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CRUSHED CONSTRUCTION AND DEMOLITION WASTE AS REACTIVE MEDIA IN PERMEABLE REACTIVE BARRIERS TO IMMOBILIZE PB(II) AND CD(II) FROM LANDFILL LEACHATE

Nguyen Lan Huong[a], Bui Xuan Khanh[b], Tran Thi Viet Nga[a,*], Nguyen Tien Dung[c]

[a] *Faculty of Environmental Engineering, Hanoi University of Civil Engineering, 55 Giai Phong road, Hai Ba Trung district, Hanoi, Vietnam*
[b] *Faculty of Chemistry, University of Science, Vietnam National University Hanoi, 19 Le Thanh Tong street, Hoan Kiem district, Hanoi, Vietnam*
[c] *Faculty of Construction Materials, Hanoi University of Civil Engineering, 55 Giai Phong road, Hai Ba Trung district, Hanoi, Vietnam*

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Abstract

This study evaluates the effectiveness of using crushed construction and demolition waste as reactive media for applying in Permeable Reactive Barriers to treat contaminated groundwater by landfill leachate. Modern analytical methods such as SEM and EDX were used to analyze the physicochemical properties of crushed demolition waste, demonstrating that the material has a porous structure on the surface that is suitable for the targeted heavy metal adsorption. For ion Pb^{2+}, the maximum adsorption amount of 114.05 mg/g was for 1-3 mm fraction and 105.05 mg/g for 3-5 mm fraction. Similarly, ion Cd (II) achieves the maximum adsorption capacity of 8.43 mg/g with 1-3 mm and 6.26 mg/g with 3-5 mm fraction. Simultaneously, this paper investigates the impact of additional parameters such as time and pH on the adsorption efficiency of the materials.

Keywords: permeable reactive barriers; landfill leachate; construction demolition waste; heavy metals; adsorption.

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1. Introduction

Vietnam is one of the fastest growing economies in the ASEAN countries with rapid urban and infrastructure development. This results in an ever-increasing amount of construction and demolition waste (CDW). The daily CDW generation exceeds 3,000 tons/day in Hanoi City (the capital of Vietnam), due to active construction activities including new constructions, renovations, and demolition of buildings [1]. Despite the growing amount of CDW daily, the recycling rate remains low (1-2%). CDW is mostly reused for landfilling, ground leveling, and backfilling on-site (construction and demolition sites) [2]. Although the reuse of CDW for wastewater treatment has been studied and researched in other countries [3–7]. For example, Wang, et al investigate the high ability of CDW to absorb Pb (II), Cd (II) and Cu (II) in urban storm water [8]. In the study by Kumara et al, autoclaved aerated concrete (AAC) wastes were proven to be an effective absorbent to remove Pb (II) and Cd (II) in wastewater [3]. Nonetheless, research on the use of CDW for wastewater treatment in Vietnam remains limited. A recent study by Son et al. [9, 10] found that AAC waste manufactured from Bac Ninh AAC factory is an effective absorbent to remove Cadmium from wastewater.

Groundwater pollution from leachate has been a critical problem from the open dumping of municipal solid waste in developing countries. According to a report by the Ministry of Construction of

*Corresponding author. *E-mail address:* ngattv@huce.edu.vn (Nga, T. T. V.)

Vietnam, there are 657 landfills in operation in Vietnam, with a total size of 4900 hectares, of which only 31% are sanitation landfills. According to the Hanoi Urban Environment Company (URENCO), each ton of waste will generate 0.43 m^3 of leachate, which means that with 5000 tons of waste per day, Nam Son landfill will generate about 2150 m^3 of leachate/day. The pollution of groundwater in the Lai Son village near the Nam Son landfill - the biggest landfill of Hanoi city - was reported by Giang et al. [11] with high COD, Cadmium, Lead, Copper, and Coliform values. In addition, all the groundwater samples indicated Cyanide concentration exceeding permissible limits.

Permeable Reactive Barrier (PBR) is one of the new technologies with a simple, low-cost, long-life method in which a wall structure is placed underground to allow contaminated seepage to flow through, attempting to supplement or replace existing methods [12, 13]. Contaminated water streams are treated by precipitation, adsorption or decomposition of pollutants when the pollutant seepage flows through the reactive zone [12]. Much research has been done and focused on the design of PRB, selection of suitable materials to construct PRB and demonstrated their effectiveness in removing heavy metals and inorganic contaminants [14–17] but only a few studies focus on using CDW as reactive barriers in PRB although the feasibility of PRBs with low-cost, locally and readily available reactive materials to treat landfill leachate has been studied by Arthanayaka et al. [18].

