Study on adsorption of lead and cadmium by non-burning ceramic granules from waterworks residual sludge

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Abstract. In this experiment, the unburned ceramiste water treatment residuals (UCWTR) was prepared by the burning-free method to study its adsorption performance on Pb and Cd. Adsorption experiments show that the quality of ceramsite improved with the height of the filler layer in the reaction column, and its penetration and depletion time are prolonged. On the contrary, the driving force of mass transfer increases with the initial concentration of water in the reaction column, and the penetration and exhaustion time is shortened. The dynamic adsorption process of Pb and Cd by UCWTR is more consistent with the Thomas model and Yoon-Nelson model, and the correlation coefficient R2 is greater than 0.9. Under the condition that the initial flow rate v is 10 mL/min, the height of the packing layer h is 10 cm, and the initial concentration C0 is 150 mg/g, the fitted dynamic saturated adsorption capacities of UCWTR for lead and cadmium are 9.22 mg/g, 11.52 mg/g, the static adsorption capacity of lead and cadmium are 1.48 times and 155 times respectively. kT decreases as the height of the filler layer and the initial concentration increase; kYN decreases as the height of the filler layer increases, and increases as the initial concentration increases; the time t required to penetrate 50% of the adsorbent increases with the height of the filler layer and decreases with the increase of the initial concentration, which indicated that the solution did not reach full contact with UCWTR during the dynamic adsorption process.

1. Introduction

Lead and cadmium are the first type of pollutants that are strictly controlled in the China Comprehensive Wastewater Discharge Standard (GB8978-1996) and the Urban Wastewater Treatment Plant Pollutant Discharge Standard (GB18918-2002) which mainly comes from mining, metal smelting, and electroplating. At the same time, some of the lead and cadmium discharged in daily life of residents will also enter the sludge of sewage treatment plants and leachate from municipal waste landfills through the municipal pipe network (Pan, 2014). Lead and cadmium in the tail gas of iron and steel, metallurgy and other industries can cause damage to the environment through atmospheric deposition and rainfall migration, and even threaten very far areas (He, 2013). The composition of wastewater containing lead and cadmium is complex and highly toxic, and it is currently a hot and difficult emphasis in wastewater treatment (Karlsson K, 2010).
At present, the treatment methods for lead-cadmium wastewater can be divided into chemical methods (mainly electrolytic methods (Zhang, 2013)), physical and chemical methods (membrane separation methods (Cai, 2019), adsorption methods (Wei, 2018) and plant methods (Lin, 2017). Among them, the adsorption method has the advantages of short reaction time, common usage, and certain selectivity to target pollutants. A large number of scholars have conducted research on the development of adsorption materials and their adsorption characteristics, and have achieved certain results. The adsorption and purification treatment of lead and cadmium pollution has made a certain contribution, but the dynamic reproducibility of the adsorption material is one of the main bottlenecks restricting its practical application.

In this essay, the self-made UCWTR is used as the adsorption material, and the lead and cadmium simulated wastewater are used as the objects. Meanwhile, the UCWTR is used as the adsorption column packing to carry out the UCWTR dynamic adsorption experiment of lead and cadmium which research content includes investigating the influence of the initial concentration, the height of the packing layer of the adsorption column on the adsorption penetration curve; using different dynamic adsorption models (Thomas model and Yoon-Nelson model) to fit the experimental data and analyze the penetration rule.

2. Methods and material

2.1. Preparation of UCWTR
Mix the residue of the water supply plant with a mass ratio of about 80% (through a 0.15 mm sieve), about 10% of cement, 10% of the stimulant (quick lime) and binder (sodium silicate within 2%) according to a certain ratio. Stir well to make raw ceramsite raw material, use spheroidizer to make ceramsite raw material with a diameter of about 5 mm, age at room temperature for 2 h, and then form by high-temperature steam curing (Liu, 2019).

2.2. Dynamic adsorption experiment
The UCWTR dynamic adsorption experiment device uses a plexiglass column as the adsorption column, with an inner diameter of 4.0 cm and an effective height of 24.0 cm. In order to prevent the outflow of ceramsite from the water inlet or water outlet, the top and bottom of the adsorption column are covered with a layer of quartz sand. This experiment was conducted at room temperature (20–25 °C). In order to make the water uniform, the lead and cadmium wastewater passed through the adsorption column through the bottom in and out, and the permeate pump was used to control the water flow. The proposed flow rate was 10 mL/min. Set three gradients of packing layer height and three gradients of initial concentration of influent to study the dynamic adsorption performance of UCWTR to lead and cadmium respectively. Samples were taken from the outlet of the adsorption column every 20 minutes to determine the content of lead and cadmium in the water.

