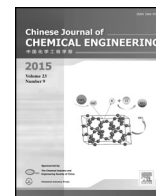




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Adsorption characteristics of construction waste for heavy metals from urban stormwater runoff☆



Jianlong Wang *, Pingping Zhang, Liqiong Yang, Tao Huang

Key Laboratory of Urban Stormwater System and Water Environment, Beijing University of Civil Engineering and Architecture, Ministry of Education, Beijing 100044, China

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ABSTRACT

Stormwater runoff has become an important source of surface water pollution. Bioretention, a low impact development measure in urban stormwater management, has been proven to be effective in the removal of pollutants from stormwater runoff, with appropriate bioretention media. In this study, construction wastes were selected as bioretention media to remove heavy metals from stormwater runoff. Static and dynamic adsorption batch experiments were carried out to investigate the adsorption of heavy metals in simulated stormwater runoff system with construction wastes in different particle sizes. The experimental results show that the pseudo-second-order kinetic model characterizes the adsorption process and the adsorption equilibrium data are well described by Freundlich isotherm model. The construction wastes used can remove heavy metals from stormwater runoff effectively, with their average removal rates all more than 90%. The particle size of construction wastes greatly influences the equilibrium time, rate and adsorption capacity for heavy metals.

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1. Introduction

With the rapid development of urbanization, increasing impervious areas interrupt stormwater infiltration channels and greatly increase stormwater runoff volume and peak flow. In addition, due to human activities, atmospheric deposition and other factors, a large number of pollutants accumulate and are discharged into the municipal stormwater sewer by stormwater runoff flushing, then enter receiving watershed. With increasing perfection of point pollution control in the urban, non-point pollution caused by stormwater runoff becomes an important source of urban watershed pollution. Researches on water quality of urban stormwater runoff show that heavy metals from stormwater runoff are important sources of surface water pollutants [1].

Heavy metals in urban stormwater runoff are mainly from vehicle exhaust, industrial soot, fossil fuel burning, sandstorm dust and corrosion of various metal facilities, and heavy metals copper, zinc, and lead are common constituents in highway runoff, usually at relatively high concentrations. Different from organic pollutants, heavy metals are difficult to be degraded in environment and easy to accumulate in

human body through food chain and other ways. Excessive intake of heavy metals in human body may irritate mucous membranes, leading to hepatic and renal damage, capillary damage, and central nervous problems [2]. A major finding of the Nationwide Urban Runoff Program was that metals (especially Cu, Pb, and Zn) are the most prevalent constituents found in urban runoff land uses such as transportation [3]. Then some developed countries started to study heavy metal pollution and its impact factors [4,5]. Bioretention technology (also known as rain garden), as one of the low impact development stormwater management measures, can not only control water quality efficiently (with removal rate of heavy metals over 90%) [6], but also has ecological function and landscape effect. It can be widely used in road green belt to control open space stormwater runoff.

Bioretention is a simulated ecological measure. It is in low-lying areas. Plants are planted to purify the first flush runoff from small watershed by replacing natural or artificial soils and reduce the runoff volume. It is mainly composed of gravel layer, filler layer, planting soil layer, covering layer, plants, and overflow device, with natural infiltration or drainage system at the bottom. Stormwater enters a bioretention unit and is purified by physical filtration, adsorption and ion exchange of soil, microbial transformation and redistribution, evaporation and absorption of plants in soil layer and filler layer [7]. Purified stormwater can supplement groundwater, or transport to municipal systems or subsequent treatment facilities through a device at the bottom of perforated collection tube. Bioretention medium has a significant effect on the removal of heavy metals (Table 1). Therefore, selection of effective and economical bioretention medium is of great importance for wide applications of bioretention technology.

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* Corresponding author.

E-mail address: wjl_xt@163.com (J. Wang).

Table 1
Removal rate of heavy metals with different bioretention media

Type	Removal rate/%						Reference
	Cu ²⁺	Zn ²⁺	Pb ²⁺	Cd ²⁺	Cr ³⁺	Ni ²⁺	
Sandy-loam	>90	>90	>90	–	–	–	[8]
Zeolite	53.4	41.8	89.2	45.0	–	–	[9]
Activated carbon	>99	>99	>99	–	>90	>90	[10]
Pinecone	55.32	39.96	86	–	–	–	[11]
Peanut shell	62	–	98	26	34	14	[12]

Construction waste consists of dregs, waste soil, waste material, general waste, and wastes from construction and repairing of buildings and structures, and pipe network construction, laying and removal [13]. Construction waste is hard and resistant to wear, frost, and water. It is chemically stable and its performance is better than that of clay, silt and lime soil. Waste bricks have a large specific surface area and micropore volume and their surfaces have a rough and porous structure. They have been used as substrates for constructed wetland, with good adsorbency and efficacy in phosphorus removal. Waste iron slag, a typical byproduct of steel production, is inexpensive and widely available. It contains a large amount of iron oxides, which are efficient in arsenic removal [14]. Therefore, construction waste may be used as adsorbent for removal of heavy metals from urban stormwater runoff.

In this study, construction wastes with different particle sizes are used for removing heavy metals (Cu, Pb, Cd) in stormwater runoff. With static and dynamic adsorption experiments, the feasibility of using construction wastes as bioretention media is discussed. This study could provide a scientific basis for effectively controlling heavy metal pollutants from stormwater runoff with construction wastes.

2. Materials and methods

2.1. Construction wastes

The construction wastes used in the experiment were mainly composed of brick, mixed with a small amount of concrete. After crush with a crusher, the wastes were sieved to achieve four different particle sizes, 2.36–4.75, 4.75–10.0, 2.36–10.0, and 0–10.0 mm. Particles were soaked and washed with distilled water to remove surface residuals, dried in an oven at 105 °C for 24 h and stored in the grinding jars until use.

