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Effects of Chelating Surfactants on Competitive Adsorption of Lead and Zinc on Loess Soil

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ABSTRACT

The study of competitive adsorption of heavy metals on soil is important for heavy metals contaminated soil remediation. However, there have been few studies on the impact of desorption reagents on heavy metal adsorption in soil. Batch adsorption studies were used to investigate the competitive adsorption mechanism of two heavy metals, Pb and Zn, on a loess soil in the presence of a new chelating surfactant, sodium N-lauroyl ethylenediamine triacetate (LED3A). Results showed that competitive adsorption equilibria of Pb and Zn were reached at 3 and 10 h, respectively. The maximum equilibrium adsorption capacities were 19.55 and 18.35 g.kg^{-1}, respectively. LED3A affected the competitive adsorption kinetics of Pb and Zn by increasing the change in external mass transfer and reducing the change in internal mass transfer. LED3A reduced Pb and Zn adsorption capacities onto the soil through competitive chelation of the heavy metals. The heavy metal chelating ability of LED3A was higher for Zn than for Pb. When its concentration was larger than 5 g.L^{-1}, LED3A showed a significant effect on the competitive adsorption of Pb and Zn. In the competitive system, the effect of Pb concentration on the Zn adsorption capacity was greater than the effect of Zn concentration on the Pb adsorption capacity. LED3A weakened the effect of Pb concentration and enhanced the effect of Zn concentration. LED3A showed a significant potential for efficiently leaching remediation of Pb and Zn co-contaminated soil.

INTRODUCTION

Heavy metal pollution in soils has become a severe issue that is endangering human health and endangering the environment (Yuan et al. 2021). The adsorption/desorption characteristics of heavy metal ions at the soil-water interface are crucial for understanding the movement and fate of heavy metals in the soil. This has major implications for studying heavy metal bioavailability and transformation in the biological chain (Peng et al. 2018). It also provides evidence for the prevention and remediation of heavy metal contaminated soils (Qiu et al. 2010, Garridorodriguez et al. 2014). As a result, numerous research on the adsorption/desorption of heavy metals on soils have been published in recent years (Wang et al 2011, Shi et al. 2013, Peng et al. 2018, Wang et al. 2020). However, most adsorption/desorption research has concentrated on diverse soil types, heavy metal species, and adsorption/desorption process variables (Muhammad et al. 2018). Meanwhile, mechanistic research on soil and heavy metal systems, as well as contaminated soil and desorbent systems, have made it difficult to explain the competitive mechanism for heavy metals between soil and desorbent. To research the influence of the desorbent on heavy metal adsorption onto the soil, it is important to investigate the coexisting system of soil, heavy metals, and desorbent. The evident link between soil, heavy metals, and desorbents can offer the theoretical foundation needed for desorbent-based chemical leaching treatment of heavy metal-contaminated soil.

Desorbents, on the other hand, are mostly inorganic chemicals (such as acids, alkalis, and salts) and chelating agents, as previously stated. Inorganic chemicals, on the other hand, cause serious damage to soil structure and are not suited for contaminated soil repair (Kou et al. 2006, Guemiza et al. 2017). Although chelating agents can effectively desorb heavy metals from contaminated soils, it is difficult to degrade these agents in the soil environment, resulting in secondary pollution (Cao et al. 2013, Kim et al. 2016). Therefore, the development of an environmentally friendly desorbent that can efficiently desorb heavy metals from soil is critical.

Novel chelating surfactants (such as sodium *N*-alkyl ethylenediamine triacetate and sodium *N*-acyl ethylenediamine triacetate) possess both surface activity and chelating properties (Wang et al. 2004). These surfactants show superior performance due to being water-soluble, non-toxic to mammals and aquatic organisms, and biodegradable in the

natural environment without causing secondary pollution (Diao et al. 2016, Qiao et al. 2016). These surfactants have been extensively used in detergents, mineral flotation, phase transfer catalysis, emulsion polymerization, and other fields (Diao et al. 2016). However, few studies have reported the effect of chelating surfactants on the adsorption of heavy metals on the soil. To further improve the efficiency of LED3A in the remediation of soils contaminated with heavy metals, the relationship among chelating surfactant, soil, and heavy metals needs to be further clarified.

Loess soil from Northwest China and two heavy metals, Pb and Zn, were chosen as the researched soil and pollutants, respectively, in this study. The competitive adsorption of Pb and Zn onto the soil, as well as the influence of a new chelating surfactant, sodium N-lauroyl ethylenediamine triacetate (LED3A), on the competitive adsorption behavior of heavy metals, were studied in a series of studies. The findings support LED3A leaching treatment of heavy-metal contaminated soils on a theoretical level.

