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# Experimental study on the application of coal gangue for landfill liner

## Etude expérimentale de l'application de la gangue de charbon pour le revêtement de décharge

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**ABSTRACT:** Clay minerals are usually used to construct the liners in landfill to prevent the leakage of leachate and the diffusion of contaminants into the groundwater. In order to conserve clay minerals, many other materials have been proposed as a substitute of clay minerals. In the present study, the feasibility of using coal gangue, an industrial solid waste, as landfill liner material is investigated through laboratory tests in terms of the mechanical, hydraulic and sorption characteristics. The results indicated that the hydraulic conductivity of the coal gangue can be less than the regulatory  $1 \times 10^{-7}$  cm/s requirement for a monolayer landfill liner with a void ratio less than 0.44. The coal gangue also showed remarkable sorption capacity on  $\text{Cd}^{2+}$ . Therefore, the coal gangue has potential to be used as landfill liners to conserve clay minerals as well as recycle industrial solid waste.

**RÉSUMÉ :** Les minéraux argileux sont habituellement utilisés comme matériau pour la construction de revêtements dans les décharges, afin d'éviter la fuite du lixiviat et la diffusion de contaminants dans les eaux souterraines. Cependant, pour conserver les minéraux argileux et limiter leur utilisation, de nombreux matériaux ont été proposés en remplacement. Cette étude examine la faisabilité d'un nouveau type de revêtement fait à partir de la gangue de charbon, qui est un déchet solide industriel. Une série de tests sont effectués en laboratoire afin d'évaluer ses caractéristiques mécaniques et hydrauliques, ainsi que sa capacité de sorption. Les résultats obtenus montrent que la conductivité hydraulique de la gangue de charbon peut être inférieure à la norme en vigueur de  $1 \times 10^{-7}$  cm/s, pour un revêtement monocouche présentant un taux de vides inférieur à 0.44. Ce nouveau revêtement a également prouvé une remarquable capacité de sorption des ions  $\text{Cd}^{2+}$ . Par conséquent, la gangue de charbon a aussi le potentiel d'être utilisé comme matériau de revêtement pour l'enfouissement ; ce qui permettrait non seulement de conserver les minéraux argileux, mais aussi de recycler un certain type de déchet solide industriel.

**KEYWORDS:** coal gangue; hydraulic conductivity; sorption capacity; initial concentration; soil-to-water ratio.

### 1 INTRODUCTION

A large amount of industrial solid wastes is produced during the exploitation and utilization of mineral resources, including tailings, coal gangue, fly ash, slag, red mud and slag (Cokca and Yilmaz 2004, Zha et al. 2008, Herrmann et al. 2009, Du et al., 2016). According to statistics, the production of industrial solid wastes in China has been increased by 230% from 1.0 billion tons/year in 2003 to 3.3 billion tons/year in 2013. The recycling, disposal and storage of these industrial solid wastes is important for the management and development of mining industry. Among these industrial solid wastes, coal gangue is one of the largest and most harmful wastes generated from the coal production process. Many methods have been proposed for the recycling of coal gangue, such as power generation, agricultural fertilizer, highway roadbeds, brick production, cement production and concrete production (Li et al. 2006, Zhang et al. 2011, Zhou et al. 2014, Wang et al. 2016). However, due to the high increasing rate of coal gangue, the accumulated amount of coal gangue still increases every year and alternative utilization method is required for the recycling of coal gangue.

With the rapid increase in the amount of municipal solid wastes produced every year, landfill has been one of the most important ways for the management of the large amount of wastes, and clay minerals are usually used as liner materials in landfill to prevent the leakage of leachate and the diffusion of contaminants into the groundwater. In recent years, many other materials have been proposed as a substitute of clay minerals in landfill liner construction in order to conserve clay resources (Nhan et al. 1996, Hettiaratchi et al. 1999, Cokca and Yilmaz 2004, Du and Hayashi 2006, Herrmann et al. 2009, Tang et al. 2009, Yang et al. 2012, Yang et al. 2013, Yang 2013, Gong 2014, Xue et al. 2014, Rubinos et al. 2015). Since coal gangue generally contains clay minerals and carbon, it has potential to be used as liner materials in landfill. Yang et al. (2013) examined the mechanical and hydraulic properties of a coal

gangue and the experimental results indicated that the coal gangue could be potential anti-seepage materials for landfill liners. However, although the coal gangue shows remarkable anti seepage characteristics, there has been limited research done on the recycle of coal gangue as liner material, and further research is still needed to understand the adsorption mechanism of heavy metals on coal gangue.