To contribute to establishing the circular economy in Vietnam, this research targets reusing of CDW to develop the low-cost treatment of contaminated ground water by landfill leachate. The CDW was utilized as reactive media in Permeable Reactive Barriers. The objectives are (1) to study the adsorption properties of selected heavy metals (Pb^{2+} and Cd^{2+}) on to crushed CDW; (2) to discuss the environmental safety of applying CDW as PRB media to remove heavy metals from contaminated ground water by landfill leachate.

2. Materials and methods

2.1. Adsorbent Preparation

To evaluate the absorption capacity of CDW to be used as reactive media to immobilize Pb (II) and Cd (II) in contaminated groundwater by landfill leachate in PRB, Crushed Demolition Waste samples were sorted from a pilot CDW recycling plant in Dong Anh, Hanoi. Samples were sieved and graded according to particle size (1-3 and 3-5 mm fractions), washed with distilled water to remove fine particles, dried and stored at room temperature.

2.2. Batch experiment

The batch adsorption test was described in Fig. 1. The stock solutions were prepared by dissolving $CdCl_2xH_2O$, Pb $(NO_3)_2$ to 1000 ppm of Cd^{2+} and Pb^{2+} solutions. The first test was to examine the equilibrium contact time (0-72 hours) with fixed initial metal ion concentration (20 g/L) and agitation speed of 100 rpm. The second test involved the study of variation in the initial metal ion concentration (100, 150, 200, 300, 400 and 500 mg/L) for Cd^{2+} and (1000, 1500, 2000, 4000, and 5000) for Pb^{2+} at the optimum contact time (based on the results of the first set), while keeping the agitation speed and sorbent dosage as in the first set. The concentration of heavy metals after adsorption was determined by taking 50 mL of the solution and separating the solid with filter paper. The remaining

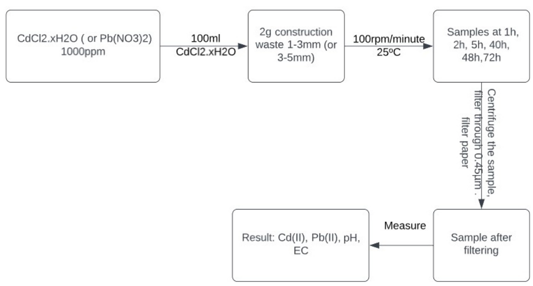


Figure 1. Batch adsorption test procedure

concentration of Pb^{2+} ions was determined through atomic absorption spectroscopy (F-AAS, Shimazu, Japan).

The amount of contaminant absorbed (Q_e) retained in the CDW phase could be specified by taking into consideration the initial and equilibrium concentrations, was calculated as follows [19]:

$$Q_e = V\frac{C_o - C_e}{M} \tag{1}$$

where C_o and C_e are initial concentration and equilibrium concentration (mg/L) of a contaminant in solution; V is solution concentration in a flask (L); and m is CDW mass in a flask (g).

We used two models to examine the sorption isotherm:

The Langmuir model:

$$q = q_{\max}\frac{bCt}{1 + bCt} \tag{2}$$

where $q_{\max}$ is the maximum adsorption capacity corresponding to monolayer adsorption (mg/g), b is the Langmuir adsorption constant (L/mg), C_t is adsorption capacity at equilibrium time.

The Freundlich model:

$$q = lgk\frac{1}{n}lgC \tag{3}$$

where k is the Freundlich constant representing the relative adsorption capacity of the adsorbent (L/g), and $1/n$ is the heterogeneity factor denoting the adsorption intensity.

