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ORIGINAL ARTICLE

Potentiality of waste human hair towards removal of chromium(VI) from solution: kinetic and equilibrium studies

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Abstract

Industrial and agricultural activities discharges huge amount of hazardous pollutants that lead to massive environmental pollution and health hazards. Keratin is a fascinating protein and useful biopolymer, which is usually found in wool, human hair, nails, feathers, etc. The present research deals with the potentiality of human hair towards removal of hexavalent chromium from aqueous solution through batch mode. The adsorbent was characterized by pH_{ZPC} and SEM study. The Cr(VI) adsorption was studied with the help of different process parameters, viz. initial concentration, contact time, adsorbent dose, pH, and temperature. Results revealed that Cr(VI) adsorption by human hair was highly pH sensitive. Maximum Cr(VI) was adsorbed from water at pH 1.0. Study of temperature effect on chromium adsorption confirmed the endothermic behaviour of the process. On the other hand, thermodynamic properties were also calculated and found that physisorption was dominant with activation energy of 0.385 kJ mol^{-1}. Kinetic study revealed that pseudo-second-order model was followed by the adsorption process. Adsorption equilibrium was analysed with Langmuir, Freundlich, and Dubinin–Radushkevich isotherm models. Results showed that the adsorption system followed both Langmuir and Freundlich isotherms with Langmuir adsorption capacity of 9.852 mg g^{-1}, which was compared with other adsorbents and observed that the performance of the present adsorbent is better than others. Finally, it can be concluded that human hair could be an alternative chief low-cost waste material for decontamination of heavy metals from an aqueous medium.

Keywords Human hair · Hexavalent chromium · Adsorption · Isotherm · Keratin · Kinetics

Introduction

Heavy metal-induced pollution has remarkably increased due to excessive discharges of heavy metals from various industries including metallurgical, steel manufacturing, fertilizer, including sponge iron, etc. (Rajapaksha et al. 2018; Gu et al. 2015). Among the different heavy metals, chromium has been specially attracted due to its both mutagenic and carcinogenic activities (Kazakis et al. 2017). Chromium exists in various oxidation states including Cr(III), which is mainly presence in the aqueous medium under acidic condition (pH 3.5) as Cr^{3+}, $Cr(OH)^{2+}$, $Cr(OH)_3$ and $Cr(OH)_4^-$. However, under oxidizing conditions Cr exists as Cr(VI)-oxoanion species such as $HCrO_4^-$ (pH 4–6) or CrO_4^{2-} (pH 8–10). The mobility of hexavalent chromium is higher than trivalent chromium in the aqueous medium (Brose and James 2013). The lower oxidation state of chromium can be precipitated after reduction of Cr(VI) (Hsu et al. 2009). In the soil enriched with Fe and Mn oxides, Cr(VI) can be adsorbed through sorption process under acidic condition (Zhang et al. 2017). Cr(VI) is a powerful oxidant which can oxidize the biomolecules and subsequently induce toxicity. The usual toxicity symptoms are necrosis, irritation of the gastrointestinal mucosa, etc. Hexavalent chromium is also known for its mutagenic and carcinogenic nature (Eleftheriou et al. 2015). Chromium can enter into the cell in the form of chromate (CrO_4^{2-}) through sulphate uptake channel. After insertion, Cr(VI) interacts with reducing biomolecules through formation of labile intermediates, and possibly this labile intermediate interacts with biomolecules including the

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bases of DNA (Feng et al. 2017). This may cause the main toxicity of hexavalent chromium.

Various technologies are available in the literature for removal of hexavalent chromium from the aqueous medium such as coagulation, precipitation, osmosis, and electrodialysis. However, all the above technologies have serious limitation due to high operational cost and generation of huge sludge. The above difficulties can be removed by using a cost-effective and high-performance technique known as adsorption (Liu et al. 2019).

Previous literature (Manjuladevi et al. 2018; Khulbe and Matsuura 2018; Mohan et al. 2005) highlighted a large number of adsorbents, which are mainly used for removal of Cr(VI) from aqueous solution. Various adsorbents such as biochar, activated carbon, and clay have been investigated (Rajapaksha et al. 2018; Nethaji and Sivasamy 2014; Khan et al. 2010). However, none of the above adsorbents showed good performance towards removal of Cr(VI) from aqueous medium. Therefore, there is a tremendous need of a suitable adsorbent, which should be easily available and greener one.