MATERIALS AND METHODS

Reagents

Sodium *N*-lauroyl ethylenediamine triacetate (LED3A, $C_{20}H_{35}N_2Na_3O_7$) with purity greater than 95.0% was purchased from Yilu Pharmaceutical Technology Co., Ltd. (Hangzhou, Zhejiang Province, China). Analytical-grade $Pb(NO_3)_2$ and $Zn(NO_3)_2$ were obtained from Damao Chemical industry Co., Ltd. (Tianjin, China). All other chemicals were of analytical grade purity.

Experimental Materials

Experimental Soil was collected from the surface soil (0-20 cm depth) of the mountain behind Lanzhou Jiaotong University (Lanzhou, Gansu Province, China). After removing gravel, branches, and leaves, the soil was naturally air-dried (20-30°C) and passed through a 0.3 mm sieve. The soil organic matter content was low (5.3 g.kg^{-1}). The soil pH was slightly alkaline (pH 8.11) and the carbonate content was 11.7 %.

Adsorption Experiments

Soil (0.05 g) was weighed into a series of conical flasks, to which 20 mL of the $Pb(NO_3)_2$, $Zn(NO_3)_2$, and LED3A solutions with different concentrations were added. Each conical flask also contained 0.01 mol.L^{-1} NaNO$_3$ to keep a constant ionic strength, and 0.1 mL of 100.0 mg.L^{-1} NaN$_3$ was added to inhibit microbial growth. The flasks were sealed immediately with a stopper and put into a shaker (TD6, Changsha pingfan Instrument and Meter Co., Ltd.,

China) at 150 rpm for a certain time at 25°C. At the end of the adsorption, the solution was centrifuged at 3500 rpm for 30 min. The supernatant was filtered by a 0.45 μm membrane for heavy metal analysis. The Pb and Zn concentrations were measured using a flame atomic absorption spectrophotometer (Spectrum AA110/220, Varian, Palo Alto, CA, USA). The adsorption capacities of Pb and Zn on the soil were calculated using Eq. 1:

$$q_t = \frac{(c_o - c_t) \times V}{m} \qquad \dots(1)$$

where q_t is the adsorption capacity of heavy metals at time t (g.kg^{-1}), c_o and c_t are the heavy metal concentrations at the beginning of adsorption and time t (g.L^{-1}), respectively, V is the volume of the heavy metal solution (L), and m is the mass of the soil sample (g).

A series of adsorption experiments using this method had been used to study the effect of LED3A on the competitive adsorption kinetics of Pb and Zn, the effect of LED3A concentration on competitive adsorption of Pb and Zn, the competitive adsorption of different concentrations of Pb and Zn and the effect of LED3A on its. All treatments were performed in triplicate, and the results were presented as average values.

RESULTS AND DISCUSSION

Competitive Adsorption Kinetics of Pb and Zn, and Effect of LED3A

The competitive adsorption kinetic results for Pb and Zn onto the loess soil were shown in Fig. 1(a) and the results of the curve fitted by the intraparticle diffusion model (b) are shown in Fig. 1(b). The adsorption capacities of Pb and Zn rapidly increased at an adsorption time within 1 h, increased slowly after 1 h, and then gradually leveled off as the adsorption time was further extended. During the rapid increase stage, more active sites were present on the surface of the soil particles along with a larger concentration gradient, resulting in a greater mass transfer force. As the adsorption time was extended, the Pb and Zn concentrations in solution and available active sites on the soil particle surfaces decreased gradually, causing a corresponding decrease in the mass transfer force. The equilibrium adsorption capacity of Pb (19.55 g.kg^{-1}) was reached at 3 h, while Zn (18.35 g.kg^{-1}) was reached at 10 h. These results indicated that Pb was preferentially adsorbed onto the soil in the competitive adsorption process and that the soil showed a higher ability to adsorb Pb than Zn.

Many studies on competitive adsorption have suggested that the inherent properties of heavy metal ions relate to their adsorption order, such as ion radius, electronegativity, first-order hydrolysis constant, hydrated ion radius,
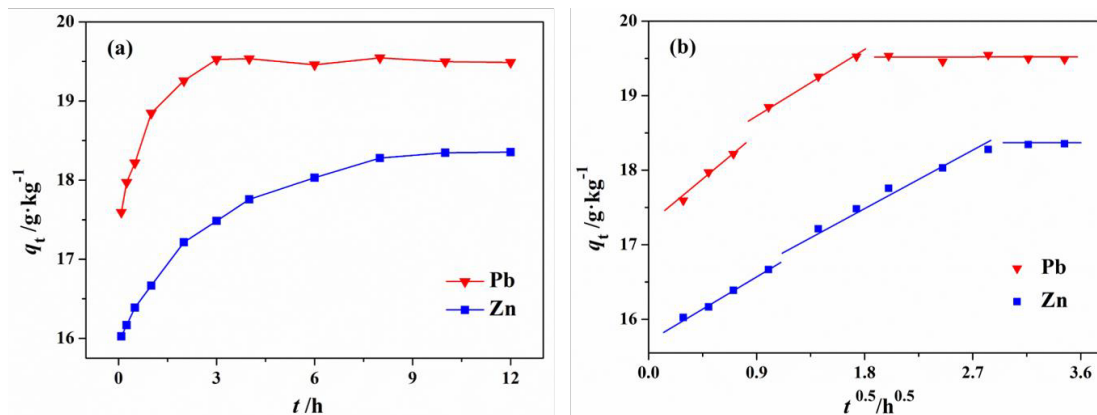
Fig. 1: Competitive adsorption kinetics of Pb and Zn (a), and the results of curve fitted by intraparticle diffusion model (b).

