Adsorption characteristics of Pb from urban stormwater runoff by construction wastes

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Abstract: Construction wastes were selected as the adsorbents, and static and dynamic adsorption batch experiments were carried out to investigate the adsorption of Pb to construction wastes with different particle size gradations in the simulated stormwater runoff system. The experimental results show that the pseudo-second-order kinetics model can better characterize the adsorption process of Pb than the pseudo-first-order kinetics model. The adsorption equilibrium data can be well fitted by the Freundlich isotherm model. The construction wastes with different tested size gradations can greatly remove Pb from stormwater runoff and their average removal rate can reach up to 99%. The construction wastes with narrow size distribution can better remove Pb but with worse permeability than those with wide size distribution. The particle size gradation of construction wastes greatly influences the equilibrium time, rate and the capacity of Pb adsorption. The equilibrium adsorption rate and capacity are 18.1 µg/min and 5.5 μ g/g, respectively, for the construction wastes with the size of 2.36 to 4.75 mm, which are the greatest among the different size gradations. The present study provides a scientific basis for effectively controlling Pb pollution from stormwater runoff and the construction wastes resource utilization.

Key words: stormwater runoff; heavy metal; construction waste; adsorption

doi: 10. 3969/j. issn. 1003 - 7985. 2014. 02. 014

W ith the rapid development of urbanization, increasing urban impervious areas interrupt stormwater infiltration channels and result in a sharp increase in stormwater runoff volume and peak flow. In addition, due to the human activities, atmospheric deposition and other factors, a large number of pollutants are accumulated and discharged into the municipal storm sewer by the stormwater runoff flushing, and as a consequence, enter the city receiving water bodies. With the increasing perfection of the city point pollution control, non-point pollution caused by stormwater runoff has become one of the important sources of watershed pollution. A large number of studies on urban stormwater runoff quality showed that heavy metals from stormwater runoff had become one of the important sources of surface water pollutants, where Pb is one of the highest pollution loads of heavy metals^[1]. Vehicle brake emissions, tire wear, building sidings (weathering of paints and metal components) and atmospheric deposition are important sources of heavy metals^[2]. Heavy metals are persistent pollutants which are widely distributed in the environment, and, thus, they are difficult to be treated due to their characteristics of persistency, wide-ranging and management difficulties. Unlike organic pollutants, heavy metals are difficult to be degraded in the environment and easy to be accumulated in the human body through the food chain and other ways and become toxic when reaching a certain concentration. The Pb in the environmental water can prevent the respiratory metabolism and photosynthesis of aquatic plants, thus affecting the quality and biomass of the plants and changing the population quantity of aquatic animals. Therefore, it becomes necessary to remove heavy metals from stormwater runoff to a feasible extent to improve the quality of water entering the local watershed in an appropriate treatment and management way.

In 1970s, researchers from the United States investigated heavy metal pollution and migration characteristics in urban stormwater runoff. From then on, some researchers from developed countries started to research on heavy metal pollution and its impact factors^[3]. As one of the low impact development (LID) stormwater management measures, bioretention technology (also known as rain garden), which uses vegetable and media to hold and treat stormwater runoff at the source, can not only efficiently control water quality (the removal rate reaches up to 90%)^[4], but also has an ecological function and land-

Received 2013-10-15.

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Foundation items: The National Natural Science Foundation of China (No. 51208022), the National Science and Technology Major Project of China (No. 2011ZX07301-004-01).

Citation: Yang Liqiong, Wang Jianlong, Zhang Xiaoran, et al. Adsorption characteristics of Pb from urban stormwater runoff by construction wastes [J]. Journal of Southeast University (English Edition), 2014, 30(2): 212-219. [doi: 10.3969/j. issn. 1003 - 7985. 2014. 02.014]

scape effects. In addition, bioretention technology can be widely used to control road green belt and open spaces stormwater runoff. Water quality is improved via permeation, soil filtering and adsorption, decomposition and biotransformation mechanisms in bioretention. Laboratory and pilot-scale bioretention box studies have proved that bioretention is an effective practice for improving the quality of urban stormwater runoff^[5]. The type of bioretention media has a significant impact on the removal of heavy metals^[6-10]. Therefore, to select an effective and economical bioretention medium is of great importance for the wide application of bioretention technology.