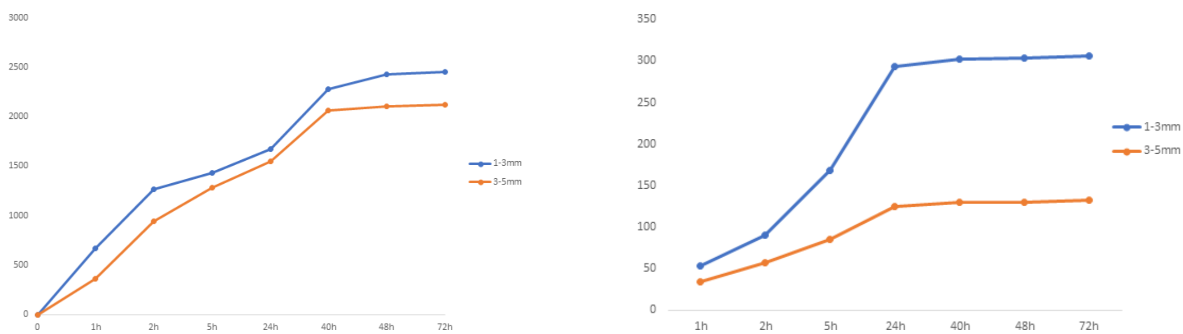
2.3. *Physical and chemical characterization of CDW*

The physical and chemical properties of CDW before and after adsorption was tested by Energy dispersive X-ray (EDX- LabX XRD-6100 instrument (Shimadzu, Japan) and Scanning electron microscopy (SEM, HITACHI-S-4800, Tokyo, Japan).

3. Results and discussions

3.1. *Results of batch experiment test*

Fig. 2 shows the effect of contact time on adsorption. Under the temperature condition of 25 °C, with the same Pb (II) concentration of 5000 ppm and the adsorbent mass of 2 g, the CDW reached adsorption equilibrium at 48 hours with the maximum adsorption capacity of 114.05 mg/g for material particles size 1-3 mm. Meanwhile, the material with the size of 3-5 mm has the maximum adsorption capacity of 105.05 mg/g at 48 hours, this value is lower than 1-3 mm size absorbent.

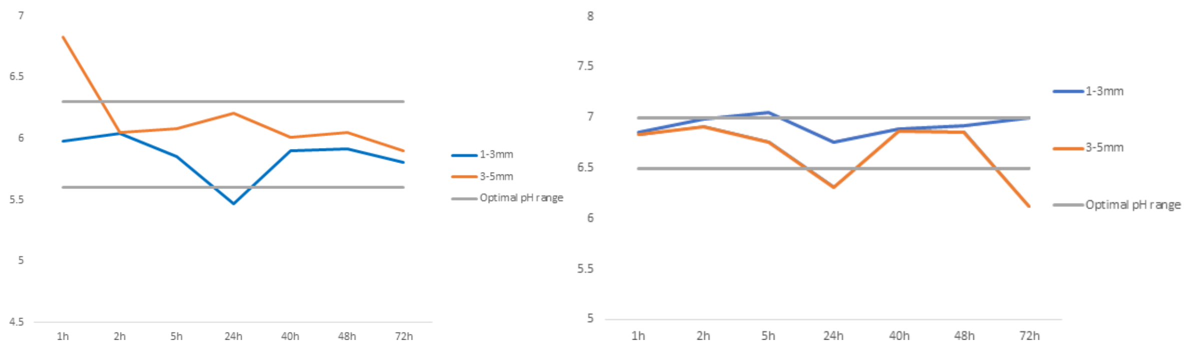


(a) Pb (II) absorption capacity of CDW

(b) Cd (II) absorption capacity of CDW

Figure 2. Pb (II) & Cd (II) absorption capacity of CDW at particle size 1-3 mm and 3-5 mm

For ion Cd (II), the CDW reached adsorption equilibrium at 24 hours with the maximum adsorption capacity of 8.43 mg/g for the particle size of 1-3 mm. Meanwhile, the CDW with the size of 3-5 mm reached the maximum adsorption capacity of 6.26 mg/g at 24 hours, lower than the 1-3 mm CDW material.