Hair is a complex tissue of animal body. It contains many chemicals which are mainly played as a main structural component. Human hair consists of proteins (65–95%), water, lipids, pigment, and trace elements (Ekop and Eddy 2010). Human hair is normally disposed as solid waste generated after hair cutting. These hairs can be used as an excellent adsorbent towards removal of heavy metals from aqueous solutions (Tan et al. 1985). Previous literature (Ingole et al. 2014) highlighted that human hair is an excellent adsorbent which removes oil from wastewater. However, there is not enough information regarding the efficacy of human hair to remove chromium from aqueous medium under varying different operating variables (pH, initial concentration, dose, contact time, temperature, etc.).

Materials and methods

Collection and preparation of adsorbent

Hair sample was collected from the different saloons of Burdwan town. After collection, the hair was washed with shampoo and dried in shade followed by cut into very small 1 mm in size by using electric cutter and stored in plastic airtight container for further use.

Characterization of adsorbent

(a) pH of zero-point charge

pH of zero-point charge was measured by following the method adopted by Bhaumik and Mondal (2015) through solid addition method. A series of 100-mL conical flask contains 50 mL of 0.1 M KNO_3 in each flask. The pH of solution was adjusted from 1.0 to 10.0 by using 0.1 (N) NaOH or 0.1 (N) HCl. Then, 0.2 g of hair was added to each flask and capped properly and sets into a shaker for 48 h and then filtered and measured the final pH of the solution.

(b) Scanning electron micrography (SEM)

The SEM (HITACHI, S-530, Scanning Electron Microscope and ELKO Engineering) study was performed with both before and after adsorption of hexavalent chromium in a fixed resolution to understand morphological change of human hair.

Preparation of Cr(VI) solution

A stock solution of $K_2Cr_2O_7$ was prepared by exactly weighing 0.2828 g of potassium dichromate in 250 mL double distilled water and finally diluted to 1 L which is equivalent to the strength of 100 mg L^{-1}. The pH of the experimental solution was adjusted by using 0.1 (N) HCl or 0.1 (N) NaOH solution.

Estimation of Cr(VI)

After performing adsorption study, the percentage of removal is calculated by using the following equation (Eq. 1):

$$\text{Removal}\,(\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \tag{1}$$

where C_0 and C_e are the initial and final concentrations of Cr(VI). The chromium concentration was measured by atomic adsorption spectrometry (LABINDIA, AS8000).

The adsorption capacity of human hair is calculated by the following equation (Eq. 2):

$$q_m\,(\text{mg/g}) = \frac{(C_0 - C_e) \times V}{M} \tag{2}$$

where V and M are the volume of the solution in litter and weight of solute taken in gram.

Regeneration study

Regeneration of the exhausted adsorbent can be done by 0.1 (M) NaOH solution. The saturated adsorbent was taken in a conical flask containing 0.1 M NaOH and placed in a magnetic starrer at 200 rpm for 30 min. After first cycle of filtration, the same thing has been done for the next 30 min.

Results and discussion

Adsorbents characterization

Analysis of pH_{ZPC}

From the pH_{ZPC} study, it is clear that all the three adsorbents showed different zero-point charge. The zero-point charge of human hair is 6.9363 (Fig. S1). The charges on the surface of adsorbents depend on these pH_{ZPC} values below and above the pH_{ZPC}; the surface of the adsorbents changes to positive and negative charges, respectively. However, at the pH_{ZPC} point, adsorbent surface has no charge at al. As per the literature, chromium species present in different oxy-anionic forms such as $CrO_4^{=}$, $HCrO_4^-$ or $Cr_2O_7^{=}$ at acidic pH and the present experiment suggests that all the three adsorbents showed maximum Cr(VI) removal at acidic pH 1 and it is below the pH_{ZPC} value. Therefore, it can be suggested that Cr(VI) adsorption is favourable when the surface of the adsorbent is positive. Almost similar observation was reported by the earlier researchers (Saha et al. 2013).