This study takes construction wastes as the research objects. Sorption of Pb to gradation construction wastes with different particle sizes in stormwater runoff is studied by static and dynamic adsorption experiments. The feasibility of the application of crushed construction wastes as bioretention media is discussed. This study can provide a scientific basis for effectively controlling the Pb pollution from stormwater runoff and the construction waste resource utilization.

1 Materials and Methods

1.1 Adsorbents and characterization

The construction wastes used in the present study are mainly composed of brick and mixed with a small amount of concrete. After crushing with a crusher, the construction wastes were sieved with a test sieve to four different particle size distributions of 2.36 to 4.75 mm, 4.75 to 10.0 mm, 2.36 to 10.0 mm and ≤10.0 mm. The particles were soaked and washed several times with distilled water to remove surface residuals, and then they were dried in an oven at 105 °C for 24 h and stored in a desiccator. The surface area and porosity factor of construction wastes were 2.269 4 m^2/g and 15%, respectively. The chemical oxide composition of the construction wastes was determined by the scanning wavelength dispersive X-ray fluorescence (XRF) using a Rigaku ZSX Primus II spectrometer. BET surface area was measured by automatic adsorption using a NOVE 4200E instrument. The chemical composition of fresh construction wastes, reported as oxides, is shown in Tab. 1. The main components of the construction wastes are oxide of silicon, calcium and aluminum, with the percentage content of 42.5%, 14.2% and 12.0%, respectively. These results are similar to the results reported by previous studies where XRF analyses of construction wastes were carried $out^{[11+2]}$. In addition, there are some other components which are probably harmful to human health and environment, such as Cr and As. However, studies have shown that there is no adverse effect of percolate.

1.2 Adsorption experiment

The stock solution was prepared with a Pb standard

 Tab. 1
 Chemical composition and physical characteristics of construction waste

Constituent	Mass percentage/%	Constituent	Mass percentage/%
Na ₂ O	0.931	Fe_2O_3	4.750
MgO	2.580	NiO	0.006 0
Al_2O_3	12.00	CuO	0.004 2
SiO_2	42.50	ZnO	0.014 5
P_2O_5	0.146 0	As_2O_3	0.002 9
K ₂ O	2.330	$\operatorname{Rb}_2 O$	0.010 3
CaO	14.20	SrO	0.0517
TiO_2	0.647 0	ZrO_2	0.035 2
Cr_2O_3	0.017 4	BaO	0.096 0
MnO	0.093 9	Bi2O3	0.009 6

solution with a concentration of $1 \text{ mg/}\mu\text{L}$. The permeability coefficients of construction wastes were measured by the hydrostatic head method (see Tab. 2). The results show that the permeability coefficients decreased with the increasing particle size gradation of the construction wastes. The static and dynamic adsorption experiments were carried out in the present study.

 Tab. 2
 Permeability coefficients of construction wastes with different size gradations

$k_1 / (m \cdot s^{-1})$	$k_2 / (m \cdot s^{-1})$
1.29×10^{-5}	1.19×10^{-5}
1.56×10^{-3}	1.50×10^{-3}
1.98×10^{-3}	1.88×10^{-3}
6.54×10^{-3}	6.22×10^{-3}
	$\frac{k_1 / (\text{ m} \cdot \text{s}^{-1})}{1.29 \times 10^{-5}}$ 1.56×10^{-3} 1.98×10^{-3} 6.54×10^{-3}

1.2.1 Static adsorption experiment

100 g construction wastes with four different particle sizes were weighed and transferred into a series of 250 mL triangle conical flasks. After adding 150 mL stock solution , the flasks were shaken on a temperature-regulated orbital platform shaker at 80 r/min. Samples were analyzed at 1, 3, 5, 10, 20, 30, 40, 60, 90, 120, 180, 240 and 300 min intervals during shaking to study the influence of reaction time on the adsorption equilibrium and determine the optimum reaction time. Prior to analysis , 50 mL samples were collected and then subsequently filtered through 0.45 μ m hydrophilic membranes to remove suspended solids , and the filtrates were stored in PVC bottles and were kept in the fridge at 4 °C for determination.