Taking the running time $t$ as the x-axis and the ratio of the effluent lead and cadmium concentration $C_t$ to the initial lead and cadmium concentration $C_0$ as the y-axis, the penetration curve is drawn. In this experiment, the concentration of lead and cadmium in the effluent reaches 10% of the lead and cadmium in the influent as the penetration point (Shahbazi A, 2013), that is, $C_t/C_0 = 0.1$. Suppose the concentration of lead and cadmium in the effluent reaches 90% of the concentration of lead and cadmium in the influent as the saturation point. At this time, the adsorbent has reached dynamic adsorption saturation (H.C., 1944), that is, $C_t/C_0 = 0.9$.

2.3. Dynamic adsorption penetration curve model
In this study, the Thomas model and Yoon-Nelson model were fitted to the dynamic adsorption data, and the corresponding parameters were obtained.

2.3.1. Thomas model. The Thomas dynamic fitting model (Hasan S H, 2010) is based on the Langmuir kinetic equation and is assumed to be derived without axial diffusion. It can describe the adsorption
process of a fixed-bed adsorption column and estimate the idealized model of equilibrium adsorption amount and adsorption rate constant. The expression equation is:

$$\frac{C_t}{C_0} = \frac{1}{1 + \exp\left[\frac{k_T v q_e m - C_0 t}{m}ight]}$$

In this equation:
- $C_1$ - concentration of outlet water, mg/L
- $C_2$ - concentration of inlet water, mg/L
- $k_T$ - thomas rate constant, mL/(mg · min$^{-1}$)
- $q_e$ - dynamic saturation adsorption capacity of ceramsite, mg/g
- $m$ - quality of UCWTR in reaction column, g
- $v$ - velocity of flow, mL/min
- $t$ - time of adsorption penetration

According to the above equation, build a model which x-axis based on ln$[(C_0/C_t) - 1]$, y-axis based on time $t$. The Thomas model diagrams under different filling layer heights and different initial influent water concentrations are calculated, and the Thomas rate constant $k_T$ under dynamic adsorption can be obtained by linearly fitting the slope and intercept in the equation. From the operation of the adsorption column to the depletion of adsorption, the total adsorption of the adsorption column Mad (Hasan S H, 2010) can be calculated from the integral area enclosed by the straight line of the penetration curve and the initial concentration, the equation can be showed below:

$$M_{ad} = \frac{v}{1000} \int_0^{t_e} (c_0 - c_t) dt$$

In this equation:
- $t_e$ - penetration time

The dynamic saturation adsorption quantity $q_e$ of UCWTR can be calculated by the following equation:

$$q_e = \frac{M_{ad}}{m}$$

According to the above two equations, the dynamic saturation adsorption capacity $q_e$ of UCWTR for dynamic adsorption of lead and cadmium can be calculated.

2.3.2. Yoon-Nelson model. The Yoon-Nelson model (Yoon Y H., 1984) can describe the penetration behavior of the adsorbate on the adsorbent. The model believes that there is a certain proportional relationship between the adsorption probability of the adsorbent substance and the probability that each adsorbed substance may be adsorbed. The Yoon-Nelson model has no restrictions on the physical characteristics of the adsorption bed (Tsai W T, 1999). The Yoon-Nelson model can briefly describe the dynamic adsorption process. Compared with other models, its dynamic adsorption parameters are simpler. The equation can be shown below:

$$\ln\left(\frac{C_t}{C_0 - C_t}\right) = k_{YN} - t k_{YN}$$

In this equation:
- $k_{YN}$ - Yoon-Nelson rate constant, 1/min$^{-1}$
- $t$ - Time required to penetrate 50% adsorbent, min

According to the equation, A model with $\ln\left(\frac{C_t}{C_0 - C_t}\right)$ as the y-axis and time $t$ as the x-axis is established, and the $k_{YN}$ and $t$ values can be obtained by linearly fitting the slope and intercept in the equation.
3. Results and analysis

3.1. Effect of packing layer height on adsorption
Figure 1 shows the penetration curves of lead adsorption by UCWTR at different filler layer heights; Figure 2 shows the penetration curves of cadmium adsorption by UCWTR at different heights of filler layers.