The chemical oxide composition of construction wastes was determined by a Scanning Wavelength Dispersive X-ray fluorescence using a Rigaku ZSX Primus II spectrometer. BET surface area was measured by automatic adsorption using a NOVE 4200E instrument, which is 2.2694 m²·g^{−1}, and the porosity factor is 15%. Chemical compositions of fresh construction wastes are shown in Table 2. The main components are silicon, calcium and aluminum oxides, with 42.5%, 14.2% and 12.0%, respectively, similar to the results of Wang *et al.* [15] and Xie *et al.* [16].

2.2. Batch adsorption experiment

According to the initial concentration of heavy metals in urban stormwater runoff [17,18], 0.5 mg·L^{−1} metal stock solution was prepared using 1 mg·μl^{−1} heavy metal standard solution provided by Analysis Center of the National Non-ferrous Metals and Electronic Materials. Then the pH of the stock solution was adjusted in the range 6.8 to 7.5, consistent with the average pH value of urban stormwater runoff [19]. The experiment was carried out at room temperature of (25 ± 1)°C.

Table 2
Chemical composition and physical characteristics of construction waste

Constituent	Composition/%
Na ₂ O	0.931
MgO	2.58
Al ₂ O ₃	12.0
SiO ₂	42.5
P ₂ O ₅	0.146
K ₂ O	2.33
CaO	14.2
TiO ₂	0.647
Cr ₂ O ₃	0.0174
MnO	0.0939
Fe ₂ O ₃	4.75
CuO	0.0042
ZnO	0.0145
Others	0.2117

2.2.1. Static adsorption experiment

100 g construction wastes with different particle sizes were weighed and transferred into a series of 250 ml triangle conical flasks, added with 150 ml stock solution, and placed at a temperature-regulated orbital platform shaker at 80 r·min^{−1}. Shaking was temporarily interrupted at 1, 3, 5, 10, 20, 30, 40, 60, 90, 120, 180, 240 and 300 min to determine the influence of reaction time on adsorption equilibrium. Under the optimum equilibrium time, 5, 10, 20, 30, 50, 70 and 100 g of construction wastes with different particle sizes were used to study the isothermal adsorption process. 50 ml samples were subsequently filtered through a 0.45 μm hydrophilic membranes prior to analysis, and the filtrate were stored in polyvinyl chloride (PVC) bottles and kept at 4 °C in the refrigerator.

2.2.2. Dynamic adsorption experiment

The construction wastes were added into experimental device. The dynamic adsorption experiment was processed with continuous water input of 4 ml·s^{−1} by a peristaltic pump. 50 ml samples were collected in sequence at 3, 5, 10, 20, 30, 60, 90, 180 and 300 min. The samples were filtered through 0.45 μm hydrophilic membranes to remove suspended matters and stored in PVC bottles. The samples were kept at 4 °C in the refrigerator for further analysis. Experimental device used for dynamic adsorption experiment is shown in Fig. 1.

2.3. Adsorption isotherm and kinetic model

2.3.1. Adsorption isotherm model

At constant temperature, sorption between solid and liquid is often described by Langmuir and Freundlich isotherm models. The equation of Langmuir adsorption isotherm can be expressed as [20]

$$\frac{C_e}{Q_e} = \frac{C_e}{Q_m} + \frac{1}{K_a Q_m} \quad (1)$$

where Q_e is the equilibrium absorption capacity (mg·g^{−1}), Q_m is the saturated adsorption capacity (mg·g^{−1}), C_e is the concentration of heavy metals at absorption equilibrium (mg·L^{−1}), and K_a is the Langmuir adsorption constant.

Freundlich isotherm model is an empirical equation based on experimental observations, assuming that the adsorption is on a complex surface. The Freundlich adsorption isotherm can be expressed as [21]

$$\lg Q_e = \frac{1}{n} \lg C_e + \lg K_f \quad (2)$$

where K_f is the Freundlich adsorption constant and n is a constant.

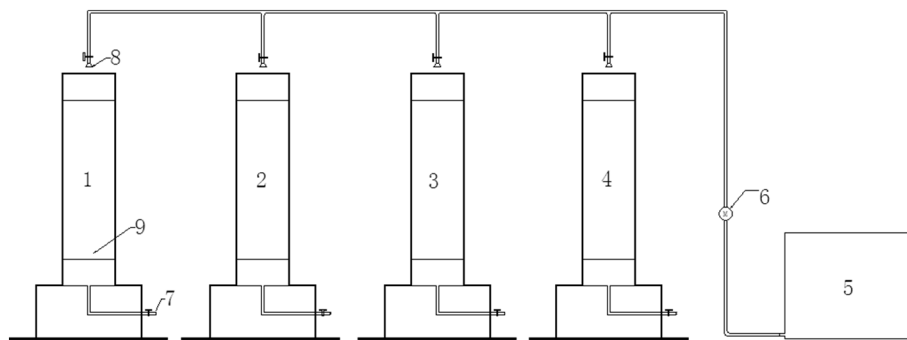


Fig. 1. Experimental device for dynamic adsorption experiment. 1—(2.36–4.75) mm construction waste; 2—(4.75–10.0) mm construction waste; 3—(2.36–10.0) mm construction waste; 4— ≤ 10.0 mm construction waste; 5—water tank; 6—peristaltic pump; 7—outlet; 8—inlet; 9—sieve plate.

2.3.2. Adsorption kinetic model

Heavy metal adsorption from stormwater runoff on the construction wastes was analyzed by pseudo-first and pseudo-second-order kinetic models. The pseudo-first-order kinetic equation is [22]

$$\lg(Q_e - Q_t) = \lg Q_e - k_1 t \quad (3)$$

where Q_t is the absorption capacity at time t ($\text{mg} \cdot \text{g}^{-1}$) and k_1 is the equilibrium rate constant. k_1 can be determined by a linear plot of $\lg(Q_e - Q_t)$ vs. t .