SEM study

Figure 1a exhibits the scanning electron micrography of human hair before adsorption of hexavalent chromium. From Fig. 1a, it is clear that human hair is layer-like structure without any deformation. However, after adsorption of Cr(VI), the surface of human hair is very rough in nature (Fig. 1b). This is perhaps due to effective interaction of Cr(VI) with protein structure (α-keratin) of hair (Zhang et al. 2018; Mahdavian 2014).

Effect of initial concentration

The removal of Cr(VI) from the aqueous solution by using human hair was studied by varying initial concentration of Cr(VI) from 10 to 30 mg L^{-1}, and the results are depicted in Fig. 3. From figure S2, it is clear that for a fixed amount of adsorbent (0.02 g), pH (1), contact time (30 min) and temperature (40 °C), percentage of Cr(VI) removal increases with increasing initial concentration from 10 to 30 mg L^{-1}. Therefore, the present results highlighted that the adsorbents support to bind the metal at a higher concentration. Results also revealed that at lower concentration (up to 15 mg L^{-1}) the Cr(VI) is not so impressive. However, at 20 mg L^{-1}, the percentage of Cr(VI) removal reached to 81.82%. Almost similar enhanced rate of adsorption of Zn^{2+} and Pb^{2+} by human hair was recorded with increasing concentration (Ekop and Eddy 2010). It was also noted that with increasing initial concentration from 10 to 30 mg L^{-1}, the adsorption capacity of the Cr(VI) in human hair increased from 11.175 to 62.365 mg g^{-1} (Fig. S2). This enhancement of adsorption with increasing initial concentration of Cr(VI) is attributed by the fact that the increase of driving force and also overcome the mass transfer resistance of metal ions between solid–liquid interphase (Khambhaty et al. 2009).

Effect of adsorbents dose

The influences of adsorbent dosage (0.01, 0.05, 0.1, 0.5 and 1.0 g human hair) on the adsorption of Cr(VI) ions at 40 °C are shown in figure S3. With the increase in biosorbent dose

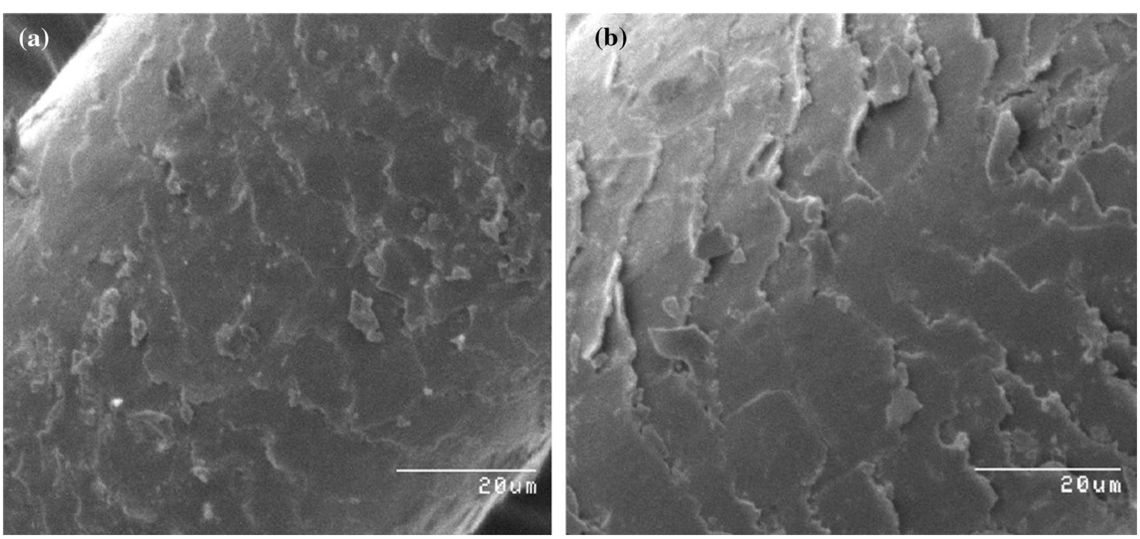

Fig. 1 **a** Human hair before Cr(VI) adsorption (×1500) and **b** human hair after Cr(VI) adsorption (×1500)

from 0.01 to 1.0 g/50 ml, the percentage of Cr(VI) removal increased from 57.43 to 98.77%. This is probably due to the availability of active binding sites where Cr(VI) ions adsorbed (Mohamed et al. 2017; Khambhaty et al. 2009). On the other hand, further increase in adsorbent dose did not cause any improvements towards adsorption of Cr(VI). This is perhaps due to the establishment of equilibrium between the ions bound to the sorbent and those remaining unabsorbed in the solution. The study results also revealed that after reaching an optimum dose, no further improvement towards removal of Cr(VI) was recorded. This can be attributed by the fact that the surface of newly added adsorbent will no longer active due to agglomeration or overlapping (Meng et al. 2017).