As can be seen from Figure 1 and Figure 2, when heights of filler layers in the reaction column is 5 cm, 10 cm and 15 cm, the penetration time of lead and cadmium are 140 min, 180 min, 220 min and 160 min, 220 min, 280 min, and the depletion time are 300 min, 320 min, 360 min, and 320 min, 360 min, and 440 min respectively. The penetration time and depletion time both increase with the height of filler layer. This is because the height of filler layer increases, the quality of the adsorbent also increases, and the flow rate of influent is 10 mL/min without change, the contact time of the ceramsite as an adsorbent and the wastewater solution is extended, so the time for ceramsite surface adsorption and internal diffusion adsorption of pollutants is also extended. The dynamic adsorption capacity of ceramsite is related to the depletion time and the height of filler layers, although appropriately increasing the height of filler layer can increase removal effect and prolong depletion time of the
reaction column, increasing the height will increase the mass transfer resistance, and the more adsorbent, the higher the running cost.

3.2 Effect of initial concentration on adsorption

Figure 3 shows the penetration curves of lead adsorption by UCWTR at different initial concentrations; Figure 4 shows penetration curves of cadmium adsorption by UCWTR at different initial concentrations.

![Figure 3](image3.png)

**Figure 3.** Penetration curves of lead adsorption by UCWTR at different initial concentrations

![Figure 4](image4.png)

**Figure 4.** Penetration curves of cadmium adsorption by UCWTR at different initial concentrations

It can be seen from Figures 3 and 4 that when the initial concentration of dynamic adsorption is set to 50 mg/L, 100 mg/L, 150 mg/L, the penetration times of lead and cadmium are 240 min, 180 min, 140 min and 300 min, 220 min, and 180 min, the exhaustion times of lead and cadmium are 400 min, 320 min, and 260 min, and 460 min, 360 min, and 300 min, respectively. The penetration time and exhaustion time of lead and cadmium decrease as the initial concentration increases. This phenomenon is due to the increase in the mass transfer driving force as the initial concentration increases, resulting in an increased exposure between the lead and cadmium ions and the UCWTR adsorption point during the adsorption process, leading to the UCWTR adsorption point being occupied in a shorter time, while the penetration curve is steeper (Zhang L. Q., 2016).
3.3. The penetration curve fitting of dynamic adsorption lead

For the experiment in Section 1.2, the Thomas fitting was performed on the data of UCWTR dynamic adsorption lead under the different height of columns and initial concentrations. The fitting results are shown in Figures 5 and 6, and the fitting parameters are shown in Table 1.

![Figure 5. The Thomas fitting of UCWTR adsorption Pb under different height of columns](image1)

![Figure 6. The Thomas fitting of UCWTR adsorption Pb under different initial concentration](image2)

| Filler layer height/(cm) | Flow rate /(mL/min) | The initial concentration(mg/L) | Initial pH | Thomas $k_T \times 10^{-4}$ | $q_e$(mg/g) | $R_T^2$ |
|-------------------------|---------------------|-------------------------------|------------|-----------------------------|-------------|--------|
| 5                       | 10                  | 100                           | 5          | 2.250                       | 6.31        | 0.9245 |
| 10                      | 10                  | 100                           | 5          | 2.027                       | 7.93        | 0.9540 |
| 15                      | 10                  | 100                           | 5          | 1.930                       | 6.96        | 0.9464 |
| 10                      | 10                  | 50                            | 5          | 2.236                       | 4.99        | 0.9248 |
| 10                      | 10                  | 100                           | 5          | 2.070                       | 8.07        | 0.9527 |
| 10                      | 10                  | 150                           | 5          | 1.964                       | 9.22        | 0.9268 |
It can be seen from Table 1 that the Thomas fit correlation coefficient $R_T^2$ of UCWTR adsorption lead exceeds 0.92, indicating that the dynamic adsorption process of UCWTR adsorption lead is more in line with the Thomas model. When the initial concentration is set to 100 mg/L and the filler layer height is 5 cm, 10 cm, and 15 cm, the $k_T$ is $2.250 \times 10^{-4}$, $2.027 \times 10^{-4}$, and $1.930 \times 10^{-4}$; When the filler layer height is set to 10 cm and the initial concentrations are 50 mg/L, 100 mg/L, and 150 mg/L, the $k_T$ is $2.236 \times 10^{-4}$, $2.070 \times 10^{-4}$, and $1.964 \times 10^{-4}$, respectively. The $k_T$ decreases with the increase of the filler layer height and decreases with the increase of the initial concentration, while the dynamic saturation adsorption quantity $q_e$ first increases and then decreases with the increase of the height of filler layer, and increases with the increase of the initial concentration, it shows that height of the filler layer increases, the mass transfer resistance increases, the penetration time prolongs, and the rate constant decreases. The initial concentration increases and the mass transfer power increases, so that the adsorption points in the adsorption column can be quickly covered, and the penetration time is reduced.