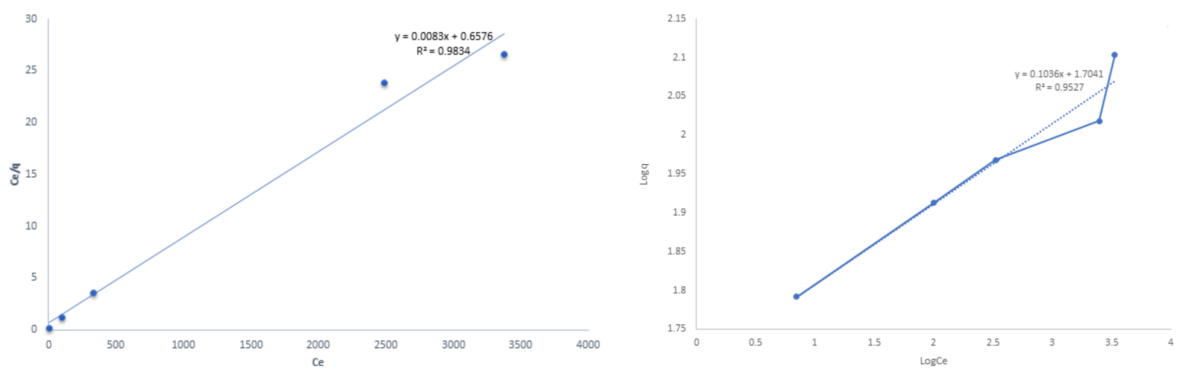


(a) Pb (b) Cd

Figure 3. pH value of CDW samples with varied sizes when adsorbing Pb, Cd

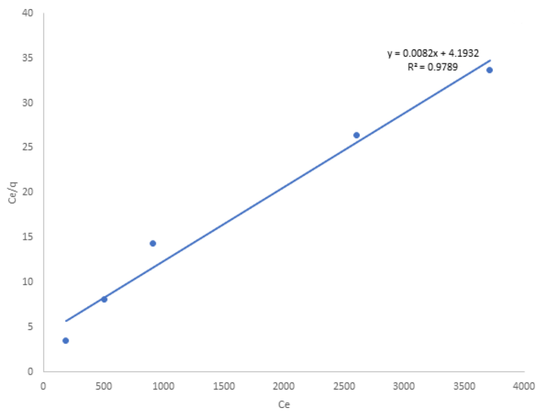
The value of pH is one of the factors that directly affect the adsorption capacity of heavy metals (Figs. 3(a) and (b)). In the paper by [20] the final partitioning of the metal species into bound to CCDW, precipitated, or remained in solution (dissolved) was studied. Referring to the results from [20] where the pH range was between 5.6 and 6.3 for Pb (II) and 6.5 and 7 for Cd (II) with the number of metals bound onto CDW was more than 80%. This trend that the higher the pH, the stronger the presence of –COOH and OH– groups on the surface of the material, the dissociation of H^+ from this functional group accelerate the accumulation of metal ion on to the surface of the absorbent by enhancing the ion exchange capacity with Pb^{2+} and Cd^{2+}. When pH is above 7, more proportion of metals ion precipitates onto the absorbent surface, which affects the chemical adsorption capacity of the absorbent.

Fig. 4 and Fig. 5 indicate the Langmuir and Freundlich adsorption isotherms of the two heavy metals Pb (II) and Cd (II). The Langmuir and Freundlich isotherm parameters were obtained from the fitting curves and the correlation coefficients (R^2) (Table 1). The heavy metal adsorption of the materials fitted both isotherms - Freundlich and Langmuir models with the R square value greater than 0.8. Heavy metal adsorption follows two principal mechanisms. The first follows specific adsorption or chemisorption such as the complexation of metal ions with specific surface functional groups. This

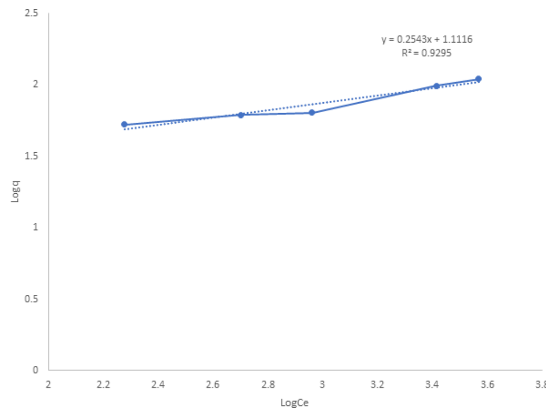


(a) Langmuir adsorption isotherm of Pb (II) by CDW size 1-3 mm

(b) Freundlich adsorption isotherm of Pb (II) by CDW size 1-3 mm



(c) Langmuir adsorption isotherm of Pb (II) by CDW size 3-5 mm



(d) Freundlich adsorption isotherm of Pb (II) by CDW size 3-5 mm

Figure 4. Langmuir adsorption isotherm of Pb (II) by CDW size 1-3 mm (a) size 3-5 mm (c); Freundlich adsorption isotherm of Pb (II) by CDW size 1-3 mm (b) size 3-5 mm (d)