Effect of pH

pH is an important factor that strongly influenced the heavy metal adsorption (Gupta et al. 2001; Park et al. 2005a). Figure S4 illustrates the percentage of adsorption capacity with the variation of pH from 1 to 10. The maximum and minimum Cr(VI) removal was recorded at pH 1 and 10, respectively, for initial concentration of 30 mg L^{-1}. At low pH, the availability of H^+ ions increases near the adsorbent surface, and subsequently, the ionic species of Cr(VI) undergo electrostatic interaction with positive surface. But at higher pH, adsorbent surface changes to negative due to excessive OH^- ions and subsequently Cr(VI) biosorption got decreased (Dehghani et al. 2016). This observation is very much consistent with the earlier work where Cr(VI) was removed by various adsorbents (Bari and Abraham 2001; Park et al. 2005b; Tewari et al. 2005).

Effect of contact time

The adsorption of chromium ions on human hair was investigated as a function of contact time (10–60 min) at constant initial concentration (30 mg L^{-1}) and fixed temperature 55 °C. As figure S5 shows, the removal efficiency of Cr(VI) ions from aqueous solution increases rapidly up to 50 min. The high removal efficiency at the initial stages is probably due to the availability of large number of active adsorption sites which saturated with time. The maximum removal (99.96%) was reported at 50 min. However, after 50 min, the removal decreased to 98.5% at 60 min. Therefore, this has been considered as the optimum contact time for adsorption of Cr(VI). Almost similar efficiency towards removal (99.66%) of Cr(VI) by quaternized chitosan microspheres at 50 min was reported by Hua et al. (2016).

Effect of temperature

Previous literature (Wang et al. 2008; Khambhaty et al. 2009) suggested that temperature is an initial factor for removal of heavy metal from aqueous solution through adsorption mechanism. Therefore, the present experiment was conducted to examine the effect of temperature on removal of Cr(VI) from aqueous solution (Fig. S6). Figure S6 shows that with increasing temperature from 30 to 55 °C, the percentage of Cr(VI) removal increases from 85.86% to 97.8% . That means percentage of Cr(VI) removal favoured with temperature. Therefore, this observation clearly suggests that Cr(VI) interaction with the human hair is absolutely endothermic one. Almost similar temperature dependency of Pb^{2+} and Zn^{2+} removal by human hair was reported by Ekop and Eddy (2010).

Isotherm study for human hair

On the basis of experimental results, biosorption isotherm was used to understand the interaction pattern of Cr(VI) with human hair, at equilibrium. The Langmuir model was based on the assumption that the adsorbate only attached to the specific sites of the adsorbent surface, suggesting that the uptake of adsorbate is absolutely unilayer without interaction between adsorbate molecules. The following form of Langmuir equation (Eq. 3) is traditionally applied:

$$q_{\mathrm{e}} = \frac{q_{\mathrm{m}} b C_{\mathrm{e}}}{1 + b C_{\mathrm{e}}} \tag{3}$$

Equation (Eq. 4) can be conveniently transformed to the following linearized form:

$$\frac{C_{\mathrm{e}}}{q_{\mathrm{e}}} = \frac{1}{q_{\mathrm{m}} b} + \frac{C_{\mathrm{e}}}{q_{\mathrm{m}}} \tag{4}$$