For the experiments in Section 1.2, the Yoon-Nelson model was fitted to the data of UCWTR dynamic adsorption lead under different height of columns and initial concentrations. The fitting results are shown in Figures 7 and 8, and the fitting parameters are shown in Table 2.

**Figure 7.** The Yoon-Nelson fitting of UCWTR adsorption Pb under different height of columns.

**Figure 8.** The Yoon-Nelson fitting of UCWTR adsorption Pb under different initial concentration.
Table 2. Parameters of Yoon-Nelson model under different conditions using linear regression analysis of Pb.

| Filler layer height/(cm) | Flow rate/(mL/min) | The initial concentration/(mg/L) | Initial pH | Yoon-Nelson kYN×10⁻² t/(min) | RYN²  |
|-------------------------|--------------------|----------------------------------|-----------|-----------------------------|-------|
| 5                       | 10                 | 100                              | 5         | 2.276                       | 219.8 | 0.9261 |
| 10                      | 10                 | 100                              | 5         | 2.102                       | 251.8 | 0.9451 |
| 15                      | 10                 | 100                              | 5         | 1.991                       | 292.9 | 0.9341 |
| 10                      | 10                 | 50                               | 5         | 1.822                       | 306.3 | 0.9739 |
| 10                      | 10                 | 100                              | 5         | 1.980                       | 248.9 | 0.9527 |
| 10                      | 10                 | 150                              | 5         | 2.027                       | 215.4 | 0.9013 |

It can be seen from Table 2 that the correlation coefficients RYN² fitted by the Yoon-Nelson model of UCWTR adsorption lead all exceed 0.90, indicating that the dynamic adsorption process of UCWTR adsorption lead is more in line with the Yoon-Nelson model. When the initial concentration is set to 100 mg/L and the filler layer heights are 5 cm, 10 cm, and 15 cm, kYN is 2.276×10⁻², 2.102×10⁻², and 1.91×10⁻²; when the filler layer height is set to 10 cm and the initial concentrations are 50 mg/L, 100 mg/L, and 150 mg/L, kYN is 1.822×10⁻², 1.98×10⁻², and 2.027×10⁻², respectively. The kYN value increases with decreasing of filler layer height and the increasing of initial concentration, and the t value increases with increasing filler layer height and decreasing initial concentration, this is because the mass transfer resistance comes from the thickness of the filler layer, the influence of the mass transfer resistance on the adsorption rate leads to an increase in the time of saturation adsorption, with the increase of initial concentration, the value of kYN increases, while the value of t decreases, and the adsorbent reaches the saturation point earlier.

3.4. The penetration curve fitting of dynamic adsorption cadmium

For the experiments in Section 1.2, the Thomas model fitting was performed on the data of UCWTR dynamic adsorption cadmium under different height of columns and initial concentrations. The fitting results are shown in Figure 9 and Figure 10, and the fitting parameters are shown in Table 3.

![Figure 9. The Thomas fitting of UCWTR adsorption Cd under different height of columns.](image-url)
Figure 10. The Thomas fitting of UCWTR adsorption Cd under different initial concentration.

Table 3. Parameters of Thomas model under different conditions using linear regression analysis of Cd.

| Filler layer height/(cm) | Flow rate/(mL/min) | The initial concentration/(mg/L) | Initial pH | Thomas kT×10^-4 | q0/(mg/g) | RT^2 |
|-------------------------|--------------------|---------------------------------|------------|-----------------|-----------|------|
| 5                       | 10                 | 100                             | 5          | 2.154           | 9.96      | 0.9544 |
| 10                      | 10                 | 100                             | 5          | 1.936           | 8.20      | 0.9675 |
| 15                      | 10                 | 100                             | 5          | 1.668           | 7.07      | 0.9629 |
| 10                      | 10                 | 50                              | 5          | 1.552           | 6.09      | 0.9549 |
| 10                      | 10                 | 100                             | 5          | 2.024           | 8.79      | 0.9699 |
| 10                      | 10                 | 150                             | 5          | 1.908           | 11.52     | 0.9161 |