It is found that adsorption kinetics of heavy metals fits better to the pseudo-second-order kinetic equation [23]:

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{t}{Q_e} \quad (4)$$

where k_2 is the equilibrium rate constant.

2.4. Aqueous sample analysis

Aqueous samples were analyzed by using a Hitachi Z-2010 Polarized Zeeman atomic absorption spectrophotometer. Calibration curves were based upon the analysis of $1 \text{ mg} \cdot \text{L}^{-1}$ heavy metal standard solution from the Analysis Center of the National Non-ferrous Metals and Electronic Materials. Cu, Pb and Cd standard solutions of 20.00, 20.00 and $3.00 \mu\text{g} \cdot \text{L}^{-1}$, respectively, were diluted by 2% (by mass) saltpeter solution. Cu and Pb calibration curves were obtained by absorbance values of 0.00, 5.00, 10.00, 15.00 and $20.00 \mu\text{g} \cdot \text{L}^{-1}$ standard stock solutions. Cd calibration curve was obtained by absorbance values of 0.00, 0.50, 1.00, 2.00 and $3.00 \mu\text{g} \cdot \text{L}^{-1}$ standard stock solutions. The correlation coefficient of Cu, Pb and Cd is 0.9997, 0.9992 and 0.9991, respectively. Concentrations of heavy metals in the solution were determined on the construction waste after adsorption, and those in reserve solutions were decided as control at each group. The efficiency of heavy metal adsorption on construction wastes was determined from the heavy-metal concentrations in the solution before and after adsorption.

The adsorption capacity and removal rate (η) are calculated as follows.

$$Q = \frac{V(C_0 - C_t)}{m} \quad (5)$$

$$\eta = \frac{C_0 - C_t}{C_0} \times 100\% \quad (6)$$

where Q is the adsorption capacity of construction wastes ($\text{mg} \cdot \text{g}^{-1}$), C_0 and C_t are the heavy-metal concentrations in the solution before and after adsorption ($\text{mg} \cdot \text{L}^{-1}$), respectively, V is the volume of sample (L), and m is mass of construction waste (g).

3. Results and discussion

3.1. Permeability of construction wastes

Table 3 gives the permeability coefficients of construction wastes measured by the hydrostatic head method. With the particle sizes of 0–10.0, 2.36–10.0, 2.36–4.75, and 4.75–10.0, the average permeability coefficients of construction wastes are 1.24×10^{-5} , 1.53×10^{-3} , 1.93×10^{-3} and $6.38 \times 10^{-3} \text{ m} \cdot \text{s}^{-1}$, respectively. Larger diameter gives better infiltration of stormwater, so the hydraulic loading shock resistance ability will be higher. The use of construction waste with larger particle size in bioretention will reduce the runoff volume and the peak flow may be more appropriate.

Table 3

Permeability coefficients of construction wastes with different size distributions

Size/mm	$k_1/\text{m} \cdot \text{s}^{-1}$	$k_2/\text{m} \cdot \text{s}^{-1}$
0–10.0	1.29×10^{-5}	1.19×10^{-5}
2.36–10.0	1.56×10^{-3}	1.50×10^{-3}
2.36–4.75	1.98×10^{-3}	1.88×10^{-3}
4.75–10.0	6.54×10^{-3}	6.22×10^{-3}

Notes: k_1 – 0.5 h; k_2 – 1.5 h.

3.2. Composition of construction wastes before and after adsorption

Fig. 2 shows scanning electron microscopy (SEM) micrographs for the surface of construction wastes before and after adsorption. The surface is smoother after adsorption, probably due to the deposition of heavy metals by physic-sorption or a progressive change in surface mineralogy. Fig. 3 shows the component concentration in construction wastes before and after adsorption, with Al decreased slightly from 6.61% to 6.00% and Ca decreased from 10.8% to 10.3%, while Mg increased significantly from 1.6% to 2.39% and Si increased from 20.8% to 22.7%. The decrease in Ca content may be related to the loss of CaO, which often results in higher pH [24]. Therefore, the change in pH of stormwater should be taken into account before the discharge. The contents of As and Cr, which are harmful to the environment and human body, change little after adsorption, with As decreased from 0.0023% to 0.001% and Cr decreased from 0.0128% to 0.0125%. The increased content of Cu after adsorption and the smoother surface of construction wastes showed that Cu was removed by adsorption. The results show that there are no detrimental effects to human health or to the environment with the application of construction wastes in stormwater runoff purification.