where q_{m} is the maximum uptake (mg g^{-1}), q_{e} the uptake capacity at equilibrium (mg g^{-1}), C_{e} the equilibrium solution concentration (mg L^{-1}), b the Langmuir constant (L/mg). The output of Langmuir constant (q_{m} and b) and correlation coefficient (R^2) are presented in Table 1. As it can be seen from Table 1, the adsorption isotherm of Cr(VI) exhibited Langmuir behaviour, which indicates a monolayer adsorption. The adsorption of Cr(VI) by human hair is well-fitted with both Freundlich and Langmuir models; therefore, the Langmuir–Freundlich equation was also applied to the data sets. Basically the combined equation of Langmuir–Freundlich is known as Sips model. Table 1 also highlights that the Freundlich constant K_{F} and n are 13.62 (mg/g) $(\mathrm{L/mg})^{1/n}$ and 1.134, respectively. Therefore, these values indicate that the adsorption capacity and adsorption intensity both are favourable (Bhaumk et al. 2012). Similarly, the higher value of regression coefficient of Freundlich isotherm suggested the multi-layer adsorption of Cr(VI) onto human hair (Varvala et al. 2016). Almost similar Cr(VI) removal was recorded by Berihum (2017) using coffee husk carbon. The

Table 1 Isotherm data for Cr(VI) adsorption using human hair

Adsorption isotherm	Equations	Parameters	Values	R^2
Langmuir isotherm	$\frac{1}{q_e} = \frac{1}{q\text{max} K_L C_e} + \frac{1}{q\text{max}}$	q_{max} K_L	9.852 0.338	0.912
Freundlich isotherm	$\log q_e = \log K_F + \frac{1}{n} \log C_e$	K_F (mg/g) (L/mg)$^{1/n}$ n	13.62 1.134	0.992
D-R isotherm	$\ln q_e = \ln q\text{max} - \frac{1}{2E^2} * \left[RT \ln\left(1 + \frac{1}{C_e}\right)^2\right]$	q_{max} E (KJ mol^{-1})	18.857 0.3846	0.721

Table 2 Summary of parameters for various kinetic models by human hair

Kinetics model	Equations	Parameters	Values	R^2
Pseudo-first order	$\log\left(q_e - q_t\right) = \log q_e - \frac{K_1 t}{2.303}$	q_e (mg g^{-1}) K_1 (min^{-1})	4.015 21.23×10^2	0.9266
Pseudo-second order	$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e}$	q_e (mg g^{-1}) K_2 (g mg^{-1} min^{-1})	1.5081 2.3779	1.00
Intraparticle diffusion	$q_t = K_1 t^{1/2} + C$	K_1 (min$^{-1/2}$) C	58.672 82.041	0.7747

Langmuir–Freundlich equation (Eq. 5) can be expressed in the following way:

$$q_e = \frac{q_s b_s C_e^{1/n_s}}{1 + b_s C_e^{1/n_s}} \tag{5}$$

Equation (Eq. 6) can be easily linearized as:

$$\ln\left[\frac{q_s}{q_e} - 1\right] = -\ln b_s - \frac{1}{n_s} \ln C_e \tag{6}$$

Adsorption kinetics for human hair

The adsorption kinetics of Cr(VI) adsorption onto human hair was evaluated by pseudo-first-order, pseudo-second-order and intraparticle diffusion kinetic equations and the constant values depicted in Table 2. From Table 2, it is clear that the experimental data are well-fitted with pseudo-second-order kinetics model with very high correlation coefficient ($R^2 = 1.00$).

On the other hand, pseudo-first-order and intraparticle diffusion model are moderately fitted with the experimental data (Table 2). These results suggest that the adsorption onto the adsorbent at specific temperature was best presented by the pseudo-second-order equation, which is based on the assumption that the rate-limiting step may be the chemisorption (Aksu 2001). Song et al. (2016) reported the same observation for removal of toxic chromium by wheat straw and *Euatorium adenophorum.* Very recently, Campos et al. (2019) reported that core–shell bimagnetic nanoadsorbents can remove hexavalent chromium from aqueous solutions and it followed the pseudo-second-order model.