The main objective of this study was to investigate the feasibility of using coal gangue as a substitute of clay minerals for landfill liners. The mechanical, hydraulic and sorption characteristics of a coal gangue were studied through a series of laboratory tests. The anti-seepage and contaminant blocking capability of the coal gangue were then analyzed.

### 2 MATERIALS AND METHODS

The coal gangue used in this study was obtained from Fuxin, Liaoning, China. The basic geotechnical properties of the coal gangue were listed in Table 1. The carbon content of the coal gangue was 7.80%.

Table 1. Basic geotechnical properties of the coal gangue.

Property	Value
Specific gravity	2.56
Liquid limit (%)	42.07
Plastic limit (%)	22.91
Plasticity index (%)	19.16
Maximum dry density ( $\text{g/cm}^3$ )	1.60

Optimum moisture content (%)	17.8
Specific surface area (m <sup>2</sup> /g)	18.89

The compressibility of the coal gangue was analyzed through consolidation test performed in a one-dimensional oedometer 8 cm in diameter according to ASTM standard D2435M-11 (ASTM 2011a). The stepwise vertical stresses were 12.5, 25, 50, 100, 200, 400, 800, and 1600 kPa.

A flexible wall permeameter was used to measure the hydraulic conductivity of the coal gangue under different consolidation pressures (ASTM D5084-10). The coal gangue was first oven dried, then mixed with water at a pre-determined water content of 10% and compacted into the sample mold in five layers with a dry density of 1.60 g/cm<sup>3</sup>, and finally saturated under a vacuum. The specimens for the permeability tests were 10 cm in diameter and 10 cm in height. The consolidation stresses used in the experiment were 50, 100, 200, 400, and 600 kPa.

A series of batch sorption tests were performed to study the sorption capacity of Cd<sup>2+</sup> on the coal gangue. Heavy metal solutions of Cd<sup>2+</sup> were prepared by dissolving a certain mass of CdCl<sub>2</sub> into 1 L deionized water respectively according to the pre-designed C<sub>0</sub>, which was set to be 100, 200, 300, 400 and 500 mg/L. A certain amount of coal gangue was then mixed with 100 ml heavy metal solution and shaking for 24 h at a constant temperature (25 °C). The soil-to-water ratio *s/w* used in this study was 5, 10, 20, 30, 40, 50, 80 and 100 g/L. The equilibrium concentration C<sub>e</sub> was measured and the sorption capacity q<sub>e</sub> (mg/g) and removal efficiency R could be calculated using the following equation,

$$q_e = \frac{(C_0 - C_e) \times V}{m} \quad (1)$$

$$R = \frac{C_0 - C_e}{C_0} \times 100\% \quad (2)$$

where V(L) is the volume of solution and m(g) is the mass of coal gangue.

### 3 RESULTS AND DISCUSSION

#### 3.1 Compressibility

The relationship of void ratio *e* and vertical stress *p* is shown in Fig. 1. The linear relationship between *e* and the logarithm of *p* can be expressed as,

$$e = -0.186 \log(p) + 1.032 \quad (3)$$

According to the above equation, the compressibility index of the coal gangue is 0.186 and the coefficient of compressibility *a*<sub>v1-2</sub> (100 ~ 200kPa) is 1.193 MPa<sup>-1</sup>, indicating that the coal gangue is high compressible.