charge-to-radius ratio (Berrin 2012, Zakaria et al. 2017, Uddin 2017). The basic physical and chemical properties of Pb and Zn were shown in Table 1. In terms of ionic radius, the order of competitive adsorption is Zn > Pb (Uddin 2017). The electronegativities of Pb and Zn are important parameters determining the order of adsorption. Oxygen atoms on the soil surface or within soil particles bind to Pb ions with higher electronegativity, forming stronger covalent bonds and facilitating Pb adsorption on the soil (Zakaria et al. 2017). Furthermore, the specific adsorption of heavy metal ions onto the soil is related to their hydrolysis ability (Berrin 2012). An increase in the negative logarithm of the first-order hydrolysis constant, pK_1, can lead to the reduction of the adsorption affinity of heavy metals. Therefore, the preferential adsorption order of heavy metal ions was inversely proportional to the first-order hydrolysis constant, Pb > Zn. Moreover, metal ions with a smaller hydrate ion radius are more easily adsorbed onto soils by ion exchange (Wang et al. 2015). As Pb had a smaller hydrate ion radius than Zn, the former showed stronger adsorption ability. In an adsorption process involving electrostatic attraction alone, a larger charge-to-radius ratio results in greater bond energy (Wang et al. 2015). Therefore, the preferential adsorption order was Zn > Pb. In the present study, the preferential adsorption order of Pb and Zn onto the loess soil was consistent with their electronegativity, hydrolysis constant, and hydrate ion radius.

The effect of LED3A on the competitive adsorption kinetics of Pb and Zn is shown in Fig. 2(a) and the results of the curve fitted by the intraparticle diffusion model are shown in Fig. 2(b). In the presence of 5 g.L^{-1} LED3A, the competitive adsorption kinetic process of Zn was also divided into rapid, slow, and equilibrium stages. However, only two stages (rapid and equilibrium) were recognized in the competitive adsorption of Pb. The equilibrium adsorption times of Pb and Zn were 1 and 6 h, respectively, which were less by 2

and 4 h, respectively, as compared to the equilibrium adsorption in the absence of LED3A. The equilibrium adsorption capacities of Pb and Zn on the soil were 16.99 and 13.38 g.kg^{-1}, respectively, indicating that Pb had a higher adsorption ability than Zn. The adsorption capacities of Pb and Zn in the presence of LED3A were reduced by 2.56 and 4.97 g.kg^{-1}, respectively, compared with those without LED3A addition. This was attributed to the coexistence of competitive adsorption of Pb and Zn onto the soil, competitive chelation of Pb and Zn by LED3A, and soil competing with LED3A for Pb and Zn in the LED3A, soil, Pb, and Zn system. LED3A has a high ability to chelate Pb and Zn, resulting in attenuated adsorption of heavy metal ions on the soil surface and within soil particles, which shortened the time to reach equilibrium. The reduction in Zn adsorption capacity was greater than that in Pb adsorption capacity, indicating that LED3A had a stronger ability to competitively chelate Zn than Pb. A similar conclusion was made by Qiao et al. (2016) who studied the LED3A as an effective washing reagent for lead (Pb)- and zinc (Zn)-contaminated soil.

Pseudo-first-order and pseudo-second-order equations were used to calculate the date for competitive adsorption kinetics, and the calculation formula was as follows (Manhirethan et al. 2018):

$$\ln(q_e - q_t) = \ln q_e - k_1 t \qquad \ldots(2)$$

Table 1: Basic physical and chemical properties of Pb and Zn ions.

Basic properties	Pb	Zn
Ion radius/pm	119	74
Electronegativity	1.8	1.65
First-order hydrolysis constant/pK_1	7.8	8.96
Hydrated ion radius/nm	0.401	0.430
Charge-to-radius ratio	1.68	2.10

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad \qquad …(3)$$

where t is the adsorption time (h), q_e and q_t are the heavy metal adsorption capacities at equilibrium and time t, respectively (g.kg^{-1}), k_1 is the first-order rate constant (h^{-1}), and k_2 is the second-order rate constant (kg.g^{-1}.h^{-1}).

