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REVIEWS ON THE APPLICABILITY OF CONSTRUCTION AND DEMOLITION WASTE AS LOW-COST ADSORBENTS TO REMOVE-HEAVY METALS IN WASTEWATER

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ABSTRACT: Removal of heavy metals from wastewater is of special concern due to the persistence of heavy metals in the environment. The industrial discharge of heavy metals adversely affects soil and water resources, aquatic organisms, and ecosystem integrity. In addition, high concentrations of heavy metals are detected in solid waste landfill leachate in many developing countries due to non-segregated waste dumping. At present, various kinds of adsorbents such as activated carbon, biomass resources, geomaterials, and industrial waste products are used to treat heavy metal-contaminated water. However, the use of construction and demolition waste (CDW) and its recycled materials to treat heavy metal-contaminated water has not been fully examined. Because the generation of CDW is increasing due to rapid urbanization and a high demand for construction, the wise use of CDW and its recycled materials is necessary. This paper reviews journal articles published from 2004 to 2016 that studied the applicability of low-cost CDW adsorbents to remove heavy metals from wastewater. In particular, it summarizes results on using CDW and recycled materials such as cement, concrete, and brick compared to those from various other adsorbents such as biosorbents, geomaterials, and their industrial products. The potential applicability of CDW and its recycled materials as heavy-metal adsorbents was examined based on a comparison table listing material properties, test conditions, and measured adsorption properties.

Keywords: Construction and demolition waste, Recycled materials, Adsorbent, Heavy metals

1. INTRODUCTION

Conventional wastewater treatment methods such as coagulation, flocculation, reverse osmosis, membrane separation, and ion exchange have long been conducted to provide better water quality and to treat wastewater before releasing it into water bodies [1]. However, these methods are expensive, and cost reduction is needed [2]. Recently, scientists have been studying using low cost and abundant adsorbents to remove heavy metals in wastewater. Natural and industrial geomaterials and biosorbents are well known sorbents, and researchers have done many experiments to identify the adsorption capacity of those materials [3].

Rapid urbanization, industrialization, and population growth in developing and developed countries produce millions of tons of construction and demolition waste (CDW) per year. As examples, Malaysia, Thailand, and Vietnam produce 1.9, 1.9, and 6.9 million tons of CDW per year, respectively, and a common treatment method is landfilling or dumping [4-5]. About 850 million tons of CDW are generated in the European Union (EU) per year, representing 31% of the total waste generated in the EU [6]. In 2014, the United States produced 534 million tons of CDW materials, and 70% of that was waste concrete [7]. Thus, researchers predict that densely populated countries like India and China will produce huge amounts of CDW materials in the future, creating many environmental problems. Therefore, investigation of the efficient usage of those abundantly available resources for different activities is timely. The objective of this review was to investigate the applicability of CDW and recycled materials as an efficient adsorbent to remove heavy metals from wastewater with respect to geomaterials and biosorbents from wastewater.

2. METHODOLOGY

A detailed literature search was carried out by selecting several key words. Those used included heavy metal adsorption, natural and industrial geomaterials, biosorbents, and CDW materials. The target medium was wastewater, and the method of adsorption was batch adsorption. The search was limited to the paper-selection criteria given in Figure 1. All papers selected were published by the Web of Science™ (Clarivate Analytics) during 2004-2016. A total of 33 references were selected of which most are journal papers from environmental engineering and material science research areas. A careful review of those selected papers was done by comparing experimental conditions, maximum adsorption capacity, and analysis methods, results for each category of adsorbents and suggesting future considerations.