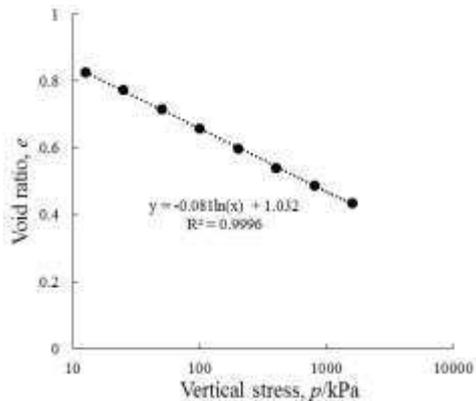


Figure 1. Relationship between void ratio and vertical stress.

#### 3.2 Hydraulic conductivity

Fig. 2 shows the hydraulic conductivity *k<sub>h</sub>* of the coal gangue with different void ratio.

The void ratio of the coal gangue sample consolidated under 50 kPa is about 0.51, and the corresponding hydraulic conductivity was about 1.6 × 10<sup>-7</sup> cm/s. With the increase in the consolidation pressure from 100 to 600 kPa, the void ratio decreased from 0.44 to 0.24, and the hydraulic conductivity decreased from 0.71 × 10<sup>-7</sup> cm/s to 0.24 × 10<sup>-7</sup> cm/s. Regarding the regulatory 1 × 10<sup>-7</sup> cm/s requirement for a monolayer landfill liner, the coal gangue has potential to be used in a liner structure with a void ratio less than 0.44. From Fig. 3, the relationship between void ratio and hydraulic conductivity for the coal gangue can be expressed as,

$$e = 2.686 + 0.32 \log(k_h) \quad (4)$$

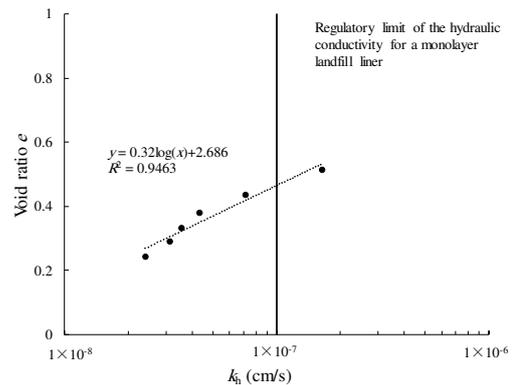


Figure 2. Relationship between void ratio and hydraulic conductivity.

#### 3.3 Sorption characteristics

The sorption isotherms of Cd<sup>2+</sup> on the coal gangue under different *s/w* are shown in Fig. 3. The sorption capacity q<sub>e</sub> increased with the increase in C<sub>0</sub> and decrease in *s/w*, which agreed with the results from previous studies.

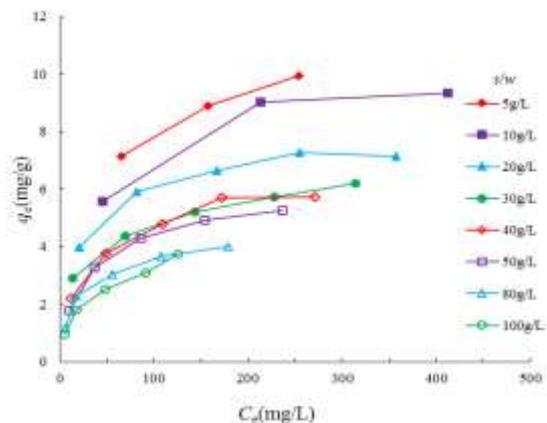


Figure 3. Sorption isotherms of Cd<sup>2+</sup> on the coal gangue.

