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Rapid removal of mercury ion (II) from aqueous solution by chemically activated eggplant hull adsorbent

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ABSTRACT

Eggplant hull (EH) as a waste material was introduced as a new adsorbent through activation process followed by a chemical treatment method using H_2O_2 and NH_3. For the first time, the EH adsorption was used for the adsorptive removal of mercury ion (Hg^{++}) from aqueous solutions and the effects of different parameters, including pH of solution, contact time, temperature, and initial concentration, on the adsorption efficiency were studied. The results revealed that the EH, as an effective adsorbent, has high adsorption activity in higher pH values. The temperature study indicated the endothermic nature of the adsorption processes. Also, the Hg (II) adsorption by EH follows the pseudo first order kinetic model and both Langmuir and Freundlich isotherm models. However, Freundlich adsorption model fitted the experimental data better than Langmuir model. The maximum adsorption capacity of EH was obtained 147.06 mg/g which was higher than most conventional adsorbents. The surface activity of EH was estimated using iodine method as 300 mg/g.

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1. Introduction

Water pollution caused by metal ions is considered one of the major environmental problems in many countries which can threaten human health and other living organisms in higher contents than admissible sanitary standards (Ayati et al. 2016). Mercury, as one of the top ten toxic metal ion, is mainly present in the wastewater effluents of various process industries such as paper, pulp and paint, oil refining, rubber processing, and fertilizer plants (Johari et al. 2016). The converting of mercury to methyl mercury, as its organic form, and its bioaccumulation in food chain makes it high toxicity which can constitute a threat to the aquatic life and living creatures (Yu et al. 2014). The exposure of human cells to methyl mercury can lead to neurological effect, mood swings, memory loss, damage of gastrointestinal tract, and the kidneys (Adams et al. 2010). Accordingly, its MCLG (Maximum Contaminant Level Goals) was specified at low level of 0.2 ppb by Environmental Protection Agency (EPA).

Thus, many researchers focused on the mercury removal by different methods (Fu and Wang 2011) such as membrane technology (Fard and Mehrnia 2016), chemical precipitation (Nguyen et al. 2016), ion exchange (Oehmen et al. 2014), coagulation (Samrani et al. 2008), and adsorption (Inbaraj and Sulochana 2006; Zabihi et al. 2010). However, some of these methods such as reverse osmosis and ion exchange are excellent techniques, but adsorption was found a superior to other methods, due to its low cost, simplicity and flexibility, ease of operation (Rafatullah et al. 2010) and removal efficiency. A wide range of adsorbents and biosorbents have been used for mercury ion removal from aqueous solutions, such as activated carbon, as commonly used sorbent (Bhatnagar et al. 2013; De et al. 2013), waste rubber (Gupta et al. 2012), carbon fibres (Nabais et al. 2006), chitosan (Reddy and Lee 2013), polymer composites (Say et al. 2008) and etc. In last decades, many researches have been done to find eco-friendly, low cost, highly active and locally available adsorbents (Rangabhashiyam et al. 2013). In this regard, agricultural wastes, including peanut hull (Liao et al. 2011), sago waste (Kadivelu et al. 2004), Walnut shell (Zabihi et al. 2009), have attracted significant interest as emerging low cost adsorbents of mercury.

In the present work, eggplant hull (EH), as one of the food wastes, was activated by a chemical treatment method using H_2O_2 and NH_3 and introduced as a novel low cost biosorbent for the ion removal from aqueous solution under varied experimental condition. Moreover, the adsorption kinetics and isotherms were studied by the well-known models.

2. Materials and methods

Mercury chloride was obtained from Merck in analytical grade. The sorbent was the eggplant hull as a waste material. It was washed with distilled water to remove impurities and dried at 180 °C for 20 h. The sample was ground and chemical treated by the solution of hydrogen peroxide (2 % vol) and ammonia (2 % vol) for 30 min, in order to extract the soluble organic compounds of hulls and enhance chelating efficiency.