1.2.2 Dynamic adsorption experiment

The construction wastes with four different particle size distributions were added into experimental columns with several steps , and compacted with each step. The dynamic adsorption experiment was operated under a condition of continuous water input with a peristaltic pump at 4 mL/s. 50 mL samples were collected at the time intervals of 3 , 5 , 10 , 20 , 30 , 60 , 90 , 180 and 300 min. The samples were filtered through 0.45 μ m hydrophilic membranes to remove the suspended matters , and then stored in PVC bottles. The samples were kept in the fridge at 4 °C for further analysis. Bioretention columns dynamic

simulation experimental set-up is shown in Fig. 1.



Fig. 1 Permeation columns used for dynamic adsorption experiment

1.3 Aqueous sample analysis

Samples were analyzed by the graphite furnace atomic absorption spectrophotometer method using a Hitachi Z-2010 Polarized Zeeman atomic absorption spectrophotometer (AAS). A calibration curve was made with the Pb standard solution of the National Standard Center for standard matter. 20.00 µg/L standard Pb solution was diluted by the standard solution from the National Standard Center with 2% nitric acid. The absorbance of the standard solution with the concentration of 0, 5.00, 10.00 , 15.00 , 20.00 μ g/L was measured by the graphite furnace automatic dilution function. The correlation coefficient of the calibration curve was 0.999 2. The concentration of Pb in the supernatant after adsorption by construction wastes was measured. The amount of sorption was calculated based on the concentration of Pb in the solution before and after adsorption.

1.4 Adsorption isotherms

Under optimum adsorption equilibrium time , 5 , 10 , 20 , 30 , 50 , 70 and 100 g of construction wastes with different particle size distributions were weighed to study the isothermal adsorption process. The adsorption capacity of the construction wastes is calculated as

$$Q = \frac{V(C_0 - C_1)}{m} \tag{1}$$

where Q is the adsorption capacity per unit mass of construction wastes , mg/g; C_0 is the initial concentration of Pb in the aqueous solution , mg/L; C_1 is the equilibrium concentration of the test solution , mg/L; V is the volume of sample , L; m is the mass of construction wastes , g.

2 Results and Discussion

2.1 Characterization of construction wastes

The scanning electron microscopy (SEM) images for the surfaces of construction wastes before and after adsorption are shown in Fig. 2. The surface of construction wastes becomes smoother after adsorption , which may be due to the deposition of Pb by physisorption , or due to a progressive change in surface mineralogy with time. The comparison of the component composition of construction wastes before and after adsorption (see Fig. 3) shows a significant decrease of the element content in Al and Ca , while showing a significant increase in Mg and Si. The significant decrease in Ca content may be related to the loss of free lime (CaO) , which results in high pH^[13]. Therefore , the changes in pH before the discharge of the stormwater should be taken into account for practical applications.



Fig. 2 SEM images of the construction wastes. (a) Before adsorption; (b) After adsorption



Fig. 3 Change of relative concentration of mineral elements. (a) C , N , O , etc.; (b) F , P , Cl , etc.

This result shows that there are no detrimental effects on human health or on the environment by application of construction wastes in stormwater runoff purification.

2.2 Adsorption isotherms

Under a constant temperature , sorption between solid and liquid is often described by the Langmuir and Freundlich isotherm model. The Langmuir adsorption isotherm (in linear form) ^[14] is given as

$$\frac{C_{\rm e}}{Q_{\rm e}} = \frac{C_{\rm e}}{Q_{\rm m}} + \frac{1}{K_{\rm a}Q_{\rm m}}$$
(2)

where Q_e is the amount of Pb adsorbed at equilibrium , mg/g; Q_m is the saturated adsorption capacity of Pb , mg/g; C_e is the final equilibrium concentration of Pb in the test solution , mg/L; K_a is the Langmuir adsorption constant.