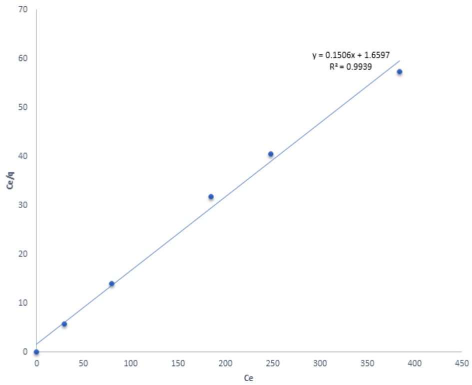
Table 1. Adsorption isotherm parameters

Adsorption of Cd (II)	Grain size 1-3 mm		Grain size 3-5 mm	
Langmuir model	q_m (mg/g)	114.05	q_m (mg/g)	105.05
	b (L/g)	0.00196	b (L/g)	0.0126
	R^2	0.9939	R^2	0.9907
	RL	0.296	RL	0.059
Freundlich model	$1/n$	0.1036	$1/n$	0.2543
	n	9.65	n	3.93
	K_f (mg/g) (L/mg)$1/n$	50.59	K_f (mg/g) (L/mg)$1/n$	12.93
	R^2	0.9702	R^2	0.986
Adsorption of Pb (II)	**Grain size 1-3 mm**		**Grain size 3-5 mm**	
Langmuir model	q_m(mg/g)	8.43	q_m(mg/g)	6.26
	b (L/g)	0.091	b (L/g)	0.074
	R^2	0.9834	R^2	0.9789
	RL	0.098	RL	0.1173
Freundlich model	$1/n$	0.0202	$1/n$	0.0517
	n	49.5	n	19.34
	K_f (mg/g) (L/mg)$1/n$	5.35	K_f (mg/g) (L/mg)$1/n$	4.35
	R^2	0.9527	R^2	0.9295

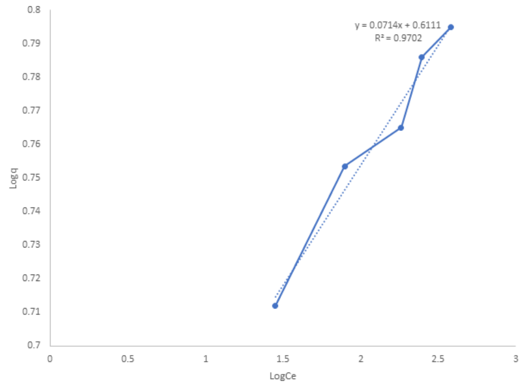
is selective, less reversible and results in a monolayer formation. The result of the R^2 value for the Langmuir model greater than the Freundlich model indicates the major process was chemisorption. The second is non-specific adsorption which is non-selective and reversible due to weaker binding energy but is not limited to monolayers [7].

3.2. Result of scanning electron microscopy

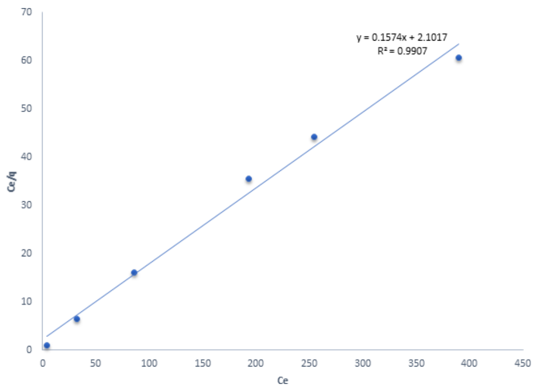
Surface morphology of materials before and after heavy metal adsorption with different ratios is shown in Fig. 6. The figure indicates that CDW is formed from unsolid cement stone with the irreg-



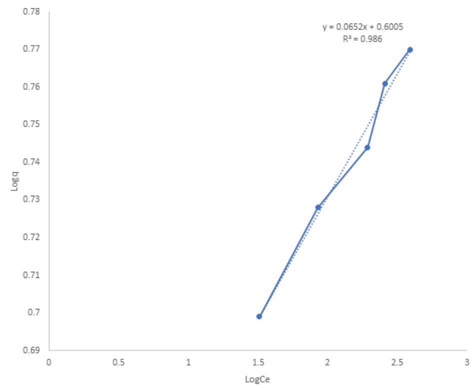
(a) Langmuir adsorption isotherm of Cb (II) by CDW size 1-3 mm