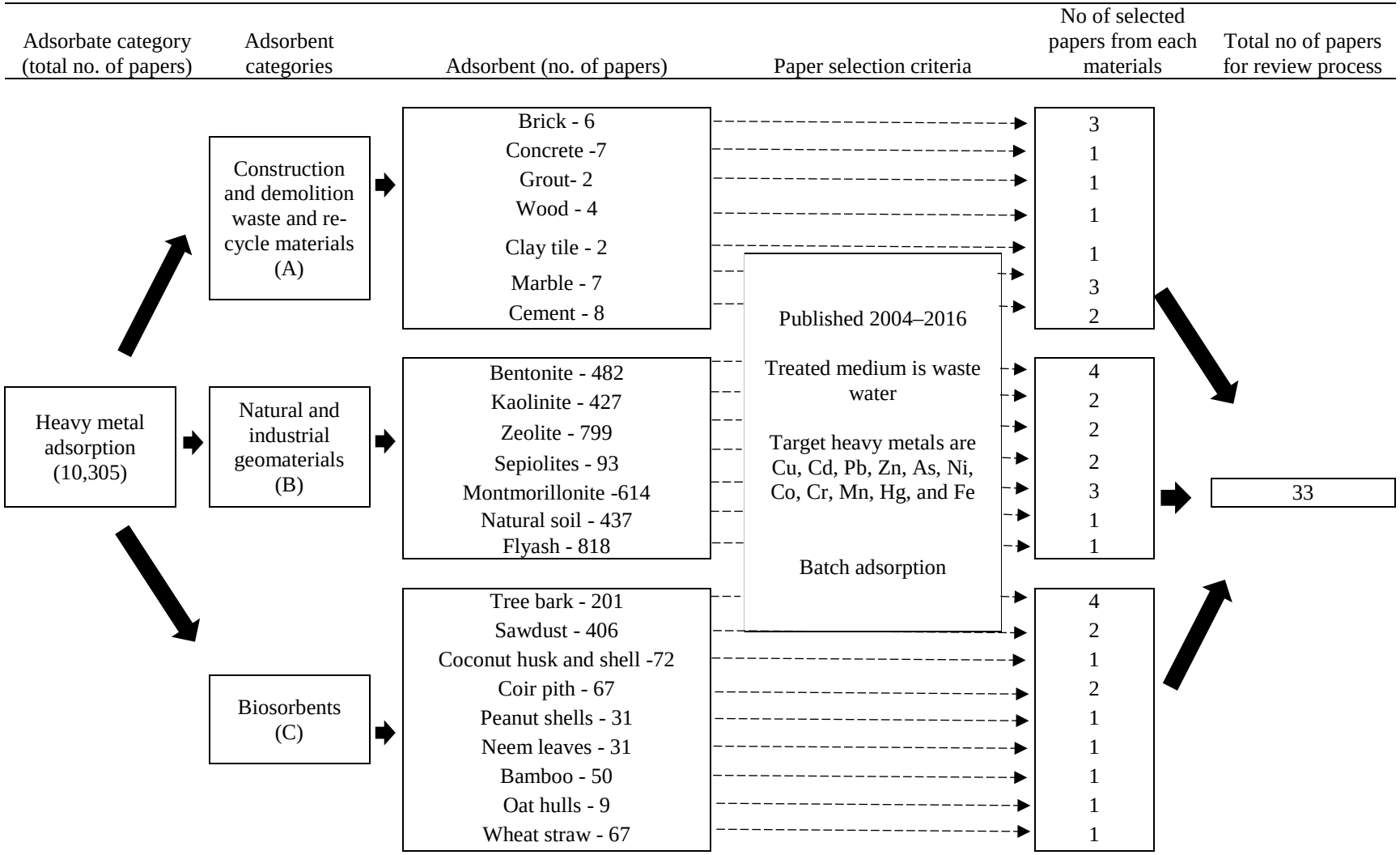

Figure 1: Procedure of journal paper selection for the review in this study

3. RESULTS AND DISCUSSION

Adsorption has been proved to be an excellent way to treat wastewater, offering significant advantages such as low-cost, availability, profitability, and ease of operation. According to Fig. 1, scientists and researchers gave highest priority to geomaterials and biosorbents (>80%), to adsorb heavy metals from wastewater in comparison to CDW and recycled materials. Furthermore, adsorption studies on concrete and cement materials are rare. Hence, investigation, comparison, and identification of material properties, test conditions, adsorption capacity, and analysis methods of various adsorption materials with respect to CDW materials are beneficial for future generations.

As shown in the Table 1, CDW [8-16], geo- [17-22] and biosorbents [23-30], major target metals including Cu, Cd (39.9%), Pb (33.3%), and Zn (24%) were studied, while less consideration was given to metals like As (12.1%), Cr, Hg, and Mn (6.1%). The particle sizes used varied for each category and metal type in reviewed papers. In more than 97% of the studies, the maximum particle size used was less than 2 mm. However, it is important to investigate the adsorption capacity of materials larger than 2 mm because in practical application powder-like materials cause higher pressure drops in filter beds. Absorbent concentration also is one of the critical parameters for higher efficiency of the adsorption process. The most common concentration was 10 g/L, and few studies used a range of concentrations to determine the optimum concentration [8,13,15] of CDW materials. Testing the effects of background electrolytes on the adsorption process also was given less consideration in each category, and common solutions were NaCl, $NaNO_3$, or $NaClO_4$ in the concentration range of 0.001–0.1 N. It is clear that CDW and recycled materials need longer equilibrium times than the other two categories of adsorbents. Basically equilibrium time depends on the target metals, adsorbent and its physical conditions, and other operation parameters. A longer equilibrium time is one of the main constraints for commercial application of those adsorbents. Therefore, further studies are needed to discover shorter equilibrium times by changing other testing conditions.