Several isotherm models have been used to analyze the equilibrium isothermal sorption process of heavy metals on soils, including Langmuir, Freundlich and Dubinin-Radushkevich (D-R) models. These models describe the relationship between the sorption capacity and the equilibrium concentration, and the equations for the three models are given below respectively,

$$\frac{C_e}{q_e} = \frac{1}{k_L Q} + \frac{C_e}{Q} \quad (5)$$

$$\log q_e = \log k_F + \frac{1}{n} \log C_e \quad (6)$$

$$\ln q_e = \ln q_{max} - k\varepsilon^2 \quad (7)$$

where  $Q$  (mg/g) and  $k_L$  (L/mg) are Langmuir constants;  $k_F$  (mg/g) and  $n$  are Freundlich constants;  $q_{max}$  (mg/g) and  $k$  ( $\text{mol}^2/\text{kJ}^2$ ) are D-R constants;  $Q$  and  $q_{max}$  both represent the maximum sorption capacity;  $\varepsilon$  is the Polanyi potential which can be expressed as,

$$\varepsilon = RT \ln \left( 1 + \frac{1}{C_e} \right) \quad (8)$$

in which  $R$  (J/(mol·K)) is the ideal gas constant;  $T$  (K) is the absolute temperature. From the D-R model, the free energy of sorption  $E$  (kJ/mol) can be calculated with the following equation,

$$E = -\frac{1}{\sqrt{2k}} \quad (9)$$

The value of  $E$  actually indicates different sorption types: the chemical sorption dominates when the absolute value is larger than 16 kJ/mol, the ion exchange dominates when the absolute value is between 8 and 16 kJ/mol, and the physical sorption dominates when the absolute value is in the range from 1 to 8 kJ/mol.

Table 2. Fitting results of the sorption isotherms.

$s/w$ (mg/g)	Langmuir model			
	$Q_m$ (mg/g)	$k_L$ (L/mg)	$R^2$	
5	11.3	0.03	0.99	
10	10.5	0.03	0.99	
20	7.5	0.05	0.99	
30	6.1	0.07	0.99	
40	5.8	0.05	0.98	
50	5.5	0.05	0.99	
80	4.1	0.07	0.99	
100	3.4	0.08	0.98	
$s/w$ (mg/g)	Freundlich model			
	$k_F$ (mg/g)	$n$	$R^2$	
5	2.56	4.07	0.99	
10	2.21	4.03	0.95	
20	2.15	4.63	0.95	
30	1.56	4.16	0.99	
40	1.06	3.16	0.98	
50	0.85	2.86	0.97	
80	0.73	2.90	0.95	
100	0.52	2.46	0.99	
$s/w$ (mg/g)	D-R model			
	$q_{max}$ (mg/g)	$k$ ( $\text{mol}^2/\text{kJ}^2$ )	$E$ (kJ/mol)	$R^2$
5	19.4	0.003	-13.1	1
10	17.5	0.003	-12.9	0.96
20	12.5	0.003	-14.1	0.97
30	10.6	0.003	-13.9	0.99
40	13.0	0.004	-12.1	0.99
50	13.3	0.004	-11.6	0.99
80	10.3	0.004	-11.9	0.97
100	11.0	0.004	-11.3	0.99

30	10.6	0.003	-13.9	0.99
40	13.0	0.004	-12.1	0.99
50	13.3	0.004	-11.6	0.99
80	10.3	0.004	-11.9	0.97
100	11.0	0.004	-11.3	0.99

In order to uniformly illustrate the impact of  $s/w$  and  $C_0$ , a new factor  $a/s$  defined as the ratio between  $C_0$  and  $s/w$  is used to analyze the sorption behavior of  $\text{Cd}^{2+}$  on the coal gangue. According to the value of  $C_0$  and  $s/w$ , the value of  $a/s$  is 1 ~ 500 mg/g. The relationships between  $a/s$  and  $q_e$ ,  $R$  are displayed in Fig. 6 and Fig. 7 respectively. The different curves in Fig. 4 and Fig. 5 could be uniformly described by one curve in Fig. 6 and Fig. 7 if the horizontal ordinate was replaced by  $a/s$ . With the increase in  $a/s$ , the sorption capacity increased and the removal efficiency decreased. The increase in  $a/s$  can be induced by the increase in  $C_0$  or the decrease in  $s/w$ , therefore the effect of the  $C_0$  and  $s/w$  are uniformly illustrated by  $a/s$ . Based on the above analysis, the new factor  $a/s$  may be used as a more essential experiment condition for both sorption experiment instead of  $C_0$  and  $s/w$ .