The Freundlich adsorption isotherm (in linear form) ^[8] is given as

$$\lg Q_e = \frac{1}{n} \lg C_e + \lg K_f \tag{3}$$

where K_i is the Freundlich adsorption constant, and n is a constant.

The static adsorption process of Pb from stormwater runoff to construction wastes is studied by the batch static experiments. The experimental data is fitted by the Langmuir and Freundlich isotherm models and their calculated parameters are summarized in Tab. 3. The Freundlich adsorption isotherms are shown in Fig. 4. Compared with the correlation coefficients, the equilibrium data for the adsorption process is better fitted by the Freundlich isotherm model than the Langmuir isotherm model. The correlation coefficients of model equations derived from different particle size gradation construction wastes are higher than 0.95. In addition, the particle size of 2.36 to 4.75 mm is fitted by the Langmuir isotherm equation, but the correlation coefficient is 0.934 3. Kragović et al.^[15] studied the removal of Pb in aqueous solutions by natural zeolite. Their results show that the adsorption of Pb by natural zeolite fits the Freundlich isotherm equation well, which is consistent with the results of the present study. However, some other types of adsorbents reported previously that the sorption data was fitted by the Langmuir adsorption model^[16-17]. Zhang et al.^[8] found that the adsorption isotherm of Pb2+ to peanut shells was better fitted by the Langmuir equation than the Freundlich equation measured by sequencing batch adsorption experiments. Therefore, there is a large difference in different bioretention media on the adsorption properties of the Pb^{2+} .

Tab. 3 Parameters of Langmuir and Freundlich equations



Fig. 4 Sorption isotherms of Pb sorption to construction wastes with different particle size gradations fitted by the Freundlich model. (a) 2.36 to 4.75 mm; (b) 2.36 to 10.0 mm; (c) 4.75 to 10.0 mm; (d) ≤ 10.0 mm

2.3 Adsorption kinetics

The adsorption of Pb from simulated stormwater runoff to the construction wastes can be analyzed by pseudofirst-order and pseudo-second-order rate expressions as applied by Liu et al^[16]. The linear expression of the pseudo-first-order equation is given as^[11]

$$\log(Q_{e} - Q_{t}) = \log Q_{e} - \frac{k_{1}}{2.303}t$$
(4)

where Q_i is the amount of Pb adsorbed at time t, mg/g; k_1 is the equilibrium rate constant, which can be determined by a linear plot of log($Q_e - Q_i$) vs. t.

Tien et al.^[18-20] found that the adsorption kinetics of heavy metals was better fitted to a pseudo-second-order kinetic model. The linear expression is given as

$$\frac{t}{Q_{t}} = \frac{1}{k_{2}Q_{e}^{2}} + \frac{t}{Q_{e}}$$
(5)

where k_2 is the rate constant at equilibrium. The adsorption kinetic fitting curves of Pb by construction wastes with different particle sizes are shown in Fig. 5. The fitting kinetic equations are shown in Tab. 4. The pseudosecond-order kinetic equation of Pb adsorption from the synthetic simulated runoff to the construction wastes fits the linear equation well (see Fig. 5) and it is significantly better than the pseudo-first-order kinetics model (see Tab. 4). Many studies at domestic and overseas on adsorption of heavy metals to saccharomyces cerevisiae^[21], natural zeolite^[22], blast furnace $slag^{[23]}$ and hazelnut shell^[24] have shown that the data of sorption kinetics fit the pseudo-second-order kinetics model^[25], which is



Fig. 5 Second-order equation for the adsorption of Pb by construction wastes

Tab.4Parameters and correlation coefficients for the kineticmodels

Size distribution/mm	$k_2 / (g \cdot mg^{-1} \cdot min^{-1})$	R^2	Kinetic model
2.36 to 4.75	3 099.50	0.9999	$Q_t = \frac{0.093\ 6t}{1\ +17.035t}$
4.75 to 10.0	87.48	0.9996	$Q_t = \frac{0.001\ 13t}{1+0.315t}$
2.36 to 10.0	132.81	0.9987	$Q_t = \frac{0.001\ 55t}{1+0.453t}$

consistent with the experimental results in the present study.