As shown in Table 2, all three categories of materials lacked data on cation exchange capacity (CEC) and point of zero charge of the materials, but these two parameters are essential to understand the adsorption potential and to explain the adsorption behavior of metals. Table 2 reveals that a wide range of initial concentrations of metal solutions was tested in the adoption studies, for example, 0.025–2000 mg/L. The selected concentrations basically depend on the target metals and wastewater solutions. Solution pH is the one of the critical parameters in adsorption experiments, and careful observation of working pH is essential for maximum adsorption, especially if adsorbent materials are concrete or cement materials [8-16]. Furthermore, when working with CDW materials, identification of the natural pH (alkaline or acidic) of CDW materials is very important because cement and concrete-like materials show high pH and brick and industrial slags show acidic pH [31]. Therefore, mixtures of those two types of materials at different ratios have the potential to provide better adsorption by controlling the optimum pH naturally. The working temperature for adsorption experiments was room temperature (25°C) in more than 80% of the studies. Therefore, study of the effects of temperature on adsorption of heavy metals on CDW materials has great potential.

As shown in Table 3, different types of mathematical models were used for data analysis in isotherm and kinetic studies. Commonly used models for isotherm studies were Langmuir and Freundlich models. For kinetic studies, pseudo first order, pseudo second order, Elovich, and intra-particle diffusion models were commonly used models. Importantly, more than 95% of studies on CDW materials used one or more models to express the adsorption behavior of heavy metals. In isotherm studies, the Langmuir model gave the best fit for more than 90% of CDW and recycled materials by representing monolayer adsorption to adsorbents. In addition, a pseudo second order model was best fitted for more than 75% of the kinetic studies. Interestingly, a few studies mentioned that the initial stage of the experiment showed a first order reaction which gradually was converted it to second order or other reaction type [8, 29].

Different types of adsorption mechanisms were reported in reviewed papers, and common mechanisms were ion exchange, diffusion (inter-particle, film, and surface diffusion), chemisorption, and inner and outer sphere complexation. It is important to investigate the adsorption mechanism of highly alkaline concrete and cement-like materials with respect to the working pH of the wastewater because heavy metals have the potential to precipitate at higher pH. Thermodynamic parameters are important to explain the adsorption process when the temperature of the medium is changed. The Gibbs free energy (ΔG) is negative for CDW and recycled materials by indicating the spontaneous nature of the adsorption process, and the negative entropy (ΔS) values suggest the probability of a favorable adsorption. In addition, only 5% of the studies in all reviewed papers carried out desorption experiments, and no studies were found for CDW materials. Also, 74% of the studies used single metal solutions, although for industrial applications, it is essential to think about binary or multiple metal solutions for adsorption studies.