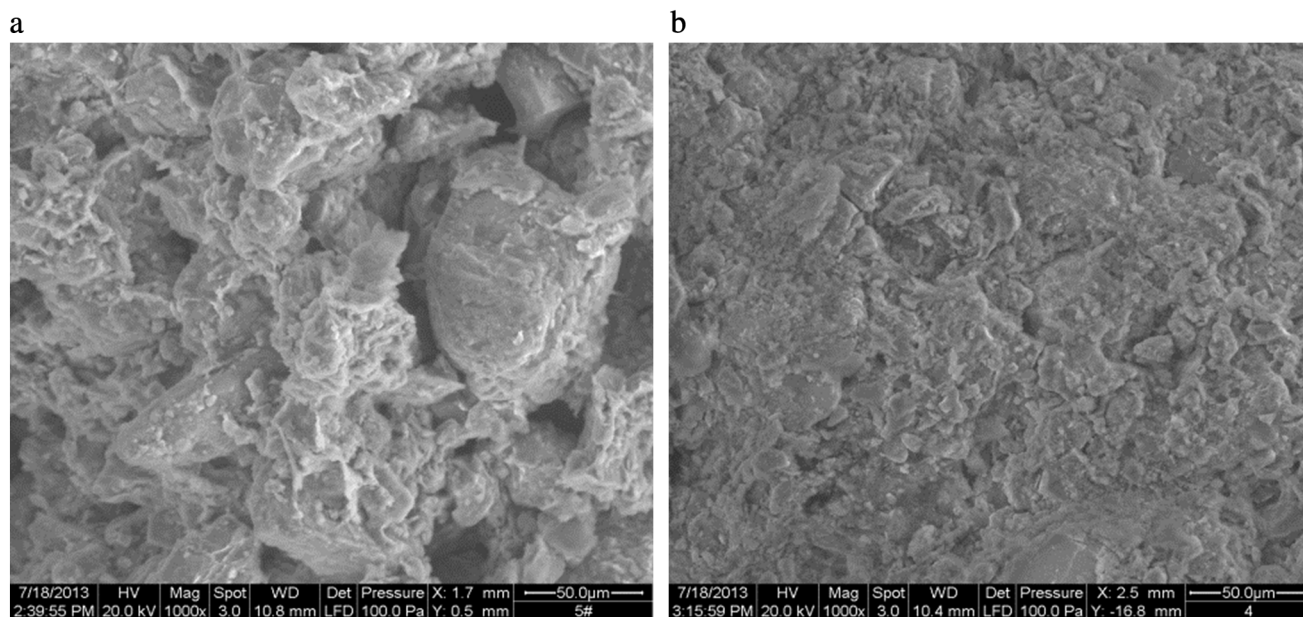


Fig. 2. SEM images of construction waste before (a) and after (b) adsorption.

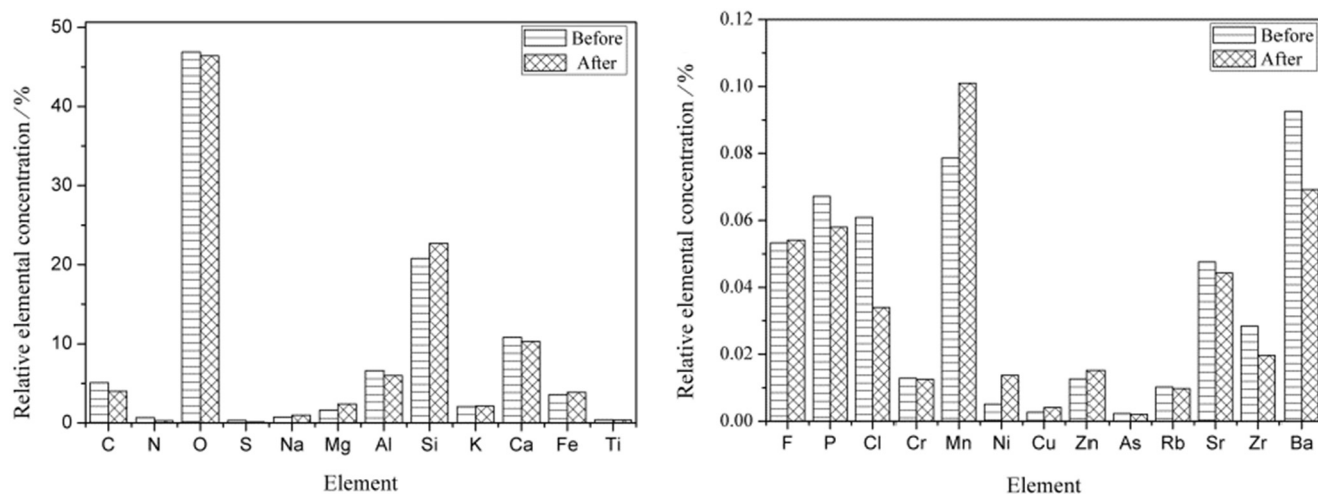


Fig. 3. Relative concentrations of mineral elements before and after adsorption.

3.3. Adsorption isotherms

For static adsorption of Cu, Pb and Cd on construction wastes with different particle sizes, Table 4 gives the Langmuir and Freundlich

adsorption isotherm parameters. The correlation coefficients are higher than 0.9000 except for Cd on the particle of 4.75–10.0 mm. Comparison of correlation coefficients shows that the Freundlich isotherm is better than the Langmuir isotherm. This is consistent with some previous

Table 4
Parameters of Langmuir and Freundlich equations

Particle size/mm		Langmuir adsorption constants			Freundlich adsorption constants		
		$Q_m/\text{mg}\cdot\text{g}^{-1}$	K_a	R^2	$1/n$	K_f	R^2
2.36–4.75	Cu	-2.262 ± 0.024	-1.193 ± 0.024	0.7283	0.785 ± 0.24	0.181 ± 0.053	0.9207
	Pb	0.0017 ± 0.0007	317.27 ± 2.78	0.9343	1.402 ± 0.52	37.58 ± 1.58	0.9936
	Cd	-2.435 ± 0.059	-3.933 ± 0.72	0.9588	1.065 ± 0.55	0.106 ± 0.031	0.9736
4.75–10	Cu	-0.720 ± 0.015	-2.258 ± 0.025	0.6917	0.234 ± 0.32	0.025 ± 0.193	0.9715
	Pb	-0.0076 ± 0.022	-20.07 ± 1.27	0.6442	0.178 ± 0.57	0.016 ± 0.49	0.9748
	Cd	1.874 ± 0.063	-10.870 ± 2.01	0.7948	2.867 ± 0.55	1.109 ± 0.16	0.8954
2.36–10	Cu	-1.423 ± 0.032	-1.468 ± 0.045	0.7827	1.369 ± 1.02	2.020 ± 1.89	0.9076
	Pb	-0.00044 ± 0.0009	-269.62 ± 1.35	0.7557	0.713 ± 0.63	0.79 ± 0.092	0.9933
	Cd	-3.821 ± 0.89	-1.822 ± 0.44	0.8101	0.742 ± 0.067	0.054 ± 0.18	0.9701
0–10	Cu	-0.803 ± 0.034	-1.374 ± 0.011	0.8393	1.806 ± 1.21	20.179 ± 19.98	0.9468
	Pb	-0.000033 ± 0.000024	-720.93 ± 5.48	0.6610	0.456 ± 0.34	0.24 ± 0.12	0.9963
	Cd	-2.434 ± 0.013	-2.327 ± 1.03	0.6501	0.641 ± 0.26	0.045 ± 0.56	0.9824

