Research Article

The Breakthrough Time Analyses of Lead Ions in CCL considering Different Adsorption Isotherms

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As the 2 m thick compacted clay liner with permeability coefficient of $1 \times 10^{-7}$ cm/s is required in the Chinese technical specifications about landfill, the performance of this compacted clay liner was analyzed considering three different adsorption isotherms (convex, straight, and concave). The effects of source concentration, adsorption mode, and waterhead on the breakthrough curve and breakthrough time of Pb$^{2+}$ were discussed. The results indicate that reducing the concentration of pollution sources is beneficial to prolonging the breakthrough time. With the waterhead of 10 m, the absolute breakthrough time, respectively, increased from 2.77 to 3.7 years (concave type isotherm), from 17.63 to 26.58 years (straight type isotherm), and from 35.43 to 59.6 years (convex type isotherm), as the source concentration decreased from 1000 mg/L to 10 mg/L. The effect of adsorption isotherm type on the performance of the barrier is very obvious: with the waterhead of 10 m, the absolute breakthrough time corresponding to the convex isotherm is more than twice that of the straight adsorption isotherm, and more than 12.8 times that of the concave isotherm. The absolute breakthrough time corresponding to 0.3 m waterhead is more than 4 times that of 10 m, and reducing the waterhead can effectively increase the breakthrough time.

1. Introduction

Compacted clay liner (CCL) is used as a common type of barrier in landfills of China. Especially in the large number of existing old landfills, CCLs are basically used as the main antifouling barriers. Compacted clay can not only be used as an important impermeable material combined with geomembrane and GCL to form a composite liner, but also be a separate liner [1]. The technical parameters of CCL are specified in many of the specifications [2–4]: the permeability coefficient of natural clay liner or modified clay liner shall not be greater than $1.0 \times 10^{-9}$ m/s, and the thickness of liners at the field bottom and the four walls shall not be less than 2 m.

As the horizontal antiseepage and antifouling structure of the landfill, the performance of the liner is directly related to the effect of groundwater pollution control. In the field of barrier performance analysis and breakthrough time evaluation, the former researchers have done a lot of research work [5–15]. Acar and Haider [8] proposed a set of design methods for soil barrier in landfills based on one-dimensional pollutant convection-dispersion model and explained the analysis methods by experiments. Shackelford [9] believed that the US specification requires that 0.9 m thick compacted clay with a permeability coefficient of $1.0 \times 10^{-9}$ m/s may not meet the antifouling requirements within the operational life of the antifouling facility (usually 40 to 50 years) and proposed that the design of liner thickness should meet the condition that the target pollutant within the design period should not exceed the concentration limit. Based on the one-dimensional convection-dispersion analytical solution of a semi-infinite thickness uniform medium, the transit-time design method for clay liner was proposed. Shackelford and Glade [10] discussed the pollution leakage of the soil downstream under different $R_d$ conditions during linear adsorption by theoretical analysis.
2. Theoretical Analyses for Performance of CCL

2.1. Model and Main Parameters. According to the requirements of specification, we took the 2 m thick clay liner with permeability coefficient of $1 \times 10^{-7}$ cm/s as the analysis object. The common adsorbent heavy metal ion Pb$^{2+}$ was selected as the target pollutant. Figure 1 is a schematic diagram of a horizontal clay liner. It is assumed that the source concentration keeps constant, the upstream of liner is the pollution source solution, and the downstream of the liner is aquifer. As one-dimensional migration of pollutants takes place in the liner, the function of the liner is to prevent pollutants in the leachate from entering the underlying aquifer through vertical seepage.

According to the on-site investigation, the water level of landfill in China is high, which can reach more than ten meters [23, 24], while the landfill specifications require that the height of the waterhead on the liner should not exceed 30 cm. Therefore, in the analyses, the waterhead difference between the upstream and downstream barriers was considered in two cases: when the waterhead difference is high, the height of the waterhead on the liner is taken as 10 m; when the waterhead difference is low, the height of the waterhead on the liner is taken as 0.3 m.