2.1. Batch adsorption

For the batch adsorption studies, 0.1 g of EH adsorbent (fraction with 0.2 mm particles in size) was added to 50 mL of Hg(II) solution with the desired concentration (in the range of 11.2-105.6 mg/L) at pH of 5 in several conical flasks. The pH was adjusted using HCl and NaOH solutions. The suspensions were agitated on mechanical stirrer (720 rpm) for specific time intervals at room temperature. Finally, the adsorbents were separated through microporous filter paper and the solutions were analyzed to obtain Hg (II) ion concentrations by Varin atomic absorption spectrophotometer (spectra-110-220/880, Australia Pty. Ltd.) equipped with a Zeeman atomizer.

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2.2. Characterization of adsorbent (eggplant hull)

The surface activity of EH toward iodine was determined by using the DIN 53582 standard method. The iodine No. of the sample was obtained as 300 mg/g. The apparent density of this sorbent was measured as 25 g/cm^3 using ASTM (D 2845-89).

3. Results and discussion

3.1. Influence of pH

The effect of the solution pH on mercury ion adsorption removal efficiency is illustrated in Fig.1, while the initial concentrations of Hg(II) ions was fixed at 52.6 mg/L. The results show that hydrogen ion concentration is an important parameter in the Hg(II) ions adsorption by EH. So that, the Hg(II) adsorptive removal percentage increases in the range of 2 to 5 and followed a plateau trend for higher pH values. The surface charge is a key factor for the metal ions adsorption (Rio and Delebarre 2003; Budinov et al. 2008). As in our previous study, the surface of food and agricultural wastes has oxygen –functional groups due to presence of lactonic, carboxylic, and phenolic groups on their surface (Zabihi et al. 2010). Furthermore, the surface of EH was treated by the solution of hydrogen peroxide (2 % vol) and ammonia (2 % vol). So it is assumed that, the oxygen-containing functional groups on the EH surface is responsible for mercury adsorption. On the other hand, the Hg(II) ions adsorption may also include surface complexes formation with the functional groups precipitation on the surface of adsorbent. In the presence of Hg (II) in solution, the following surface complexes may be formed (Boehm et al. 1966; Boehm 1994):

$$2C_xOH + Hg^{2+} \rightarrow (C_xO)_2Hg^{2+} + 2H^+ \quad (1)$$

$$2C_xOOH + Hg^{2+} \rightarrow (C_xCOO)_2Hg^{2+} + 2H^+ \quad (2)$$

At pH<4 (acidic medium), the reverse reactions may occurred and mercury ion dissolved in the solution.

3.2. Effect of temperature and contact time

Fig. 2 shows the effect of contact time on the Hg(II) ions adsorption at different temperature. As can be seen, the process is very fast and the ion concentration sharply decreased to attain equilibrium just within 2 to 7 min. This rapid uptake of ions by EH clarifies that it can be considered to apply in economical wastewater treatment. Also, the Hg(II) ions concentration decreased with the increase in temperature indicating an endothermic nature of the adsorption processes. The adsorption capacity enhanced by increasing the temperature suggesting that more surface active sites are available for adsorption at higher temperature which might be attributed to the pore size change as well as diffusion rate enhancement in an endothermic process.

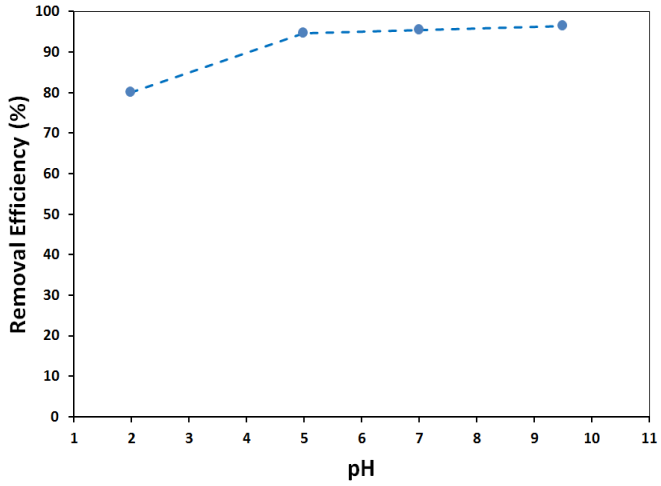

Fig.1. Effect of pH on the removal efficiency of Hg(II) ion (EH dosage=0.1 g, C_0=52.6 mg/l, T=25 ˚C, t=5 min).