Table 3 Thermodynamics parameters for adsorption of Cr(VI) by human hair

Temperature (K)	$\Delta G°$ (KJ mol^{-1})	$\Delta H°$ (KJ mol^{-1})	$\Delta S°$ (KJ mol^{-1})
303	−420.216		
313	−725.165	$+3 \times 10^{-4}$	+67.0149
318	−726.246		
323	−2044.332		
328	−1151.517		

Thermodynamic study for human hair

The thermodynamic parameters for the obtained equilibrium data on temperature variation by the use of equations (Eqs. 7–9) were evaluated. The equilibrium constant K_c is calculated based of C_{Ae} and C_e values:

$$K_c = \frac{C_{Ae}}{C_e} \tag{7}$$

where C_{Ae} indicates adsorption in mg L^{-1} at equilibrium and C_e is the equilibrium concentration of the metal in mg L^{-1}. The respective values of other thermodynamic parameters such as $\Delta H°$ and $\Delta S°$ were obtained from the slope and interpret of the plot of $\log K_c$ against $1/T$ (Eq. 8), revealing that the values of free energy ($\Delta G°$) at different temperatures are obtained using Eq. 9.

$$\log K_c = \frac{\Delta S°}{2.303RT} - \frac{\Delta H°}{2.303RT} \tag{8}$$

where T is the temperature in Kelvin and R is the gas constant (KJ mol^{-1} K^{-1}).

The entire results for the thermodynamic parameters are presented in Table 3. From Table 3, it is clear that both $\Delta H°$ and $\Delta S°$ are positive; the positive value of $\Delta H°$ for Cr(VI) removal by human hair confirms that the adsorption process is endothermic in nature and positive $\Delta S°$ indicates the adsorption process is spontaneous. However, the spontaneity of chromium adsorption is also supported by the free energy value at different temperatures (Table 3), according to equation (Eq. 9).

$$\Delta G° = -RT \ln K_c \tag{9}$$

Results indicate that there is more negative value of ΔG at higher temperatures. That is the adsorption of Cr(VI) by human hair is favourable at higher temperatures. This is perhaps because the adsorption reaction is absolutely endothermic in nature. Almost similar results were reported by Ekop and Eddy (2010) for removal of Zn^{2+} and Pb^{2+} ions from aqueous solution by using human hair.

Regeneration study

In wastewater treatment process, the regeneration of exhausted adsorbent is an extremely important factor (Mohamed et al. 2017). The saturated adsorbent was washed several times with 0.1 (M) NaOH followed by distilled water and dried in shade. Then, activated adsorbent was again used for removal of Cr(VI) from aqueous solution. The present results highlighted that about 69% Cr(VI) can be removed by regenerated adsorbent. The current research (Su et al. 2019) demonstrated that exhausted activated carbon with hexavalent chromium can be regenerated by 1 M HCl.

Comparison with other published keratinous substances

Hexavalent chromium adsorption along with other heavy metals by various keratinous substances has been compared with respect to the results of adsorption capacity, and it is presented in Table 4. From Table 4, it is clear that wool showed greater than four times of Cr(VI) adsorption than human hair. The performance of animal hair exhibited higher adsorption capacity of heavy metals than human hair. Table value also revealed that human hair is better performance for lead removal than chromium. Hassan and Davies-McConchie (2012) demonstrated that wool fibre has lower adsorption capacity for arsenic(V).

Table 4 Comparison of heavy metal uptake by various keratinous substances

Adsorbents	Heavy metal	Adsorption capacity (mg g^{-1})	References
Human hair	Pb^{2+}	33.0	Mahdavian (2012)
Goat hair		24.0	
Ship hair		30.0	
Wool	Cr^{6+}	41.2	Dakiky et al. (2002)
Modified wool	Fe^{2+}	37.0	McNeil (2001)
	Zn^{2+}	11.0	
Wool fibres	As^{5+}	0.22	Hassan and Davies-McConchie (2012)
Human hair	Cr^{6+}	9.852	Present work

Conclusion

The present study results revealed that human hair was effective in the removal of Cr(VI) from aqueous medium. The efficacy of human hair was tested through batch study with variation of initial concentration, adsorbent dose, contact time pH, and temperature. The equilibrium of Cr(VI) adsorption was nicely fitted with Langmuir and Freundlich isotherms with adsorption capacity of 9.852 mg g^{-1}. The pseudo-second-order kinetics is well-fitted with regression coefficient 1.00. Thermodynamic of Cr(VI) adsorption by human hair is endothermic in nature. Finally, it can be concluded that human hair could be an inexpensive material for removal of hexavalent chromium from aqueous solution. Further study can be done towards removal of other metals from aqueous solutions.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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