2.4 Effect of construction wastes with different particle size gradations on Pb removal

2.4.1 Adsorption capacity

The adsorption capacities of construction wastes with different particle sizes are shown in Fig. 6. At the initial stage, the adsorption rate increases dramatically with the increase of time until reaching a plateau. The size distributions of adsorbent significantly influence the adsorption efficiency of heavy metals^[26]. Construction wastes with sizes smaller than 10.0 mm reach equilibrium in the shor– test time (about 1 h) . The equilibrium adsorption capacities of construction wastes with particle sizes smaller than 10.0 mm , 2.36 to 10.0 mm , 2.36 to 4.75 mm and 4. 75 to 10.0 mm for Pb sorption are 4.1, 3.4, 5.5 and 3. $6 \ \mu g/g$, respectively. The adsorption capacity of construction wastes with particle size of 2.36 to 4.75 mm is the largest (see Fig. 6(a)), probably because of their larger specific surface area. Dynamic experimental results (see Fig. 6(b)) show that the construction wastes with four different size distributions for Pb sorption reach equilibrium within 10 min. Their equilibrium adsorption capacities (i.e., 1.8, 2.1, 2.1 and $2.3 \mu g/g$, respectively) are much smaller than the data derived from static experimental results. Therefore, the construction wastes have



Fig. 6 Changes in adsorption capacity of construction wastes with different particle sizes on Pb adsorption. (a) Static experiment; (b) Dynamic experiment

not reached adsorption saturation. The data show that construction wastes with 2.36 to 4.75 mm can be used as a bioretention media to remove Pb from stormwater run-off.

2.4.2 Adsorption rate

Changes in adsorption rates of construction wastes with different particle sizes on Pb adsorption are depicted in Fig. 7. The adsorption rate equation is given as

$$\nu = \frac{m_i}{t} \tag{6}$$

where ν is the adsorption rate , mg/min; m_t is the amount of Pb adsorbed within time t, mg; and t is the sampling interval time, min.



Fig.7 Changes in adsorption rates of construction wastes with different particle size gradations on Pb adsorption. (a) Static experiment; (b) Dynamic experiment

As shown in Fig. 7(a) , the adsorption rates of Pb adsorption to construction wastes with the size of 2. 36 to 4.75 mm and smaller than 10.0 mm decrease rapidly. The adsorption rates of Pb adsorption to construction wastes with 2.36 to 10.0 mm first increase and then decrease , while construction wastes with 4.75 to 10.0 mm first decrease and then increase until reaching a plateau , which may be due to the unevenly distributed size grading of construction wastes in this study. The stable adsorption rates of Pb sorption to construction wastes with four particle sizes are 18.1, 1.4, 5.6 and 1.2 μ g/min, respectively. The adsorption rates of construction wastes in the dynamic adsorption process (see Fig. 7(b)) decrease gradually. The adsorption rates

are basically identical , and stable adsorption rates (i.e., 0.8, 0.6, 0.7 and $0.7~\mu g/min$, respectively) are much smaller than the data derived by the static experimental data. With the increase of bioretention media depth , the Pb concentration of stormwater decreases and may be harmful to the environment.

2.4.3 Removal rate

The average removal rates of construction wastes with different particle sizes and their maximum values are summarized in Tab. 5. The equation of removal rate is given as

$$\eta = \frac{C_0 - C_t}{C_0} \times 100\%$$
 (7)

Tab.5 Average removal rates of construction wastes with different particle sizes and their maximum values

E	Size	Removal	Equilibrium
Experiment	distribution/mm	rate/%	time/min
	2.36 to 4.75	98.7	30
c:	4.75 to 10.0	99.8	300
Static	2.36 to 10.0	99.9	60
	≤10.0	100.0	< 5
	2.36 to 4.75	99.9	5
D ·	4.75 to 10.0	99.9	10
Dynamic	2.36 to 10.0	99.9	10
	≤10.0	100.0	< 5