results, such as Cu adsorption on soils [11] adsorption of Cu on pine-cone [25], and adsorption of Pb on natural zeolite [26–28], while heavy-metal adsorption on other adsorbents follows the Langmuir isotherm equation [27]. The K_f value in the Freundlich equation is a typical index for capacity of adsorbents for heavy-metals. Table 4 shows that the adsorption capacity of construction waste is related to particle size.

3.4. Adsorption kinetics

The adsorption kinetic profiles of heavy metals on construction wastes with different particle sizes are shown in Fig. 4 and the kinetic model parameters are given in Table 5. The adsorption fits the pseudo-second-order kinetic model well. The equilibrium adsorption capacity calculated with the kinetic equation is consistent with experimental data. The chronological order to reach adsorption equilibrium is consistent with the order of k_2 . However, the adsorption process and adsorption mechanism of different adsorbents are different for different heavy metals. Covelo *et al.* studied the heavy metal adsorption by humic umbrisols and showed that pseudo-first-order kinetic model fitted to Cu adsorption better [29]. Investigations for adsorption of heavy metals on *saccharomyces cerevisiae* [30], natural zeolite [31], blast furnace slag [32], and hazelnut shell [33] showed that the sorption

Table 5
Parameters in the pseudo-second-order kinetic model for heavy-metal adsorption on construction wastes

Particle size/mm		$k_2/\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1}$	$Q_e/\text{mg}\cdot\text{g}^{-1}$	R^2	Dongle kinetic model
2.36–4.75	Cu	121.52	0.0125	0.9999	$Q_t = \frac{0.0189t}{1+1.517t}$
	Pb	3099.50	0.0055	0.9999	$Q_t = \frac{0.0936t}{1+17.035t}$
	Cd	13.394	0.0125	0.9993	$Q_t = \frac{0.00209t}{1+0.167t}$
4.75–10	Cu	16.197	0.0082	0.9985	$Q_t = \frac{0.00110t}{1+0.133t}$
	Pb	87.48	0.0036	0.9996	$Q_t = \frac{0.00113t}{1+0.315t}$
	Cd	10.073	0.0046	0.9837	$Q_t = \frac{0.000216t}{1+0.0467t}$
2.36–10	Cu	46.574	0.0101	0.9998	$Q_t = \frac{0.00475t}{1+0.470t}$
	Pb	132.81	0.0034	0.9987	$Q_t = \frac{0.00155t}{1+0.453t}$
	Cd	6.222	0.0058	0.9828	$Q_t = \frac{0.000210t}{1+0.0361t}$

kinetics could be better described by pseudo-second-order kinetics model.

3.5. Effect of particle size on heavy metal removal

3.5.1. Adsorption capacity

It is shown that particle size of sorbent influences the adsorption of heavy metals [34]. Adsorption capacities of construction