(b) Freundlich adsorption isotherm of Cb (II) by CDW size 1-3 mm



(c) Langmuir adsorption isotherm of Cb (II) by CDW size 3-5 mm

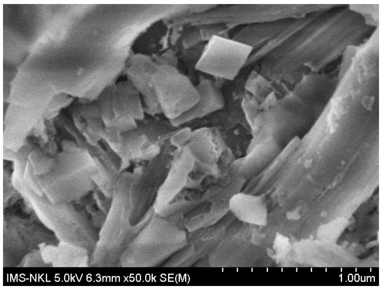


(d) Freundlich adsorption isotherm of Cb (II) by CDW size 3-5 mm

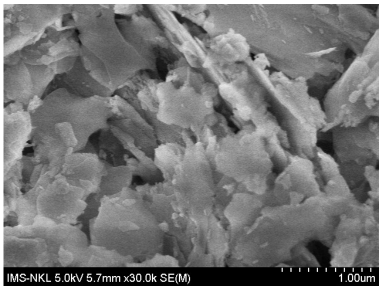
Figure 5. Langmuir adsorption isotherm of Cd (II) by CDW size 1-3 mm (a) size 3-5 mm (c); Freundlich adsorption isotherm of Cd (II) by CDW size 1-3 mm (b) size 3-5 mm (d)

ular, fractured and littered surface of CDW before adsorption. Large aggregate particles (stone), and small aggregate (sand) are all covered with cement. The crystals have regular sizes, clearly shown on the SEM image of the CDW sample (a). Samples of materials after adsorption of Pb (II) and Cd (II) gain rougher surfaces, Cd and Pb crystals adhere and cover tightly. The surface of Pb–CCF comprises a reticular, lead-bearing, calcium silicate network and rare, lead-bearing, calcium-rich, distorted polygons [21]. The morphology of the lead-bearing network is representing Hydro Silicate Calcium (CSH) – one form of the four CSH morphologies in cement hydration. The exchange of ion Pb^{2+} and Ca^{2+} resulted from the isomorphic substitution by Pb^{2+} for Ca^{2+} in the calcium silicate hydrate phase. On the other hand, the surfaces of cadmium-laden CDW indicated a cadmium-bearing cancellated network and platy foils. The surface morphology of the CDW samples is porous, making them suited for absorbing heavy metals in wastewater. Pb (II), Cd (II) adsorption resulted in an appearance of Pb, Cd on the surface of the materials.

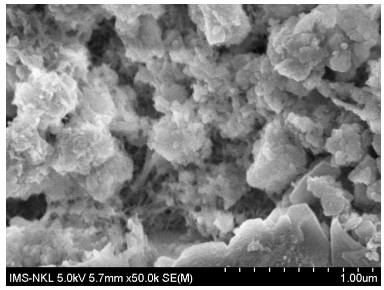
Fig. 7 shows the chemical composition of the samples. The CDW before adsorption composition is calcium in the range 15.41-17.36%, silicon 6.21%-10.82%; oxygen accounted for 51 - 60.49%. These elements are the typical Portland cement components [20]. In the samples after adsorption of Pb (II), Cd (II) appeared the adsorbed metals such as Pb 8.7% and Cd 1.77%. Based on the atomic



(a) SEM images of CDW before adsorption test

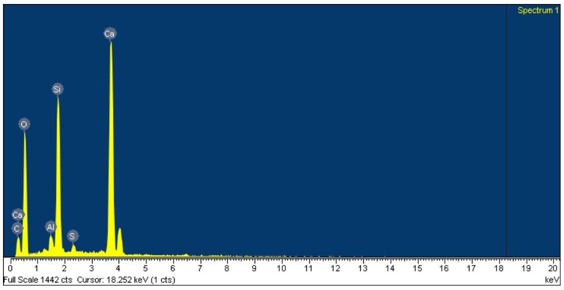


(b) CDW after adsorption of Pb

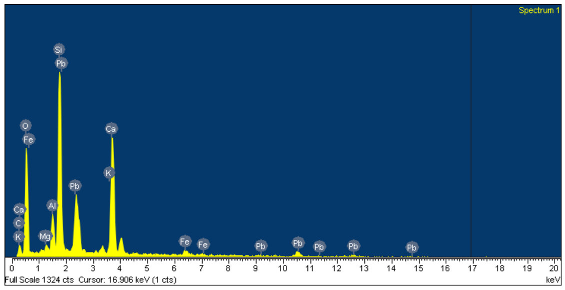


(c) CDW after adsorption of Cd (II)