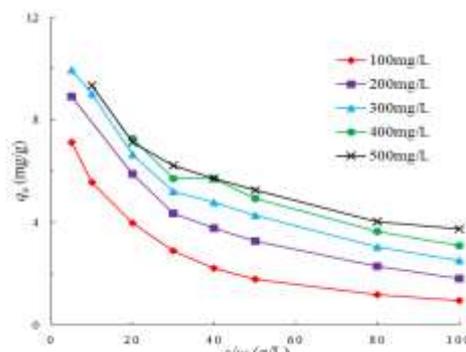


Figure 4. Effect of soil-to-water ratio on the sorption capacity of  $\text{Cd}^{2+}$ .

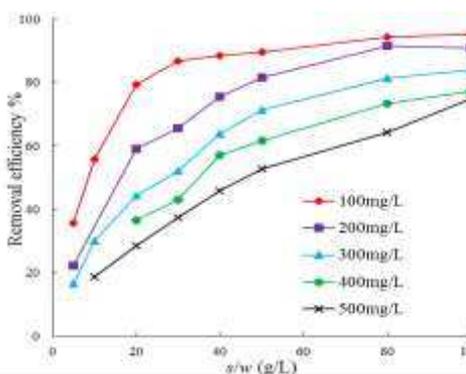


Figure 5. Effect of soil-to-water ratio on the removal efficiency of  $\text{Cd}^{2+}$ .

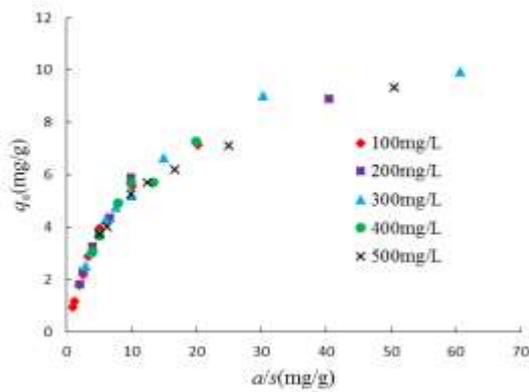


Figure 6. Effect of  $a/s$  on the sorption capacity of  $Cd^{2+}$ .

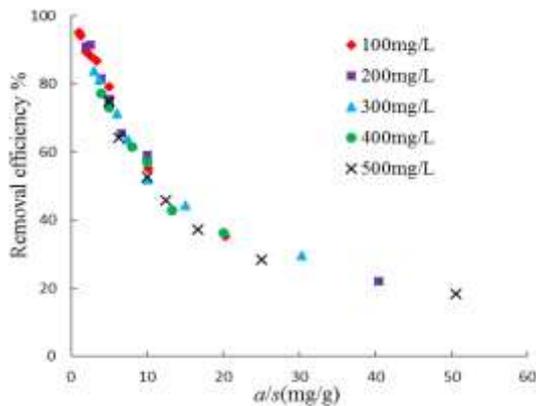


Figure 7. Effect of  $a/s$  on the removal efficiency of  $Cd^{2+}$ .

#### 4 CONCLUSIONS

In the present study, a series of laboratory experiments were performed to study the feasibility of using coal gangue as landfill liner material in terms of mechanical, hydraulic and sorption characteristics. The following conclusions can be drawn.

The hydraulic conductivity of the coal gangue can be less than the regulatory  $1 \times 10^{-7}$  cm/s requirement for a monolayer landfill liner with a void ratio smaller than 0.44.

The effect of initial concentration and soil-to-water ratio on the sorption characteristics of  $Cd^{2+}$  on the coal gangue can be uniformly illustrated by the new factor defined as the ratio between the initial concentration and soil-to-water ratio. The coal gangue showed remarkable sorption capacity on  $Cd^{2+}$ , and the desirable hydraulic and sorption properties indicated that the coal gangue has potential to be used as landfill liner materials.

#### 5 ACKNOWLEDGEMENTS

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