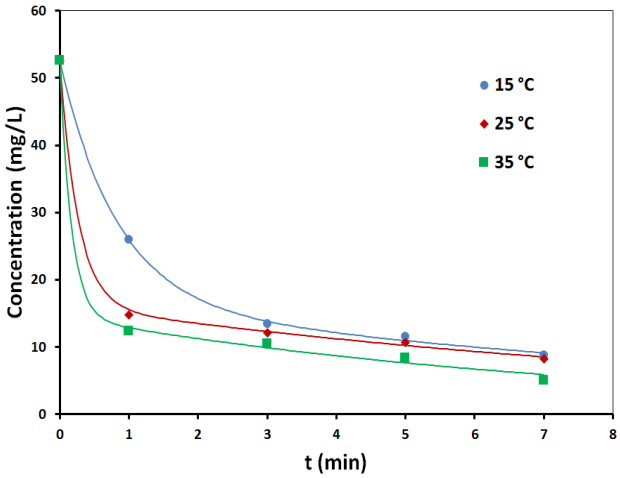

Fig. 2. Effect of contact time on the Hg(II) ion removal at different temperature (C_o=52.6 mg/l, sorbent dose= 0.1 g, T=25 ˚C, pH= 5).

3.3. Effect of initial Hg(II) ion concentration

In this investigation, the effect of initial Hg (II) concentration on the EH adsorption performance was studied using solutions with various concentrations of 11.2, 24.6, 52.6 and 105.6 mg/l at 25 ˚C (Fig.3). It is clear that the Hg (II) ions adsorption amount by EH adsorbent increases with increasing the initial ion concentration. It was very similar to the obtained results of many other Hg(II) ion adsorptive removal studies (Carrott et al. 1998; Namasivayam and Kadirvelu. 1999; Zhang et al. 2004). It might be due the higher probability of collisions between ions and EH particles by increasing the ions concentration.

As can be seen, the ions solutions were equilibrated at first 5 min. Therefore, it was obtained as equilibrium time in other batch experiments. The amount of Hg(II) ions adsorptive uptake (q, mg/g) was calculated by:

$$q = \frac{(C_o - C_e)V}{m} \quad (3)$$

where C_0 and C_e are the initial and equilibrium mercury ions concentrations (mg/L), respectively, V is the solution volume (L), and m is the mass of adsorbent (g). In the presence of 0.1g adsorbent and pH of 5, the equilibrium adsorbed amounts of Hg (II) at 11.2, 24.6, 52.6 and 105.6 mg/L initial concentrations were measured as: 1.8, 8.3, 22.05 and 48.1 mg/g, respectively.

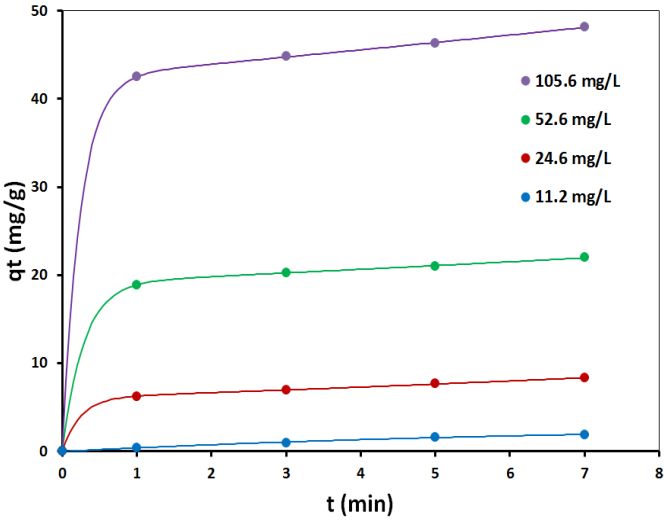

Fig. 3. Effect of initial concentration on the mercury adsorption (EH dose=0.1g, T=25 ˚C, pH=5).