As can be seen from Tab. 5, construction wastes with different particle sizes show a good purification effect on Pb removal from stormwater runoff and up to a 90% Pb removal rate is achieved. In general , particles with smaller size have larger specific surface area and better surface adsorption properties. As a consequence, the adsorption capacity is higher for construction wastes of smaller sizes than those of larger sizes^[27]. Our conclusion is consistent with the general conclusions reported previously. The removal rates derived from dynamic experiments are higher than those from static experiments. At the initial stage of dynamic experiments, the removal rate of heavy metal increases rapidly with the increase of time. Construction wastes with four particle sizes reach equilibrium within 10 min, which indicates that the adsorption of Pb to construction wastes is a fast reaction process that can be completed in a short time. As a medium for bioretention facilities, construction wastes can be used to remove Pb from urban stormwater runoff due to their great adsorption capacities.

2.5 Influencing factors of adsorption

Numerous studies domestically and overseas showed that the impact factors of solid adsorption effects on Pb were mainly the adsorption time, the amount of adsorbent, the initial concentration and the pH value. With the increase of the adsorption time, the adsorption removal rate of the particulate matter for Pb in the solution gradually increases until reaching equilibrium where the removal rate does not change significantly^[28] and may even decrease to a certain extent. The increase of adsorbent dosage may also increase the removal rate for Pb adsorption. However , the removal rate may decrease if too much adsorbent is used. Therefore , taking into account the cost , it is desirable to optimize the dosage of adsorbent. The pH of the solution is an important factor of Pb adsorption. The optimum adsorption pH values are different for different adsorbents^[29-30]. Previous studies have showed that competition occurs between different heavy metal ions and adsorbents adsorb heavy metals selectively^[31]. In practical applications , the design of parameters should be optimized based on the influencing factors of heavy metal removal.

3 Conclusions

1) Construction wastes with different particle sizes show a good effect on Pb adsorption , and their average removal rate reaches up to 99%. The removal efficiency of construction wastes increases with the increase of size grading , but the penetrating ability of construction wastes turns to worse. The adsorption removal effect of construction wastes with different sizes on Pb adsorption follows in the order: $\leq 10.0 \text{ mm} > 2.36 \text{ to } 4.75 \text{ mm} > 2.36 \text{ to } 10.0 \text{ mm} > 4.75 \text{ to } 10.0 \text{ mm}.$

2) The adsorption isotherm analysis shows that the adsorption of Pb to construction wastes adsorption fits well with the Freundlich isotherm model and the correlation coefficients are 0.976 8 to 0.996 3. The adsorption kinetic process is well described by the pseudo-second-order kinetic equation and the correlation coefficients are 0.999 6 to 0.999 9.

3) In the engineering projects, construction wastes can be applied as the adsorption media of bioretention facilities, constructed wetlands and other ecological measures for the removal of Pb from stormwater runoff. The present study provides a scientific basis for effectively controlling Pb pollution from stormwater runoff and the construction wastes resource utilization.

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建筑垃圾对城市雨水径流中 Pb 的吸附特性

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摘要: 以砖混建筑垃圾为研究对象,采用人工模拟雨水,通过静态和动态吸附实验研究了不同粒径粒级建筑 垃圾对雨水径流中 Pb 的吸附效果.实验结果表明: 准二级动力学模型比准一级动力学模型能更好地描述建 筑垃圾对 Pb 的吸附过程; Freundlich 等温模型能较好地拟合其等温吸附过程; 不同粒径粒级建筑垃圾均对 雨水径流中的 Pb 具有较好的净化效果,去除率高达 99%, 粒径粒级越小,对 Pb 的净化效果越好,但其渗透 性能越差; 建筑垃圾的粒径粒级对 Pb 的吸附平衡时间、吸附速率和吸附量具有重要影响,粒径 2.36~4.75 mm 的建筑垃圾对 Pb 的平衡吸附速率和平衡吸附量最大,分别为 18.1 μg/min 和 5.5 μg/g. 上述研究结果 为城市雨水径流中 Pb 污染的有效控制以及建筑垃圾资源化提供了科学依据.

关键词:雨水径流;重金属;建筑垃圾;吸附

中图分类号: TU993.1