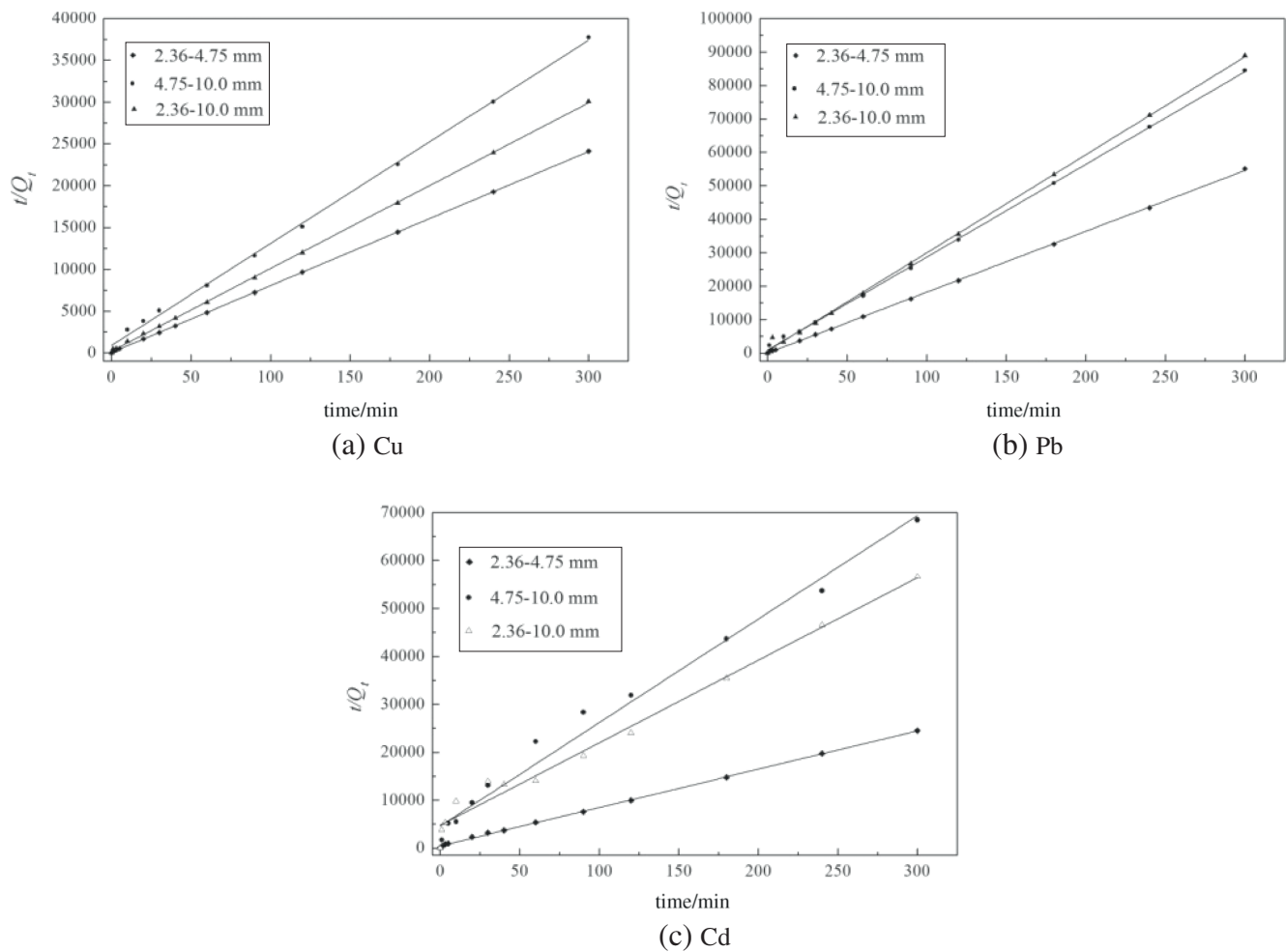


Fig. 4. Curves for pseudo-second-order kinetics of heavy-metal adsorption on construction wastes.

wastes with different particle sizes are shown in Fig. 5. The adsorption capacity increases dramatically with time until reaching a plateau. The time for reaching equilibrium is the shortest for Cu and Pb on construction wastes with particles of 0–10.0 mm, 10 and 5 min, respectively. The equilibrium time on Cu is shorter compared with previous studies in soils [35]. The equilibrium time on Cd is longer, so that construction wastes have a stronger affinity on Cu and Pb than Cd. The equilibrium adsorption capacities of Cu, Pb and Cd on construction wastes of 2.36–4.75 mm is the maximum, while those on particles of 0–10.0 mm and 4.75–10.0 mm are smaller. Generally, smaller particles present larger surface area and better adsorption performance, so the adsorption efficiency will be higher [36]. The present experiment gives a similar conclusion.

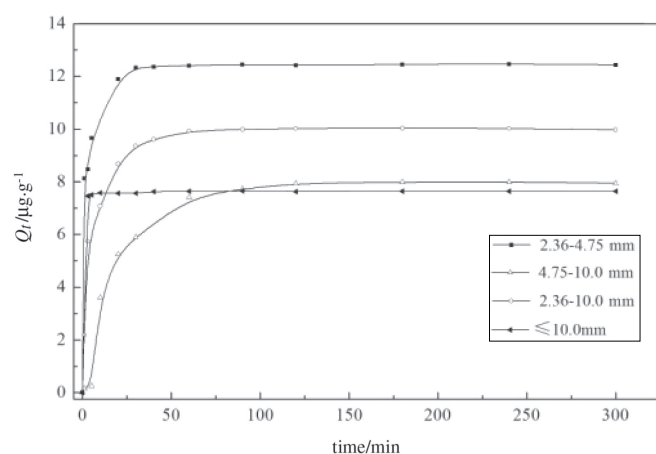
3.5.2. Adsorption rate

Adsorption rates of heavy metals on construction wastes with different particle sizes are shown in Fig. 6. The variations of adsorption rates with construction waste particles of 2.36–4.75 mm and 0–10.0 mm for Cu, Pb and Cd are similar, decreasing first and then becoming steady. Differently, the adsorption rate of Cu on

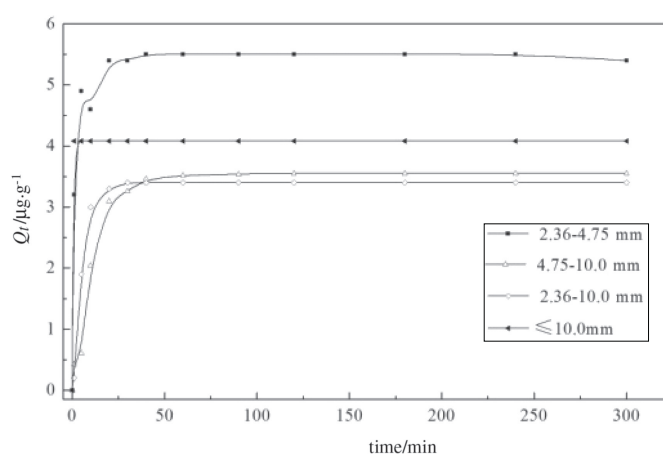
construction waste of 4.75–10.0 mm first increases and then decreases until reaching a plateau. For Pb, the adsorption rate on particles of 2.36–10.0 mm increases first and then decreases, and that on 4.75–10.0 mm particles first decreases, then increases, and tends to be steady. Stable adsorption rates of Cu, Pb and Cd are different with different particle sizes, but their largest stable adsorption rates all occur in 2.36–4.75 mm particles. The results show that the change of stable adsorption rates of heavy metals is consistent with the variation of adsorption capacity. Therefore, construction wastes with larger adsorption capacity and better adsorption rate for heavy metals can be used as bioretention media to purify urban runoff stormwater.