Three source concentrations were considered in the analyses: 10 mg/L, 100 mg/L, and 1000 mg/L. According to the literature reports, the concentration range of Pb$^{2+}$ in the leachate from abroad landfill is 0.001–5 mg/L [25, 26], while that in the leachate from the landfill of China is 0.196–57.08 mg/L [27, 28]. Therefore, according to the domestic situation, 10 mg/L is taken for low concentration. Compared with the landfill, the content of heavy metal ions in the waste liquid of slag yard and tailings pond is much higher. Chang et al. [29] surveyed several major heavy metal mining areas of Youxi lead-zinc mine, Liancheng lead-zinc mine, and Liancheng manganese mine in Fujian and found that the highest content of Mn, Zn, Pb, and Cd in the soil of the mining area, respectively, reached 92546, 27454, 23792, and 248 mg/kg. Ji et al. [30] analyzed the composition and content of heavy metals in 12 electroplating sludge samples from different electroplating enterprises. The results show that the main heavy metal species in the sludge are Cr, Cu, Ni, Pb, Zn, and Sn, and their content ranges from 40 to 14230, 22 to 16000, 14 to 20000, 5 to 120, 37 to 91000, and 32 to 19000 mg/kg. If the content of heavy metals in the solid phase is so high, the concentration of heavy metals in the corresponding pore water will also be very high. Therefore, in order to consider the effect of the source concentration value on the migration of heavy metals, the other two high concentrations in the analysis were taken as 100 mg/L and 1000 mg/L, respectively. The remaining parameters are as follows [31, 32]: the barrier porosity $n$ was taken as 0.62, the dry density was taken as 1.04 g/cm$^3$, the water content was taken as 0.6, the effective diffusion coefficient of Pb$^{2+}$ was taken as $3.3 \times 10^{-10}$ m$^2$/s, and the dispersity was taken as 0.04 m.

2.2. Scheme for Analyses. In order to study the effect of various parameters on the breakthrough time of the CCL with different adsorption models, three types of adsorption isotherms were selected: convex, straight, and concave. Langmuir sorption isotherm, linear sorption isotherm, and Freundlich sorption isotherm were used as representative models, respectively.

12 cases were set up for analysis in this paper, as shown in Table 1. Cases 1–3, Cases 4–6, and Cases 7–9 aim to compare the impact of different source concentrations (1000 mg/L, 100 mg/L, and 10 mg/L) on breakthrough time. Cases 1–3 are Straight isotherms, Cases 4–6 are concave isotherms, and Cases 7–9 are convex isotherms. The values of Cases 1–3 adsorption parameters refer to the results of kaolin adsorption experiments on lead in Zeng [32]. The parameters of the Langmuir adsorption isotherm (convex) of Cases 4–6 are $Q_0 = 7$ mg/g, $b = 0.00148$ L/mg, and the parameters of the Freundlich model (concave) of Cases 7–9 are $K_F = 0.000132$ L/g, $n_F = 1.5$. The maximum adsorption amount of 1000 mg/L on the adsorption isotherm...
corresponding to the parameters of Langmuir model and Freundlich model is equal to that of the linear model ($R_d = 8$) and can represent the convex and concave isotherms, respectively. Therefore, Case 1, Case 4, and Case 7, Case 2, Case 5, and Case 8, and Case 3, Case 6, and Case 9 can compare the effects of different adsorption modes, and the corresponding source concentrations are 1000 mg/L, 100 mg/L, and 10 mg/L, respectively.