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3.4. Adsorption Isotherm

The adsorption isotherms study of Hg(II) ions removal by EH was carried out by varying the initial Hg(II) ions concentration in the range of 11.2 to 105.6 mg/L at room temperature. The obtained data were correlated with two well-known Freundlich and Langmuir isotherm models for further investigation.

3.4.1. Freundlich model

The Freundlich model, which often gives better fit for adsorption from liquid phases, can be expressed as (Freundlich. 1906):

$$\log q_e = \log K_f + \frac{1}{n}\log c_e \quad (4)$$

where q_e is the equilibrium capacity (mg/g), C_e is the equilibrium concentration of solution (mg/L) and K_f and n are the Freundlich constants. The Freundlich plot of Hg(II) adsorption on EH is shown in Fig. 4a. The plot gives a good fit to the data with correlation coefficient of R^2 = 0.9994. Usually, good adsorbent illustrates 1<n<10. Smaller value of n attributes to better adsorption performance and formation of relatively strong bonds between the adsorbate and adsorbent surface.

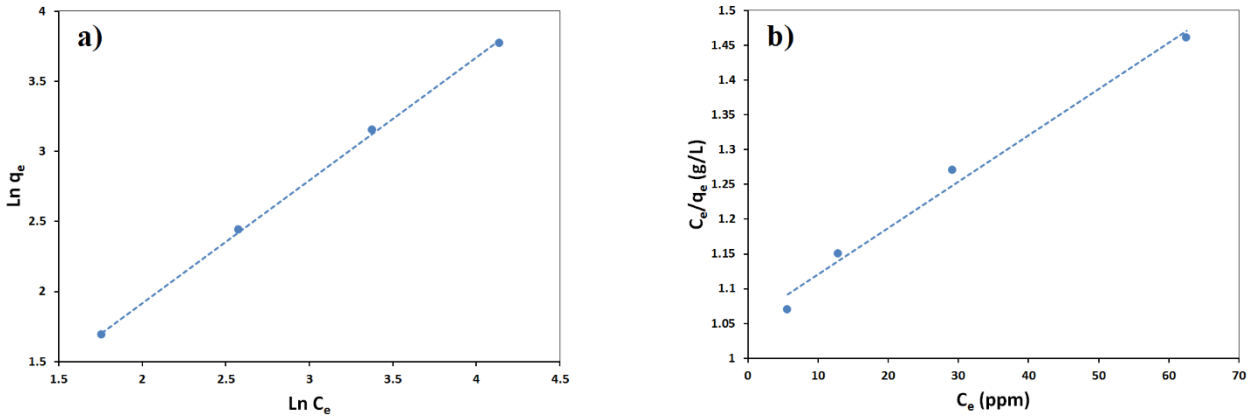

Fig. 4. a) Freundlich isotherm, and b) for adsorption of mercury by EH at 25 ˚C.

3.4.2. Langmuir model

The Langmuir adsorption isotherm was used as follow (Langmuir 1918):

$$\frac{C_e}{q_e} = \frac{1}{q_m b} + \frac{1}{q_m} C_e \quad (5)$$

where q_m is the maximum monolayer capacity of adsorbent (mg/g), obtained by complete monolayer coverage) and b is the Langmuir constant (1/mg). In the present work, we found that the Langmuir isotherm model (shown in Fig. 4b) gives a fairly good fit the experimental data of mercury ions adsorption by EH (R^2 =0.9849). The fitted data of these two models are summarized in Table 1.

Table 1. Measured parameters of the isotherm models for adsorption of Hg (II) onto EH.

Langmuir model			Freundlich model		
q_m (mg/g)	b (l/mg)	R^2	K_f (mg/g)	N	R^2
147.06	0.00648	0.98	1.160	1.136	0.99

From the results, the Freundlich isotherm model predicted the experimental data slightly better than the other isotherm model. The monolayer adsorption capacity of Hg (II) ions on EH was calculated 147.06 mg/g.

