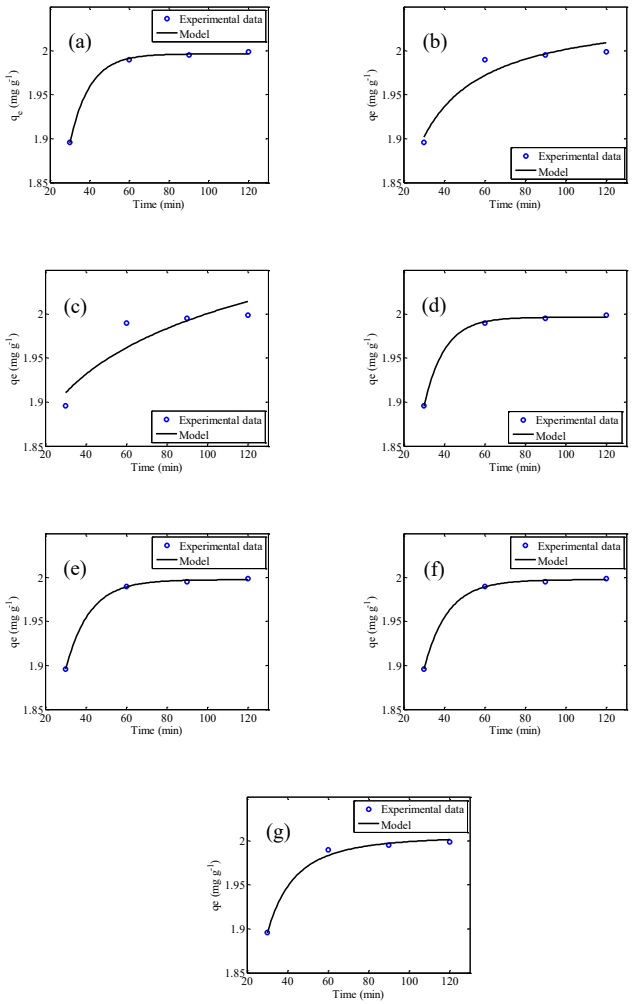

Figure 3. Experimental and simulated data using (a) the pseudo-first-order, (b) pseudo-second-order, (c) Power, (d) Avrami, (e) mixed 1,2-order, (f) fractal-like pseudo-first-order, and (g) fractal-like pseudo-second-order models.

Table 1. Optimum parameters for all employed kinetic models used in this study.

Model	Parameter	Value
Pseudo-first-order	q_e	1.996
	k_1	0.100
Pseudo-second-order	q_e	0.213
	k_2	2.048
Power	k_p	1.678
	v_p	0.038
Avrami	q_e	1.996
	k_{av}	0.315
	n_{av}	0.315
Mixed 1,2-order	q_e	1.998
	k	0.083
	f_2	0.399
Fractal-like pseudo-first-order	q_e	1.998
	k_{flfo}	0.148
	α	0.883
Fractal-like pseudo-second-order	q_e	2.006
	k_{flso}	0.003
	α	2.361

Moreover, Power model exhibited the lowest performance compared to others. Average ranking (AR) of all kinetic models for describing adsorption of AgNPs using the proposed activated carbon is given in Table 2. It was observed from the table that the Power kinetic model performed the highest average ranking and the mixed 1,2-order model exhibited the lowest average ranking. It indicated that the models performed the lowest and the highest performances, respectively.

In general, the experimental data can be well described using mixed 1,2-order followed by fractal-like pseudo-first-order, pseudo-first-order, Avrami, fractal-like pseudo-second-order, pseudo-second-order, and Power kinetic models. This is evidence that the pseudo-second-order that is commonly employed for various AgNPs adsorption data with the best performance is inappropriate for this work [14].