Case 1 and Case 10, Case 2 and Case 11, and Case 3 and Case 12 are the analysis groups for comparing the impact of waterhead on migration, respectively. Case 1 and Case 10 are straight isotherms, Case 2 and Case 11 are convex isotherms, and Case 3 and Case 12 are concave isotherms. Considering the effect of waterhead is not only the direct effect of waterhead on breakthrough time, but also the effect of velocity on adsorption [32–34]. Therefore, the adsorption parameters of different waterheads are different; refer to Zeng [32] for specific values. Case 11 and Case 12 take the adsorption isotherm, respectively, higher than Case 4 and Case 7. The Langmuir model parameters of Case 11 are taken as $Q_0 = 12$ mg/g, $b = 0.00148$ L/mg, and the Freundlich model parameters of Case 12 are taken as $K_F = 0.000217$ L/g, $n_f = 1.5$. At the same time, the maximum adsorption amounts corresponding to 1000 mg/L on the adsorption isotherm of Case 10, Case 11, and Case 12 are equal. Therefore, similar to Case 1, Case 4 and Case 7, Cases 10–12 can compare the effects of different adsorption modes at low waterhead. The specific adsorption model parameters used in the analyses are shown in Figure 2.

### 3. Analyses and Calculation Methods

The migration of pollutants in soil mainly involves convection, diffusion, mechanical dispersion, and adsorption processes. The one-dimensional convection-dispersion process considering equilibrium adsorption can be described by the following formula:

$$ R_d \frac{\partial C_r (x, t)}{\partial t} = D_n \frac{\partial^2 C_r (x, t)}{\partial x^2} - v_r \frac{\partial C_r (x, t)}{\partial x}, \quad (1) $$
where \( v_s \) is the advection velocity (L/T), \( D_h \) is the coefficient of hydrodynamic dispersion (L²/T), \( \alpha \) is the dispersity, \( R_d \) is the retardation factor, \( \tau \) is the tortuosity factor, \( D_d \) is the free diffusion coefficient (L²/T), \( D_m \) is the mechanical dispersion (L²/T), \( \rho_d \) is the dry density (M/L³), \( K_d \) is the distribution coefficient (L³/M), \( \theta \) is the volumetric water content, \( C_w \) is the resident concentration (M/L³) in pore water, \( t \) is the time (T), and \( z \) is the distance from the source (L).

For one-dimensional migration, the expressions of \( C_r \) concentration in pore water and \( C_e \) concentration in effluent solution are discussed in relevant literatures [35, 36]. Zeng [36] discussed boundary conditions by experiments and recommended the best expressions of \( C_r \) and \( C_e \).

According to the adsorption model, the relationship between total soil concentration and pore water concentration can be established.

### 3.1. Linear Sorption Isotherm.

According to the linear model, it is assumed that there is a relationship between the adsorption concentration in the soil and the concentration of the pore solution:

\[
C_s = K_d C_w, \tag{2}
\]

where \( C_s \) is the mass of pollutants absorbed per unit of soil, \( K_d \) is the partition coefficient, and \( C_w \) is the pore water concentration.

According to the mass balance, the total amount of pollutants in the soil is

\[
m_s C_{sto} = \frac{m_w w_w}{\rho_w} C_w + m_s C_s, \tag{3}
\]

where \( m_s \) is the mass of soil particles, \( C_{sto} \) is the total concentration of pollutants in the soil, and \( w \) is the water content. The following formula is processed:

\[
C_{sto} = \frac{w_w}{\rho_w} C_w + C_s. \tag{4}
\]

The following formula is obtained by substituting (2) into equation (4):

\[
C_{sto} = \frac{w_w + K_d}{\rho_w} C_w \tag{5}
\]

In saturated soil, \( S_e = 1 \), so

\[
C_{sto} = \frac{n}{\rho_d} R_d C_w. \tag{6}
\]

Substituting the above relationship into the original pore water convection-dispersion-adsorption equation, the control equation of the pollutant mass concentration in the soil is as follows:


\[ R_d \frac{\partial C_{sto}}{\partial t} = D_h \left( \frac{\partial^2 C_w}{\partial x^2} \right) - v_s \left( \frac{\partial C_w}{\partial x} \right). \]  

(7)

Therefore, the original analytical solution of pore water only needs to be multiplied by \((n/\rho_d)R_d\), which is the analytical solution of the total concentration in the soil, so the analytical solution can be used to fit the total concentration profile.

3.2. Nonlinear Sorption Isotherm. If the sorption of heavy metal on kaolin meets nonlinear isotherm, the relationship between the total concentration in the soil and the pore water concentration can still be obtained.