3.5. Kinetic studies

The kinetic adsorption data was modeled with pseudo- first- order (Eq. 6) (Lagergren 1898) and pseudo- second –order (Eq. 7) (Ho and McKay 2000) equations which are given as:

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \quad (6)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (7)$$

where q_e and q_t are the amounts of Hg (II) ions uptake by adsorbent (mg/g) at equilibrium and t time (min), respectively, and k_1 is the first-order rate constant (1/min) and k_2 is the second-order rate constant (g/mg min).

The values of different parameters determined from these two kinetic models with their corresponding correlation coefficients are presented in Table 2. The correlation coefficients of the second-order model at all temperatures are more than 0.99, showing that this model can well explain the kinetic adsorption data of mercury on EH.

3.6. Comparison with other adsorbents

The adsorption capacity to EH toward the Hg(II) ion removal was compared with the adsorption capacities of other adsorbents from the literature, as given in Table 3, It shows that the adsorption capacity of eggplant hull for Hg (II) ion is comparable with the many conventional adsorbents.

Table 2. Comparison of the parameters of the (a) pseudo-first-order and (b) pseudo-second-order kinetic models at different temperatures.

(a)

T (˚C)	K_1 (1/min)	q_e (mg/g), Exp.	q_e (mg/g), Cal.	%D	R^2
15	0.3388	21.80	24.15	10.77	0.91
25	0.2538	22.05	21.27	3.53	0.99
35	0.2347	23.55	22.27	5.43	0.98

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(b)

T (°C)	K_2 (g/mg min)	q_e (mg/g), Exp.	q_e (mg/g), Cal.	% D	R^2
15	0.0508	21.80	24.04	10.27	0.99
25	0.3664	22.05	26.54	20.36	0.99
35	0.3937	23.55	29.06	23.39	0.99

Table 3. Monolayer adsorption capacity of various adsorbents for mercury.

Adsorbent type	q_m (mg/g)	Ref.
Activated carbon (fertilizer waste)	3.62×10^{-3}	(Mohan et al. 2001)
Fuller's earth	1.145	(Oubagaranadin et al. 2007)
Silico-aluminous ashes	3.2	(Rio and Delebarre. 2003)
Sulfo-calcic ashes	4.9	(Rio and Delebarre. 2003)
Carbon aerogel	34.96	(Kadirvelu et al. 2008)
Coal adsorbents (Bolluca)	37	(Ekinci et al. 2002)
Sago waste carbon	55.6	(Kadivelu et al. 2004)
Activated carbon	69.44	(Oubagaranadin et al. 2007)
Coal adsorbent (Mengen)	92	(Ekinci et al. 2002)
Activated carbon (Indian almond)	94.43	(Inbaraj and Sulochana. 2006)
Coal adsorbents	105	(Ekinci et al. 2002)
Activated carbon (antibiotic waste)	129	(Oubagaranadin et al. 2007)
Eggplant hull (EH)	147.06	The present study
Furfural	174	(Mohan et al. 2001)
Carbon fibers (Acrylic, Kynol)	290-710	(Nabais et al. 2006)

4. Conclusions

The foregoing study has revealed the feasibility of using a new sorbent derived from eggplant hull (EH) for the rapid removal of Hg(II) ions from aqueous solutions. The adsorption capacity of the EH was obtained 147.06 mg/g at pH of 5.0 for the adsorbent particles with 0.88 mm diameter. The adsorption kinetics and equilibrium data for this process were well described by pseudo-second-order and Freundlich models respectively. The EH was found a cheap and available adsorbent which gives a rapid sorption of mercury ion.

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Nomenclature

q	Amount adsorbed
C_0	Initial metal ion concentration
C_e	Equilibrium concentration
V	Volume of the solution
q_e	Equilibrium amount adsorbed
K_f	Freundlich constants related to adsorption capacity
n	Freundlich constants related to adsorption intensity
q_m	Langmuir constant related to the maximum adsorption capacity
b	Langmuir constant related to the energy or net enthalpy of adsorption
q_t	Amount adsorbed at time t

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