It can be seen from Table 3 that the Thomas fit correlation coefficient R_T^2 of UCWTR adsorption of cadmium exceeds 0.91, indicating that the dynamic adsorption process of UCWTR adsorption cadmium is more in line with the Thomas model. When the initial concentration is set to 100 mg/L and filler layer heights are 5 cm, 10 cm, and 15 cm, the k_T is 2.154×10^-4, 1.936×10^-4, and 1.668×10^-4, respectively; when the filler layer height is set to 10 cm and the initial concentrations are 50 mg/L, 100 mg/L, and 150 mg/L, the k_T is 1.552×10^-4, 2.024×10^-4, and 1.908×10^-4, respectively. The Thomas fit of UCWTR adsorption of cadmium has a similar rule to that of lead adsorption. The reaction rate constants k_T and q_0 both increase as the height of the filler layer decreases and the initial concentration increases.

For the experiments in Section 1.2, the Yoon-Nelson model was fitted to the data of UCWTR dynamic adsorption cadmium under different columns heights and initial concentrations. The fitting results are shown in Figure 11 and Figure 12, and the fitting parameters are shown in Table 4.
Figure 11. The Yoon-Nelson fitting of UCWTR adsorption Cd under different height of columns.

Figure 12. The Yoon-Nelson fitting of UCWTR adsorption Cd under different initial concentration.

Table 4. Parameters of Yoon-Nelson model under different conditions using linear regression analysis of Cd.

| Filler layer height/(cm) | Flow rate/(mL/min) | The initial concentration/(mg/L) | Initial pH | Yoon-Nelson |
|-------------------------|--------------------|----------------------------------|------------|-------------|
|                         |                    |                                  |            | kYN×10⁻²    | t/(min)     | RYN²       |
| 5                       | 10                 | 100                              | 5          | 2.216       | 241.3      | 0.9584     |
| 10                      | 10                 | 100                              | 5          | 2.062       | 295.2      | 0.9746     |
| 15                      | 10                 | 100                              | 5          | 1.798       | 346.9      | 0.9750     |
| 10                      | 10                 | 50                               | 5          | 1.574       | 370.3      | 0.9615     |
| 10                      | 10                 | 150                              | 5          | 2.042       | 294.0      | 0.9703     |
| 10                      | 10                 | 150                              | 5          | 1.956       | 258.3      | 0.9275     |

It can be seen from Table 4 that the correlation coefficients RYN² fitted by the Yoon-Nelson model of cadmium adsorption by UCWTR all exceed 0.92, indicating that the dynamic adsorption process of cadmium adsorption by UCWTR is more in line with the Yoon-Nelson model. When the initial concentration is set to 100 mg/L and the filler layer heights are 5 cm, 10 cm, and 15 cm, the kYN is
2.216×10^{-2}, 2.062×10^{-2}, and 1.798×10^{-2}; When filler layer height is set to 10 cm and the initial concentrations are 50 mg/L, 100 mg/L, and 150 mg/L, kYN is 1.574×10^{-2}, 2.042×10^{-2}, and 1.956×10^{-2}, respectively. The Yoon-Nelson fitting of UCWTR to cadmium has a similar rule to that of UCWTR to lead. The t increases with the filler layer height increases and the initial concentration decreases, and kYN increases with the filler layer height decreases and the initial concentration increases.

4. Conclusion
The study on the adsorption of lead and cadmium using UCWTR as the adsorption material shows that:
(1) According to the dynamic adsorption experiment of UCWTR for lead and cadmium, with the increase of the height of the filler layer in the reaction column, the quality of the ceramsite is improved, and the penetration time and depletion time are extended; as the influent concentration in the reaction column increases, the driving force for mass transfer increases, the UCWTR adsorption point is occupied in a shorter time, and the penetration time and exhaustion time become shorter.
(2) The dynamic adsorption process of lead and cadmium adsorption by UCWTR is more in line with Thomas model and Yoon-Nelson model, and the correlation coefficient R^2 is greater than 0.9. Under the conditions of initial flow rate v=10 mL/min, filler layer height h=10 cm, and initial concentration C_0=150 mg/g, the fitted dynamic saturation adsorption capacities of UCWTR for lead and cadmium are 9.22 mg/g, 11.52 mg/g, the static adsorption capacity of lead and cadmium is 1.48, 155 times of its. k_T decreases with the increase of the filler layer height and the initial concentration; kYN decreases with the increase of the filler layer height and increases with the increase of the initial concentration; the time required to penetrate 50% of the adsorbent t increases with the increases of filler layer height and decreases with the increase of initial concentration, indicating that the solution did not reach full contact with UCWTR during the dynamic adsorption process.

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