The fitting results were summarized in Table 2. The pseudo-second-order kinetic model had a markedly higher correlation coefficient (R^2) than the pseudo-first-order kinetic model when fitting the competitive adsorption process of Pb and Zn. The maximum adsorption capacity (q_{cal}) calculated by fitting to the pseudo-second-order kinetic model was much close to the measured value (q_{exp}). Therefore, the competitive adsorption process of Pb and Zn onto the soil was accurately described by the adsorption mechanisms included in the pseudo-second-order kinetics, such as external liquid film diffusion, surface adsorption, and intraparticle diffusion. Moreover, k_2 was higher for Pb than for Zn, indicating preferential adsorption of Pb onto the soil. In the presence of LED3A, the competitive adsorption kinetic process of Pb and Zn was described by the pseudo-second-order kinetic model in terms of either the R^2 of fitting or the closeness of q_{cal} and q_{exp}. LED3A showed an excellent ability to chelate Pb and Zn in the LED3A, soil, Pb, and Zn system. Therefore, both

q_{cal} and q_{exp} were lower than those without LED3A addition. In the presence of LED3A, the k_2 values of Pb and Zn were 74.23 and 1.72 kg.g^{-1}.h^{-1}, respectively, which were markedly higher than those without LED3A addition. This showed that the presence of LED3A affected the competitive adsorption rate of Pb and Zn onto the soil.

To explain the mechanism of competitive adsorption between Pb and Zn in the absence and presence of LED3A, the data in Fig. 1(a) and 2(a) was fitted using the intraparticle diffusion equation (Eq. 4) (Qiao et al. 2015):

$$q_t = k_d t^{0.5} + a \qquad \qquad …(4)$$

where q_t is the heavy metal adsorption capacity at time t (g·kg^{-1}), k_d is the intraparticle diffusion rate constant (kg.g^{-1}.h$^{-0.5}$), and a is the intercept which is the boundary layer and gives an idea about the thickness of the boundary layer, i.e. the larger the intercept, the greater the boundary layer effect.

The fitting results were shown in Fig. 1 (b) and 2 (b). The competitive adsorption of Pb and Zn onto the soil was a continuous multistage diffusion process in both the soil-Pb-Zn and soil-LED3A-Pb-Zn systems, which agreed with the conclusions from the competitive adsorption kinetic curves. The first linear stage reflected a rapid boundary layer diffusion process, the second linear stage reflected a slow pore diffusion process, and the third one reflected a final

Table 2: Parameters of pseudo-first-order and pseudo-second-order kinetic equations.

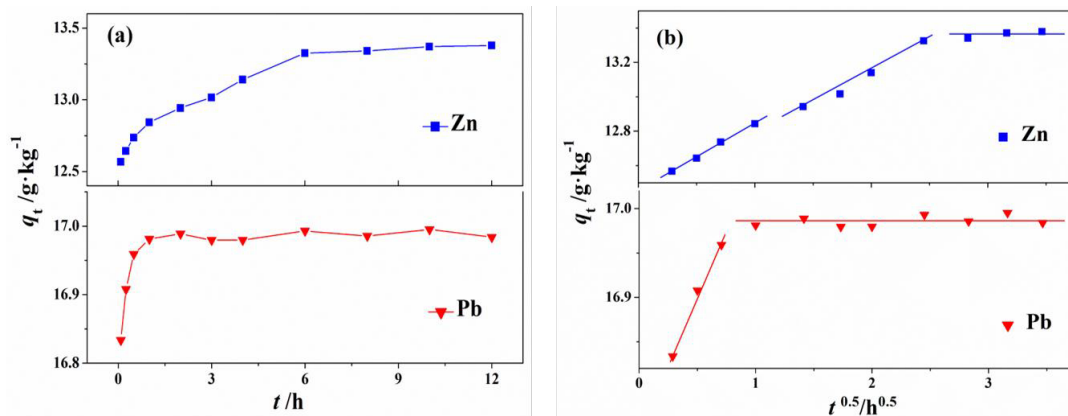| Adsorption type	q_{exp}/g.kg^{-1}	Pseudo-first-order kinetic equation			Pseudo-second-order kinetic equation		
		k_1/h^{-1}	q_{cal}/g.kg^{-1}	R^2	k_2/kg.g^{-1}.h^{-1}	q_{cal}/g.kg^{-1}	R^2
Pb	19.55	1.32	2.70	0.9834	2.49	19.54	0.9999
Zn	18.35	0.55	3.33	0.9552	0.58	18.45	0.9999
LED3A+Pb	16.99	1.33	0.12	0.9236	74.23	16.98	1.0000
LED3A+Zn	13.38	0.50	2.22	0.9796	1.72	13.41	0.9999

Fig. 2: Competitive adsorption kinetics of Pb and Zn in the presence of LED3A (a), and the results of curve fitted by intraparticle diffusion model (b).