3.6. Removal of heavy metals from stormwater runoff with construction wastes

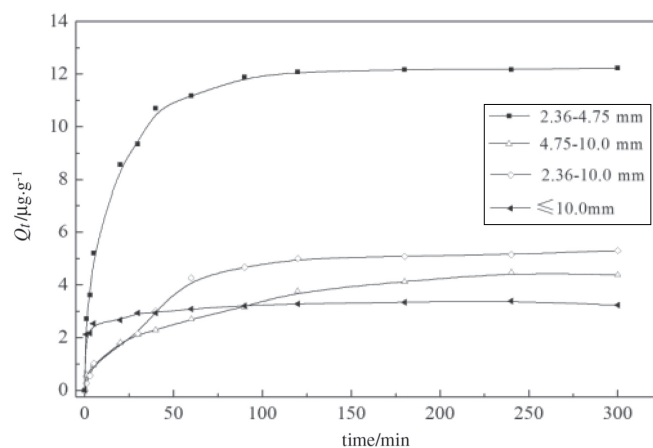
Fig. 7 shows the removal rate of heavy metals by dynamic experiments. The removal rates of Cu, Pb and Cd with construction wastes in different particle sizes increase gradually with time. The curves of equilibrium time for removing Cu, Pb and Cd are similar, increasing rapidly and then being stable. The removal sequence for Cu is construction



(a) Cu



(b) Pb



(c) Cd

Fig. 5. Heavy-metal adsorption capacity of construction wastes with different particle sizes.

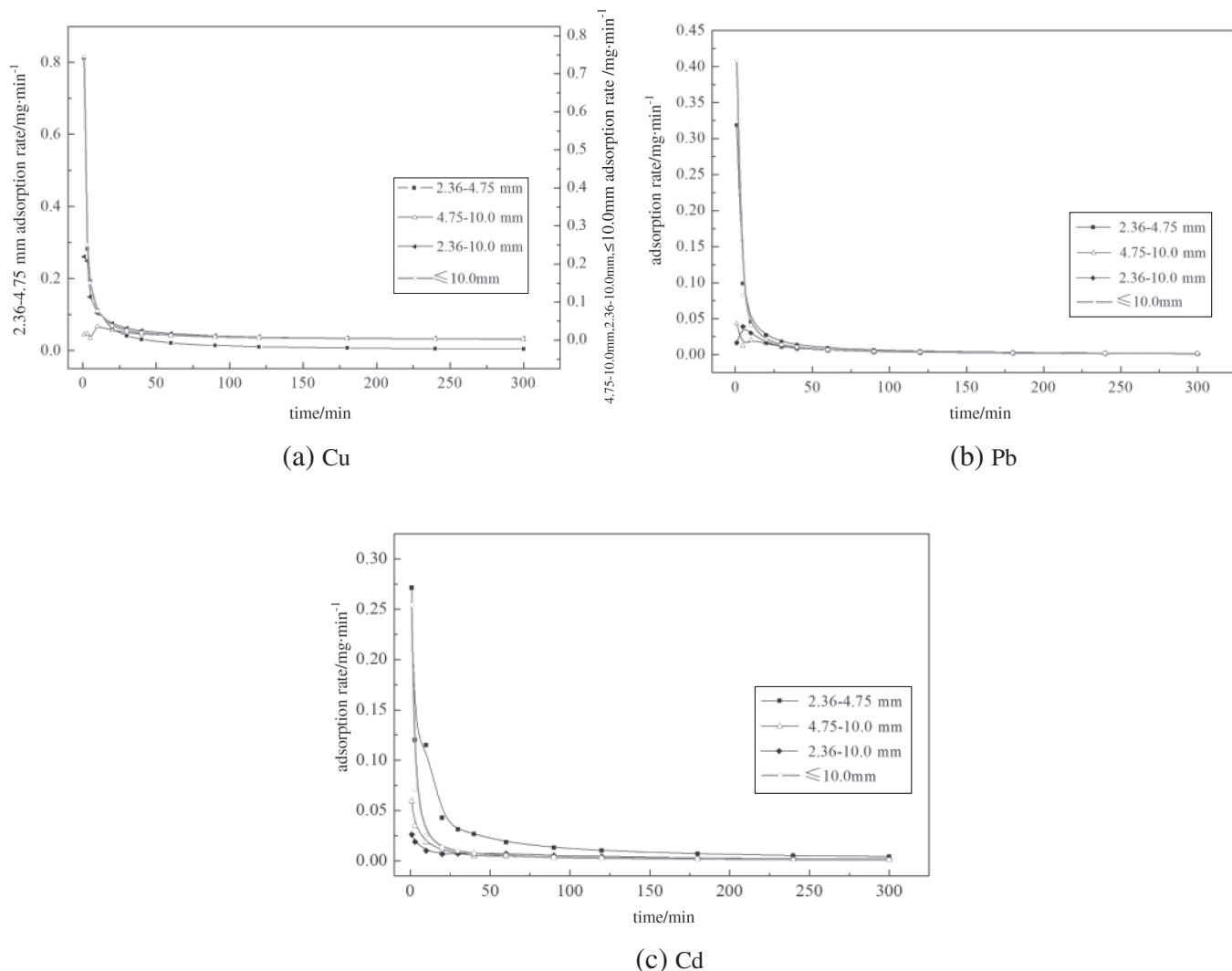


Fig. 6. Heavy-metal adsorption rates of construction wastes with different particle sizes.

wastes $2.36-10.0\text{ mm} > 2.36-4.75\text{ mm} > 0-10.0\text{ mm} > 4.75-10.0\text{ mm}$. The equilibrium time for removing Pb is all within 10 min and the removal rates are all close to 100%. The removal sequence for Cd is construction wastes $0-10.0\text{ mm} > 2.36-10.0\text{ mm} > 2.36-4.75\text{ mm} > 4.75-10.0\text{ mm}$.