(A) Langmuir nonlinear sorption isotherm

\[ q_e = \frac{Q_0 b C_w}{1 + b C_w}, \]

\[ R_d = 1 + \frac{\rho_d}{n} \frac{Q_0 b}{(1 + b C_w)^2}, \]  

(8)

\[ C_{sto} = q_e + C_w \ast V = \frac{Q_0 b C_w}{1 + b C_w} + C_w \ast \frac{w}{\rho_w}. \]

(B) Freundlich nonlinear sorption isotherm

\[ q_e = K_F C_w^{n_f}, \]

\[ R_d = 1 + \frac{\rho_d}{n} K_F n_f C_w^{n_f - 1}, \]  

(9)

\[ C_{sto} = q_e + C_w \ast V = K_F C_w^{n_f} + C_w \ast \frac{w}{\rho_w}. \]

For nonlinear sorption isotherm, because the total concentration \( C_{sto} \) and pore water concentration \( C_w \) are not simple linear relationships, it is impossible to obtain an analytical solution of the total concentration, but the difference method or the simulation software for pollutant migration can be used to simulate the total concentration. The transports with nonlinear adsorption model in this paper were analyzed using the contaminant migration module CTRAN/W in GeoStudio. The boundary conditions in the software were set the same as those in the analytical model: the inflow flux is continuous, and the infinite distance gradient of the outflow is 0. Firstly, \( C_w \) of pore water concentration was solved by CTRAN/W, and then \( C_{sto} \) profile was obtained according to the relationship between \( C_w \) and \( C_{sto} \) presented above.

4. Results and Discussion

4.1. Effect of Source Solution Concentration on Barrier Performance and Breakthrough Time. Figures 3(a)–3(c) show the comparison of effluent concentrations corresponding to different source concentrations of straight type (linear model), convex type (Langmuir model), and concave type (Freundlich model), respectively. Figure 3 shows the relative breakthrough time corresponding to the relative breakthrough standard, which is the time of the outflow concentration reaching 10% of the source concentration, expressed as \( t_{0.1} \). In the “Groundwater Quality Standards” (GB/T 14848-93) [37], class IV water quality standard based on the requirements of water used in agricultural and industrial is specified as follows: the upper limit of the concentration of Pb²⁺ is 0.1 mg/L, which is the same as the concentration limit of lead emission in the existing and new domestic waste landfills as stipulated in the “Pollution Control Standard for Domestic Waste Landfills” (GB 16889-2008) [38]. Therefore, the concentration of 0.1 mg/L Pb²⁺ was taken as the standard for evaluating the absolute breakthrough of the barrier. The breakthrough time corresponding to the absolute breakthrough standard (\( C_w = 0.1\) mg/L) is characterized by \( t_a \). The specific results are shown in Table 1. By comparing the effluent curve, we can see that, corresponding to different adsorption models, the effect of source concentration on the migration of pollutants is different: the outflow concentration curves corresponding to the straight adsorption isotherm of different source concentrations coincide with each other completely; in the convex isotherm group, the curve of outflow concentration with high source concentration appears the latest, while the curve of outflow concentration with low source concentration on the right appears the earliest, which is on the left most in the Figure 3(b); On the contrary, the case with small source concentration in concave isotherm group has the earliest outflow concentration curve, and the case with large source concentration has the latest outflow concentration curve.