competitive equilibrium process. The fitting parameters were listed in Table 3. The R^2 values of linear fitting at each stage were greater than 0.99. The fitted apparent rate constant, k_{d1}, was always greater than k_{d2} for the competitive adsorption of Pb and Zn in the absence or presence of LED3A, indicating that intraparticle pore diffusion was the controlling step for the competitive adsorption rate of the heavy metals. However, the linear plot of q_t vs. $t^{0.5}$ did not pass the origin throughout the competitive adsorption process, indicating that intraparticle diffusion was not the only mechanism controlling the competitive adsorption rate. The competitive adsorption of Pb changed from a three-stage to two-stage process after LED3A addition, suggesting that LED3A reduced the porosity diffusion of Pb during competitive adsorption due to competing with soil for Pb. Moreover, the a_1 and a_2 values in the fitting results were reduced to different degrees after the LED3A addition, indicating that LED3A increased the change in external mass transfer and decreased the change in internal mass transfer, which shortened the equilibrium time of the competitive adsorption process of Pb and Zn. This occurred because LED3A has good solubility and forms soluble chelates LED3A-Pb and LED3A-Zn after competitive chelation with Pb and Zn, which reduces the competitive adsorption of Pb and Zn on the surface of soil particles and within the pores.

Effect of LED3A Concentration on Competitive Adsorption of Pb and Zn

The molecular structure of LED3A comprises three carboxyl groups and two tertiary amino groups. LED3A can form stable and soluble chelates with Pb and Zn ions by coordinating through oxygen and nitrogen atoms. Furthermore, introducing a fatty acyl group into the molecular structure means that micellization can enhance greatly affect its selectivity for chelating heavy metals. The chelation of heavy metals (M, namely Pb and Zn) by LED3A is illustrated as follows:
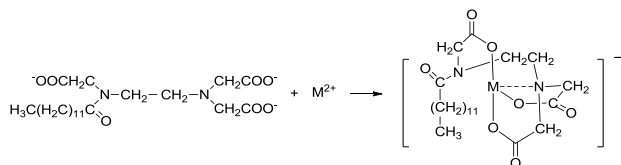
Adding LED3A to the soil and heavy metal solution,

LED3A could chelate M^{2+} to keep them in the solution system. Soil can make M^{2+} leave the solution system through adsorption. Therefore, LED3A and the soil compete for the heavy metal ions in the system, which affected the adsorption of heavy metal ions onto the soil. The effect of LED3A on the adsorption of Pb and Zn onto soil can be described by q_e:

$$\triangle q_e = q_e - q_L \qquad \text{...(5)}$$

where q_e is the equilibrium adsorption capacity of soil for Pb or Zn in the absence of LED3A (g.kg^{-1}) and q_L is the equilibrium adsorption capacity of soil for Pb or Zn in the presence of LED3A (g.kg^{-1}).

The effect of the LED3A solution concentration on the competitive adsorption of Pb and Zn onto soil is shown in Fig. 3. As the concentration of LED3A increased from 0.1 to 0.7 g.L^{-1}, the adsorption capacities of Pb and Zn decreased. In contrast, Δq_e increased from 1.21 to 1.47 g.kg^{-1} and 1.64 to 4.74 g.kg^{-1} for Pb and Zn, respectively. When the LED3A concentration was increased to 5 g.kg^{-1}, the adsorption capacities of Pb and Zn continued to decrease and Δq_e reached maximum values of 3.37 and 8.90 g.kg^{-1}, respectively. When the LED3A concentration was greater than 5 g.kg^{-1}, the adsorption capacities of Pb and Zn reached a competitive equilibrium state, because the ability of LED3A to chelate Pb and Zn was related to the LED3A concentration (Qiao et al. 2016). The critical micelle concentration (CMC) of LED3A is 0.71 g.L^{-1} (Qiao et al. 2016). In the LED3A, soil, Pb, and Zn system, when the LED3A concentration was lower than the CMC, LED3A mainly existed as monomers in the aqueous solution, which were less competitive for Pb and Zn adsorption than the soil. When the LED3A concentration exceeded the CMC, LED3A mainly existed as micelles in the aqueous solution, which showed a significantly increased ability to chelate Pb and Zn. As a result, the adsorption capacities of Pb and Zn on soil were significantly decreased, while the Δq_e was significantly increased. Comparing Δq_e for the two heavy metals, it was found that Δq_e for Pb was markedly lower than that of Zn. This was due to the different competitive abilities of soil and LED3A micelles for Pb and Zn. When the LED3A concentration is greater than 5 g.L^{-1}, LED3A exhibited the strongest effect on the competitive adsorption of Pb and Zn onto the soil.

Table 3: Kinetic parameters of intraparticle diffusion model.