Dynamic and static experimental results show that the construction waste presents good purification efficiency for heavy metals from stormwater runoff and the average removal rates are all more than 90%. Therefore, the construction waste can be used as bioretention medium to effectively remove heavy metal pollutants from urban stormwater runoff.

Some studies show that the main factors influencing heavy-metal adsorption on solids are contact time, adsorbent dose, initial concentration of solution, pH value, and temperature. The removal rate by adsorption increases gradually with contact time until reaching equilibrium. Decrease of removal rate with time is minimal [36]. Larger adsorbent dosage also increases the removal rate of heavy metals. The pH is an important factor in heavy-metal adsorption. The best pH value for adsorption varies with adsorbent [37,38]. In applications, rainfall duration, depth of bioretention media, pH of runoff stormwater, and heavy metal concentration influence the removal efficiency of heavy metals from

stormwater runoff, so design parameters should be optimized for these factors.

4. Conclusions

Static adsorption isotherms show that adsorption of heavy metals by construction wastes accords with the Freundlich isotherm adsorption equation and the adsorption kinetics is consistent with the pseudo-second-order dynamics equation.

The dynamic experimental results show that construction wastes with different particle sizes all have good adsorption and removal effect on heavy metals, with the average removal rates more than 90%. Large particle size gives better purification efficiency for heavy metals.

In practice, construction wastes with different particle sizes that have better permeability and higher removal rate for heavy metals can be used as the adsorption medium of ecological measures such as bioretention facilities and constructed wetland. It can control heavy metal pollutants from urban stormwater runoff effectively and provide a new way for resource utilization of city construction waste.

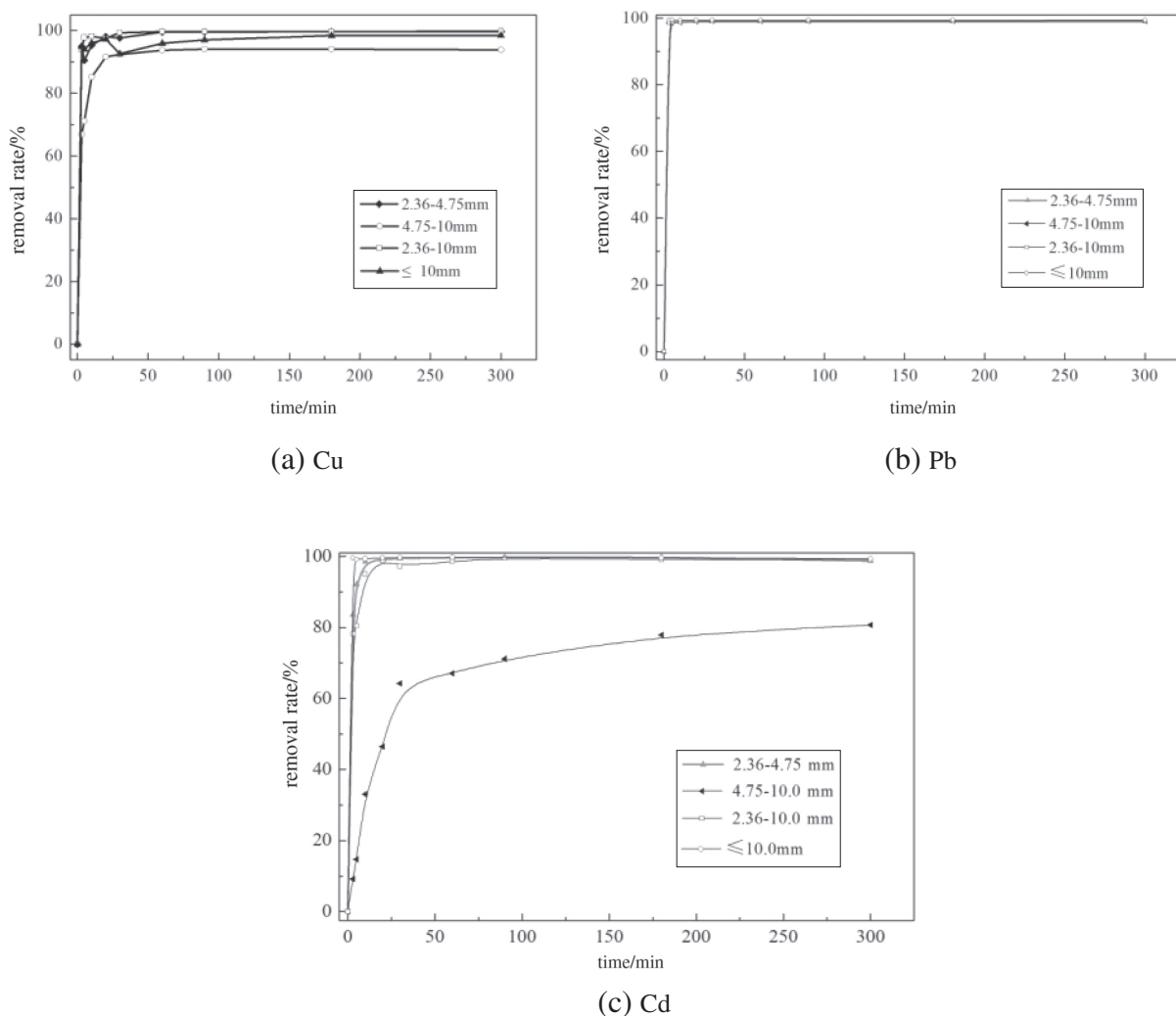


Fig. 7. Removal rate of construction wastes with different particle sizes for heavy metal adsorption.

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