Figure 4 shows the breakthrough time corresponding to different source concentrations of straight type (linear model), convex type (Langmuir model), and concave type (Freundlich model). There are two kinds of breakthrough time including the breakthrough time \( t_0 \) corresponding to absolute breakthrough standard (\( C_w = 0.1\) mg/L) and the breakthrough time \( t_{0.1} \) corresponding to relative breakthrough standard (\( C_w = 10\%C_0 \)). As shown in Figure 4, the breakthrough time \( t_{0.1} \) corresponding to the upper convex type (Langmuir model) decreases with the increase of the source concentration, and \( t_{0.1} \) corresponding to the concave type (Freundlich model) increases as the source concentration increases; the breakthrough times \( t_{0.1} \) corresponding to the linear models of different source concentrations are equal. The \( t_a \) corresponding to the three adsorption models decreases with the increase of the source concentration. And the convex type (Langmuir model) has the largest effect. The breakthrough time of the upper convex type significantly reduces as the concentration increases. Then, the linear model has the medium effect, and the concave type (Freundlich model) has the smallest effect. Source concentrations decreased from 1000 mg/L to 100 mg/L, \( t_a \) increased by 0.33 years (concave type), 3.65 years (straight type), and 10.84 years (convex type); source concentration decreased from 100 mg/L to 10 mg/L, \( t_a \) respectively, added
Figure 3: Outflow concentration curve of different concentrations. (a) Cases 1–3 (straight type). (b) Cases 4–6 (convex type). (c) Cases 7–9 (concave type).

Figure 4: Relationship between $t_{a0}$, $t_{a0.1}$ and source concentration in Cases 1–9.
0.6 years (concave type), 5.3 years (straight type), and 13.33 years (convex type).

Therefore, reducing the concentration of pollution source is beneficial to prolonging the breakthrough time: when the adsorption mode is convex, reducing the source concentration has the most obvious effect on prolonging the breakthrough time; when the adsorption mode is concave, reducing the source concentration has little effect on prolonging the breakthrough time. In order to improve the service performance of the barrier and prolong the breakthrough time, the concentration of the pollution source in the site should be reduced as much as possible in the project. Strongly adsorbent materials can be added to the solution of the pollution source to reduce the ion concentration in the solution. Or a thin layer of high adsorption material can be placed upstream of the barrier to reduce the concentration of pollutants entering the barrier.

4.2. Effect of Different Adsorption Models on Barrier Performance and Breakthrough Time. The comparison of the outflow concentrations corresponding to different adsorption models with source concentrations of 1000 mg/L, 100 mg/L, and 10 mg/L is shown in Figure 5(a)–5(c), respectively. When the source concentration is 100 mg/L and 10 mg/L, the outflow concentration curve corresponding to the convex type (Langmuir model) appears the latest, and the outflow concentration curve corresponding to the downward concave type (Freundlich model) appears earliest. When the source concentration is 1000 mg/L, the outflow concentration of the concave type (Freundlich model) appears first, then the linear model, and the convex type (Langmuir model) the latest. The three curves have common intersection points. After the intersection point, the outflow concentration corresponding to the convex type (Langmuir model) is the highest and reaches equilibrium first, the straight type is in the middle, and the concave type (Freundlich model) is the lowest. Figure 5(d) is a comparison of Case 10, Case 11, and Case 12, with the same rules as in Figure 5(a).

The comparison between \( t_a \) and \( t_{0.1} \) is shown in Figure 4. As shown in Figure 4, at the same source concentration, \( t_a \) and \( t_{0.1} \) corresponding to the three adsorption models always have the same pattern: the cases with convex type have the longest breakthrough time, followed by the cases with straight type and the cases with concave type. That means that \( t_a \) and \( t_{0.1} \) both correspond to the positional relationship of the isotherm: the higher the adsorption isotherm, the longer the breakthrough time, and the lower the adsorption isotherm, the shorter the breakthrough time. The \( t_a \) corresponding to the convex type isotherm is more than twice the \( t_a \) corresponding to the straight type isotherm, and more than 12.8 times that corresponding to the concave type isotherm, and the gap increases as the source concentration decreases.

The effect of adsorption isotherm on the antifouling performance of the barrier is very obvious: the cases with the convex type isotherm have the longest breakthrough time, followed by those with the straight type isotherm, and the cases with the concave type isotherm have the shortest breakthrough time. Therefore, in the project, choose the material whose isotherm has a convex shape, or modify the material to change its adsorption isotherm so that its adsorption isotherm becomes an obvious convex shape and, thus, can effectively extend the breakthrough time of the barrier.