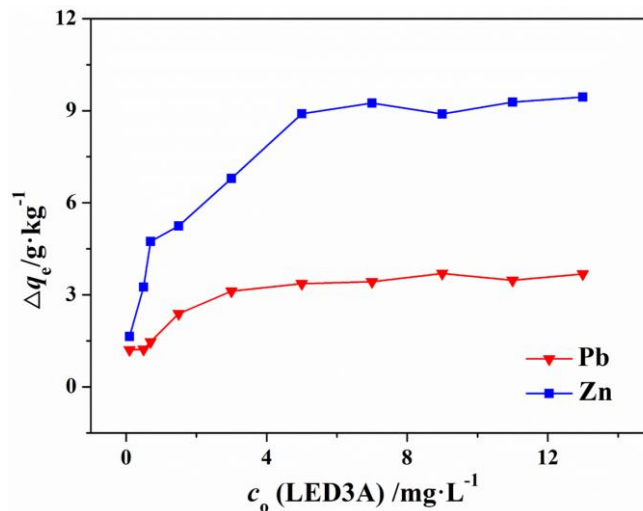| Adsorption type		k_{d1}/g.kg^{-1} h$^{-0.5}$	a_1	R^2	k_{d2}/g.kg^{-1}.h$^{-0.5}$	a_2	R^2
Pb		1.49	17.19	0.9933	0.93	17.92	0.9991
Zn		0.92	15.74	0.9968	0.75	16.19	0.9952
LED3A	Pb	0.30	16.75	0.9950	-	-	-
	Zn	0.39	12.45	0.9991	0.37	12.38	0.9925

Fig. 3: Effect of LED3A concentration on competitive adsorption of Pb and Zn onto loess soil.

Competitive adsorption of different concentrations of Pb and Zn, and the effect of LED3A

The competitive adsorption capacity of heavy metal ions on adsorbents is related to the ion concentration. (Dou et al. 2020). The Pb (Zn) concentration was set to 0.05 g.L^{-1} and the Zn (Pb) concentration was changed to determine the effect of Zn (Pb) concentration on Pb (Zn) adsorption onto the soil. As shown in Fig. 4, the adsorption capacity of Pb onto the soil was generally not affected by the change in Zn concentration, remaining stable at 19.44 g.kg^{-1}. Nevertheless, the adsorption capacity of Zn decreased with increasing Pb concentration and generally stabilized at 17.6 g.kg^{-1} with a Pb concentration of 0.25 g.L^{-1}, demonstrating obvious competitive adsorption.

According to the results by Abdelfattah and Wada (1981) and Wilcke et al. (1998), the soil particle surface is markedly heterogeneous, in which all adsorption sites can be divided into groups with high binding energy and low binding energy. Adsorption firstly occurs at high-energy adsorption sites during the initial stage, whereas low energy adsorption sites start to adsorb heavy metal ions as the heavy metal concentration is further increased (Dong et al. 2010, Gao et al. 2017). Generally, heavy metal ions adsorbed by electrostatic interaction have low binding energies, and heavy metal ions adsorbed specifically have high binding energies. The above analysis showed that as the Zn concentration increased, it was difficult for Zn ions to compete with Pb ions to be adsorbed at high energy and low energy adsorption sites. However, when the Pb concentration increased, Pb ions competed with Zn ions to be adsorbed at low-energy adsorption sites and some Zn ions adsorbed at high energy adsorption sites. Therefore, in the competitive system, the soil showed a higher ability for specific adsorption of Pb relative to that of Zn.
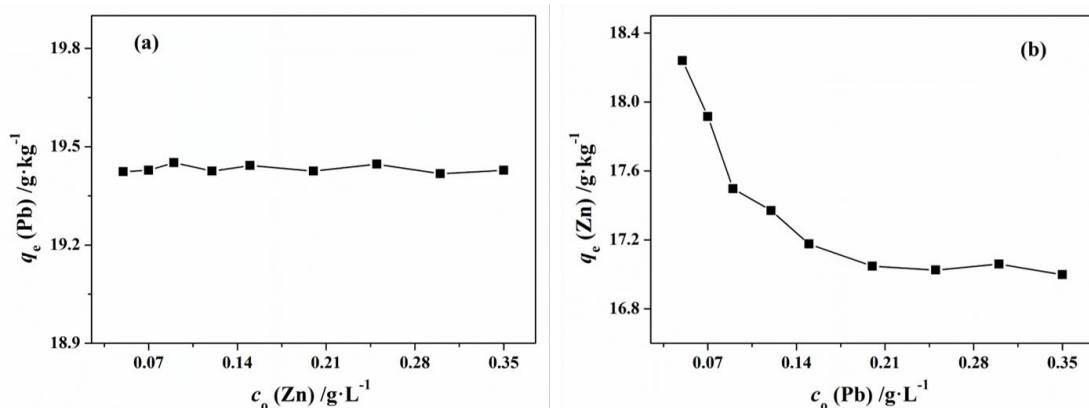

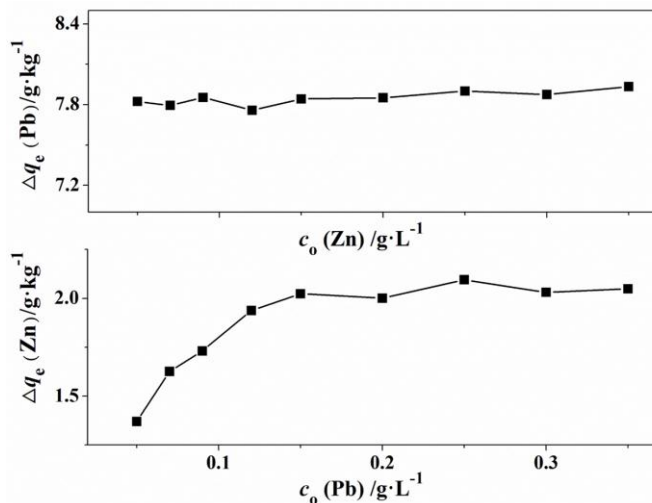Fig. 4: Competitive adsorption of different concentrations of Pb and Zn onto loess soil.