Table 2. Average ranking (AR) of all kinetic models

Model	R^2	RMSE	E_{max}	E_{min}	MAPE	MAD
A	3	3	4	3	3	3
B	6	6	6	6	6	6
C	7	7	7	7	7	7
D	4	4	5	4	4	4
E	1	1	2	1	2	1
F	2	2	1	2	1	2
G	5	5	3	5	5	5

A-G refer to Pseudo-first-order, Pseudo-second-order, Power, Avrami, Mixed 1,2-order, Fractal-like pseudo-first-order, and Fractal-like pseudo-second-order models.

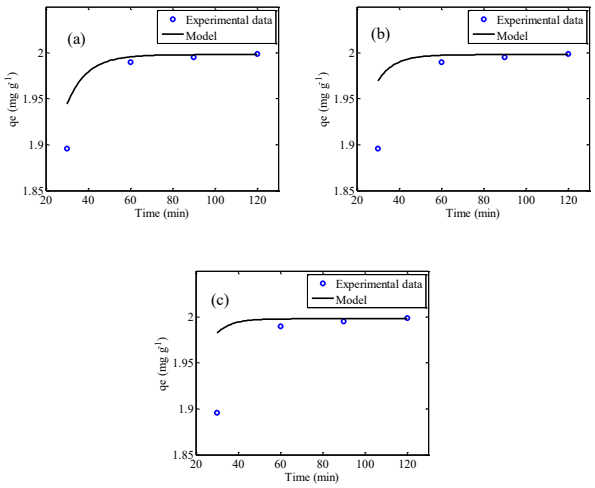

Figure 4. Effects of k variation on the performance of the mixed 1,2-order model for (a) 25, (b) 50, and (c) 75% variations.

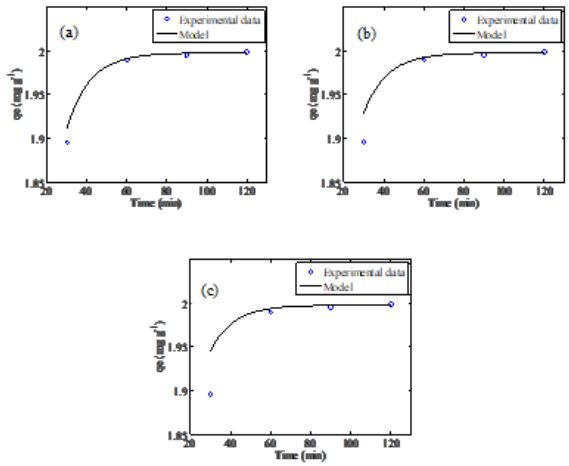

Figure 5. Effects of f_2 variation on the performance of the mixed 1,2-order model for (a) 25, (b) 50, and (c) 75% variations.

3.4. Sensitivity analysis for the best kinetic model.

This study confirmed that the mixed 1,2-order kinetic model exhibited the highest performance compared to all models. To investigate the model sensitivity by variation of its parameters

such as k and f_2, this work evaluated the effects of increasing parameters from 25% to 75% on the performance (see Figures 4 and 5).

Figures 4 and 5 show the model performance under the variated parameters. This study confirmed that the variation of k from 25 to 75% significantly affected R^2, MINE, and MAPE. It indicates that it is more sensitive to these statistical parameters compared to others. Conversely, variation of k did not affect significantly RMSE, E_{max}, and MAD. In general, it was found that the change of k is more sensitive to R^2. Moreover, the variation of f_2 is more sensitive only on the E_{min} compared to other statistical indicators.

4. CONCLUSION

The aim of this study was to propose and evaluate the activated carbon to remove AgNPs from the water environment. This study found that the proposed activated carbon can successfully remove AgNPs. The kinetic behaviors can be well described using the mixed 1,2-order kinetic model compared to other investigated kinetic models. For the sensitivity analysis, increasing k of the mixed 1,2-order kinetic model exhibited to be more sensitive to R^2, E_{min}, and MAPE. In addition, an increasing f_2 of the model was more sensitive only to E_{min}. This study is beneficial for the development of natural adsorbent materials for removal of pollution in the environment.

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