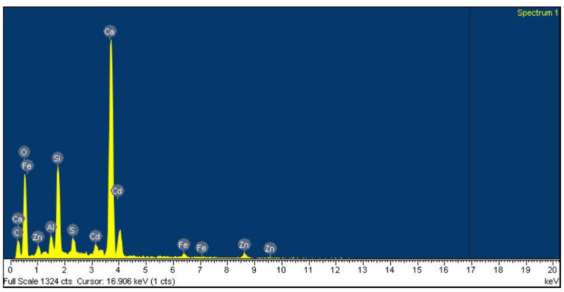
Figure 6. SEM images of CDW before adsorption test, CDW after adsorption of Pb (II), CDW after adsorption of Cd (II)



(a) EDX figure of CDW before adsorption test



(b) EDX figure of CDW after Pb (II) adsorption



(c) EDX figure of CDW after Cd (II) adsorption

Figure 7. EDX figure of CDW before adsorption test, CDW after Pb (II) adsorption, CDW after Cd (II) adsorption

composition, the values of the adsorbed metals Pb (II) and Cd (II) were lower than calculated, indicating the partial metal leakage during the adsorption process. The reason for leakage is the metals present in the loose floc precipitate on to the surface of the adsorbent [21]. In material samples, with high Ca^{2+} content from 15-17%, the material's ability to adsorb heavy metals is excellent. This heavy metal adsorption process is mainly ion exchange.

3.3. Environmental safety of using CDW as reactive media for PRB to treat landfill leachate

The leaching test for two CDW samples was examined at a certified lab according to USEPA 1311. Table 2 shows the measured values were much smaller than those of environmental standards, i.e., QCVN 07:2009/BTNMT for hazardous waste. All the harmful metals were detected below the accepted standard and the results are in agreement with previous studies by Le et al. and Shin et al.

[6, 22] From the result of the batch experiment, CDW is proven to be an efficient sorbent and support its use as a reactive media in a PRB for the remediation of landfill leachate with lead, cadmium. In addition, high pH and high acid neutralization capacity materials such as CDW will enhance heavy metal immobilization and increase the capacity to neutralize acid input, therefore preventing and controlling the leaching of heavy metal.

Table 2. Results of leaching test

Parameters	CDW1 1-3 mm	CDW2 3-5 mm	QCVN 07:2009/BTNMT
Cr	< 0.003	< 0.003	5
Co	< 0.003	< 0.003	80
Ni	< 0.003	< 0.003	70
Cu	< 0.003	< 0.003	
Zn	0.01	0.009	250
As	< 0.003	< 0.003	2
Se	< 0.003	< 0.003	1
Mo	< 0.004	< 0.004	350
Cd	< 0.003	< 0.003	0.5
Pb	< 0.003	< 0.003	15

4. Conclusions

The effects of pH, particle size, and contact time on the heavy metal adsorption process of CDW materials were confirmed in this study. Equilibrium times of 48 hours and 24 hours were observed for the removal processes of Pb (II) and Cd (II) respectively. The mechanism of interaction of Pb (II) with CDW was via diffusion into the cement matrix. The optimum pH range for was between 5.6 and 6.3 for Pb (II) and 6.5 and 7 for Cd (II). The physicochemical properties of materials are determined by modern analytical methods such as scanning electron microscopy (SEM), and energy dispersive method (EDX). The results show that the initial material sample has a surface morphology with pores, suitable for the adsorption of heavy metals in contaminated ground water. The maximum adsorption amount of 114.05 mg/g was for 1-3 mm grain size and 105.05 mg/g for 3-5 mm grain size. Similarly, the maximum adsorption capacity of 8.43 mg/g with a grain size 1-3 mm and 6.26 mg/g with a grain size of 3-5 mm was recorded for Cd (II). In addition, the results of isothermal adsorption studies show that the Langmuir and Freundlich models describe accurately the adsorption process of heavy metals. According to the findings of this study, CDW is an efficient and safe sorbent, and its usage as a reactive media in a PRB for the immobilization of Pb (II) and Cd (II) in contaminated groundwater by landfill leachate is feasible.

Acknowledgments

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