Fig. 5: Competitive adsorption of different concentrations of Pb and Zn in the presence of LED3A.

The effect of Pb concentration on Zn adsorption capacity was greater than that of Zn concentration on Pb adsorption capacity, and the increase in Pb concentration inhibited Zn adsorption onto the soil.

The effect of LED3A on the competitive adsorption of Pb and Zn at different concentrations is described by Eq. 5. As shown in Fig. 5, the Δq_e values representing the LED3A induced change in Pb and Zn adsorption capacities were greater than zero, indicating different extents of reduction in Pb and Zn adsorption capacities caused by LED3A. In general, the Δq_e of Pb did not change with increasing Zn concentration and remained 7.85 g.kg^{-1}, which suggested that the Pb adsorption capacity was not affected by the Zn concentration, mainly due to competition between LED3A and soil for Pb. In contrast, the Δq_e of Zn increased with increasing Pb concentration, with the greater reduction in Zn adsorption capacity mainly due to competition between LED3A and soil for Zn, and Pb competing for Zn adsorption sites. When the Pb concentration was >0.15 g.L^{-1}, the Δq_e of Zn generally remained 2.04 g.kg^{-1}. The Pb concentration for the Zn adsorption equilibrium was substantially lower in the presence of LED3A than in the absence of LED3A, implying that the competition for Zn adsorption sites between LED3A and soil was stronger than that of Pb. In the competitive LED3A, soil, Pb and Zn system, the Δqe of Pb was greater than that of Zn, implying that LED3A increased the influence of Zn concentration on Pb adsorption capacity while decreasing the effect of Pb concentration on Zn adsorption capacity.

CONCLUSION

The preferential adsorption order for Pb and Zn was connected to their electronegativity, hydrolysis constant, and hydrate ion radius, with Loess having a larger competitive adsorption capability for Pb than Zn. LED3A shortened the equilibrium time of competitive adsorption and reduced Pb and Zn adsorption capacities on the soil by 2.56 and 4.97 g.kg^{-1}, respectively. The competitive adsorption kinetic processes could be best described by the pseudo-second-order kinetic equation. For Pb, Zn competitive adsorption, intra-particle diffusion was the rate control step, but it was not the only rate control mechanism. As LED3A concentrations >5 g.L^{-1}, LED3A showed the greatest effect on the competitive adsorption of Pb and Zn. LED3A decreased the effect of Pb concentration on Zn adsorption capacity while increasing the effect of Zn concentration on Pb adsorption capacity. LED3A showed significant potential for chelating a lot of heavy metals from contaminated soil.

REFERENCES

Abdelfattah, A. and Wada, K. 1981. Adsorption of lead, copper, zinc, cobalt, and cadmium by soils that differ in cation-exchange materials. Europ. J. Soil Sci., 32: 271-283.

Berrin, T. 2012. Significance of thermodynamic and physical characteristics on permeation of ions during membrane separation: Hydrated radius, hydration free energy, and viscous effects. Sep. Purif. Technol., 86: 119-126.

Cao, M., Hu, Y., Sun, Q., Wang, L., Chen, J. and Lu, X. 2013. Enhanced desorption of PCB and trace metal elements (Pb and Cu) from contaminated soils by saponin and EDDS mixed solution. Environ. Pollut., 174: 93-99.

Diao, J.R., Zhao, B.W., Ma, F.F., Wang, X. and Ding, W.J. 2016. Solubility enhancement of phenanthrene using novel chelating surfactant. Chem. Papers, 70: 375-383.

Dong, X., Li, C., Li, J., Wang, J., Liu, S. and Ye, B. 2010. A novel approach for soil contamination assessment from heavy metal pollution: A linkage between discharge and adsorption. J. Hazard. Mater., 175: 1022-1030.

Dou, W., Liu, J. and Li, M. 2020. Competitive adsorption of Cu^{2+} in Cu^{2+}, $Co^{2+,}$ and Ni^{2+} mixed multi-metal solution onto graphene oxide (GO)-based hybrid membranes. J. Mol. Liq., 12: 114516.

Gao, M., Lv, M., Gong, X., Song, W., Qi, Y. and Li, Z. 2017. Effect of temperature and aging on the adsorption and desorption of menadione in soil. Environ. Prog. Sustain. Energy, 36: 1997-1004.