4.3. Effect of Waterhead on Barrier Performance. Figures 6(a) and 6(b) show the pore water concentration profiles and the total concentration profiles corresponding to different waterheads, respectively, and the corresponding time is 50 years. As mentioned above, the effect of different waterheads discussed here considers the effect of flow rate on adsorption.

The adsorption parameters used in low waterhead cases are larger than those in high waterhead cases. As shown in Figure 6(a), the relationship of pore water concentration is obvious, and the model with a low waterhead has to a shallow concentration front. As shown in Figure 6(b), the distribution of the total concentration profile in the soil is different from that of the pore water concentration. The pollutant migration front of the low waterhead cases (Case 10, Case 11, and Case 12) is shallow, and the corresponding total concentration front is also shallow, but as shown in Figure 2, the adsorption isotherm with a low waterhead is higher, the adsorption is larger, and the total concentration in the soil at the top of the liner is larger. Therefore, the front of the low waterhead case is shallower, but the total concentration value at the top is larger. So, there is an intersection between the total concentration profile of the low head and that of the high head.

The outflow breakthrough curves corresponding to different waterheads (10 m (Cases 1, 4, 7), 0.3 m (Cases 10, 11, 12)) are shown in Figure 5(a) and 5(d). Obviously, the breakthrough curve with low waterhead flows out late. Figure 7 shows the absolute breakthrough time \( t_a \) and relative breakthrough time \( t_{0.1} \) corresponding to different waterheads. As shown in Figure 7, the \( t_a \) and \( t_{0.1} \) corresponding to the 0.3 m head are far greater than 10 m. It can be found that the \( t_a \) corresponding to the 0.3 m waterhead is about 4 times more than the 10 m waterhead, and \( t_{0.1} \) is more than 6 times. Therefore, reducing the waterhead can effectively increase the breakthrough time. It is considered that the decrease of velocity can increase the adsorption of barrier material; that is to say, the decrease of waterhead can not only reduce the convection effect, but also increase the retardation effect of barrier. In the project, methods such as adding membranes to reduce the permeability of the barrier...
Figure 5: Comparison of the outflow concentration of different adsorption isotherms. (a) Cases 1, 4, 7 ($C_0 = 1000$ mg/L). (b) Cases 2, 5, 8 ($C_0 = 100$ mg/L). (c) Cases 3, 6, 9 ($C_0 = 10$ mg/L). (d) Cases 10, 11, 12 ($C_0 = 1000$ mg/L).

Figure 6: Comparison of pore water concentration and total soil concentration with different waterheads (50 years). (a) Pore water concentration profiles. (b) Total concentration in soil profiles.
can be considered to reduce the seepage velocity in the clay barrier [39].

5. Conclusion

The service performance of 2 m thick CCLs that meet the specifications is analyzed considering three type isotherms (convex, straight, and concave) for the first time. The effects of source concentration, isotherm type, and waterhead on the breakthrough time of Pb\(^{2+}\) are discussed. And the following conclusions can be drawn.

Reducing the concentration of the pollution source is beneficial to prolong the breakthrough time. When the source concentration decreased from 1000 mg/L to 10 mg/L, the absolute breakthrough time, respectively, increased by 0.93 years (concave type isotherm), 8.95 years (straight type isotherm), and 24.17 years (convex type isotherm). The effect of adsorption mode on the performance of the barrier is very obvious. The absolute breakthrough time corresponding to the convex type isotherm is more than twice that of the straight type isotherm, and more than 12.8 times that of the concave type isotherm. Reducing the waterhead can effectively increase the breakthrough time.

In order to improve the service performance of the barrier and prolong the breakthrough time, the following methods can be used in the project: add strong adsorption materials into the pollution source solution to reduce the concentration; or lay a thin layer of highly absorbent materials upstream of the barrier to reduce the pollutant concentration entering the barrier; select the material with convex type, or modify the material to change its adsorption mode from concave type or linear to convex type; take some measures such as adding membranes to reduce the permeability of the barrier to reduce the seepage velocity in the soil barrier.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest regarding the publication of this paper.

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