Garridorodriguez, B., Cutillasbarreiro, L., Fernándezcalviño, D., Ariasestévez, M., Fernándezsanjurjo, M.J., Alvarezrodríguez, E. and Núñezdelgado, A. 2014. Competitive adsorption and transport of Cd, Cu, Ni and Zn in a mine soil amended with mussel shell. Chemosphere, 07: 379-385.

Guemiza, K., Coudert, L., Metahni, S., Mercier, G., Besner, S. and Blais, J.F. 2017. Treatment technologies used for the removal of As, Cr, Cu, PCP and/or PCDD/F from contaminated soil: A review. J. Hazard. Mater., 333: 194-214.

Kim, E.J., Jeon, E.K. and Baek, K. 2016. Role of reducing agent in the extraction of arsenic and heavy metals from soils by use of EDTA. Chemosphere, 152: 274-283.

Kou, S., Lai, M.S. and Lin, C.W. 2006. Influence of solution acidity and $CaCl_2$ concentration on the removal of heavy metals from metal-contaminated rice soils. Environ. Pollut., 44: 918-925.

Manirethan, V., Raval, K., Rajan, R., Thaira, H. and Balakrishnan, R.M. 2018. Kinetic and thermodynamic studies on the adsorption of heavy metals from aqueous solution by melanin nano pigment obtained from marine source: Pseudomonas stutzeri. J. Environ. Manag., 214: 315-324.

Muhammad, A.K., Sardar, K., Ding, X., Anwarzeb, K. and Mehboob, A. 2018. The effects of biochar and rice husk on adsorption and desorption of cadmium onto soils with different water conditions (upland and saturated). Chemosphere, 193: 1120-1126.

Peng, L., Liu, P., Feng, X., Wang, Z., Cheng, T., Liang, Y., Lin, Z. and Shi, Z. 2018. Kinetics of heavy metal adsorption and desorption in soil: developing a unified model based on chemical speciation. Geochim. Cosmochim. Acta, 224: 282-300.

Qiao, H.T., Liu, Y., Dong, Y.H., Li, S., Wang, P. and Jin, T. 2015. Adsorption of sulfamonomethoxine antibiotics to cucurbit[6]uril polymer: kinetics and thermodynamic studies. Desalin. Water Treat., 54: 113-121.

Qiao, H.T., Zhao, B.W., Diao, J.R., Huang, L.P., Zhong, J.K. and Ma, F.F. 2016. Removal of lead and zinc from contaminated soil by a novel chelating surfactant. Clean Soil Air Water, 44: 1191-1197.

Qiu, R.L., Zou, Z.L., Zhao, Z.H., Zhang, W.H., Zhang, T., Dong, H.Y. and Wei, X.G. 2010. Removal of trace and major metals by soil washing with Na_2EDTA and oxalate. J. Soils Sed., 10: 45-53.

Shi, Z., Toro, D.M.D., Allen, H.E. and Sparks, D.L. 2013. A general model for the kinetics of heavy metal adsorption and desorption on soils. Environ. Sci. Technol., 47: 3761-3767.

Uddin, M.K. 2017. A review on the adsorption of heavy metals by clay minerals, with a special focus on the past decade. Chem. Eng. J., 308: 438-462.

Wang, P., Peng, H., Liu, J., Zhu, Z., Bi, X., Yu, Q. and Zhang, J. 2020. Effects of exogenous dissolved organic matter on the adsorption-desorption behaviors and bioavailabilities of Cd and Hg in a plant-soil system. Sci. Total Environ., 728: 138252.

Wang, X., Zhao, J., Yao, X. and Chen, W. 2004. Synthesis and properties of N-hexadecyl ethylenediamine triacetic acid. J. Coll. Interface Sci., 279: 548-551.

Wang, Y., Tang, X.W., Wang, H.Y. and Liu, W. 2011. Sorption and desorption behaviors of heavy metal Mn(II) on loess soil. Chinese J. Geotech. Eng., 33(Supp1): 369-373.

Wang, Z., Huang, G., An, C., Chen, L. and Liu, J. 2015. Competitive adsorption characteristics of water-quenched blast furnace slag (WBFS) towards Cu^{2+}, Cd^{2+} and Zn^{2+}. Chem. Ind. Eng. Prog., 34: 4071-4078.

Wilcke, W., Müller, S., Kanchanakool, N. and Zech, W. 1998. Urban soil contamination in Bangkok: Heavy metal and aluminum partitioning in topsoils. Geoderma, 86: 211-228.

Yuan, X., Xue, N. and Han, Z. 2021. A meta-analysis of heavy metals pollution in farmland and urban soils in China over the past 20 years. J. Environ. Sci., 101: 217-226.

Zakaria, M., Sofiane, M., Salima, S.B., Geraldin, A., Abdelhamid, E. and Derdour, A. 2017. Triazole containing magnetic core-silica shell nanoparticles for Pb^{2+}, Cu^{2+} and Zn^{2+} removal. Arab. J. Chem., 10: 1039-1051.