Table 1: Summary of methodology of adsorption studies

Category of adsorbent	Adsorbent	Target metal	Particle size (μm)	Adsorbent concentration (g/L)	Background solution	Ionic strength (mol/L)	Equilibrium time for isotherm (min)	Ref.
A	Brick	As^{5+}	≤ 300	10-40	DIW	(-)	60	[8]
	Concrete	Cu^{2+}	63-200	5	NaCl	0-0.34	120	[9]
		Zn^{2+}	45-4760	10	DW	(-)	540-1200	[10]
		Cu^{2+}, Zn^{2+}, Pb^{2+}	≤ 250	25	DW	(-)	7200 (Cu^{2+}), >7200 (Zn^{2+}), 2880(Pb^{2+})	[11]
	Grout	Pb^{2+} ,Cd^{2+}	≤ 80	20	NaCl	0.1	90	[12]
	Wood	Cr^{6+}	1-100	1-5	DW	(-)	40	[13]
	Clay tile	Zn^{2+}	45-4760	10	DW	(-)	1200	[10]
	Marble	Cd^{2+}	25 –63	2	DDW	(-)	300	[14]
		Pb^{2+}	25 –63	1	DDW	(-)	120	[15]
		As^{5+}	50	10-40	DIW	(-)	60	[8]
	Cement	Cd^{2+}, Zn^{2+}, Cu^{2+} , Pb^{2+}	2000	40	$NaNO_3$	0.01	60	[16]
		As^{5+}	1400-3000	10-40	DIW	(-)	60	[8]
B	Bentonite	Cd^{2+} , Pb^{2+}	≤ 855	10	DIW	(-)	150 (Cd^{2+}), 250 (Pb^{2+})	[17]
	Kaolinite	Ni^{2+} , Mn^{2+}	100-500	2-10	DDW	(-)	180	[18]
	Zeolite	Pb^{2+}, Cu^{2+}, Zn^{2+} , Cd^{2+}	90-300	5	$NaClO_4$	0.01-0.05	<1440	[19]
	Sepiolite	Pb^{2+}	20-53	2-20	DW	(-)	360	[20]
	Montmo-rillonite	Co^{2+}	≤ 200	0.2-1.4	$NaClO_4$	0.001-0.1	120	[21]
	Alluvial soil	Pb^{2+} , Cd^{2+}	2000	100	MQW	(-)	Pb^{2+} 360, Cd^{2+} 60	[22]
C	Bark	Cd^{2+}	≤ 500	4	Na_2SO_4	0-0.07	25	[23]
	Sawdust	Zn^{2+}, Cu^{2+}, Cd^{2+}, Fe^{2+}, Ni^{2+} , Mn^{2+}	400-1000	2-40	DW	(-)	30	[24]
	Coconut shell	Pb^{2+} , Cd^{2+}	≤ 75	100	MQW	(-)	Pb^{2+} 360, Cd^{2+} 60	[22]
	Coir pith	Cd^{2+}	≤ 96	0.25-3	NaCl	0.001-0.1	180	[25]
	Peanut shell	Hg^{2+} , Cd^{2+}	100-200	8	DIW	(-)	120	[26]
	Neem leaves	Cr^{6+}	850-1500	4-16	DW	(-)	4020	[27]
	Bamboo	As^{3+}, As^{5+}	63-125	0.01-0.15	DDW	(-)	1800 for As^{3+}, 2130 for As^{5+}	[28]
	Oat hulls	As^{5+}	360-430	0.015	DIW	(-)	1440	[29]
	Wheat straw	As^{3+}, As^{5+}	*	0.5-3	Fe_3O_4	0.5	720	[30]

DW-distilled water, DIW- deionized water, DDW- doubly distilled water, MQW- Milli-Q water, (-)- no ionic strength, *data not available

Table 2: Summary of methodology of adsorption studies

Category of adsorbent	Adsorbent	Target metals	Point of zero charge	CEC of adsorbent (mmol/100 g)	Concentration range (mg/L)	Initial pH	Temperature (˚C)	Ref.
A	Brick	As^{5+}	*	*	0.1-1	2-9	25	[8]
		Cu^{2+}	*	*	200	1.5-6	25-45	[9]
		Zn^{2+}			40-90	2.5-4.5	25	[10]
	Concrete	Cu^{2+}, Zn^{2+}, Pb^{2+}	*	*	873(Cu^{2+}), 837 (Zn^{2+}), 1041(Pb^{2+})	5-7	20	[11]
	Grout	Pb^{2+}, Cd^{2+}	*	453.2	20 (Cd^{2+}), 200 (Pb^{2+})	5	25	[12]
	Wood	Cr^{6+}	*	*	10-50	1-11	10-50	[13]
	Clay tile	Zn^{2+}			40-90	2.5-4.5	25	[10]
	Marble	Cd^{2+}	8.3	*	20-60	7	25	[14]
		Pb^{2+}	8.3	*	500-1500	7	25	[15]
		As^{5+}			0.1-1	2-9	25	[8]
	Cement	Cd^{2+},Zn^{2+}, Cu^{2+}, Pb^{2+}	*	52.2	50-1600	3-9	25	[16]
		As^{5+}	*	*	0.1-1	2-9	25	[8]
B	Bentonite	Cd^{2+}, Pb^{2+}	*	61.0	2-500	5.2 (Pb^{2+}), 8 (Cd^{2+})	0-60	[17]
	Kaolinite	Ni^{2+}, Mn^{2+}	5.5	11.3	100-500	2-8	27-50	[18]
	Zeolite	Pb^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+}	*	*	5-20	6	25	[19]
	Sepiolites	Pb^{2+}	6.4	11.8	300	2-9	20-40	[20]
	Montmorillonite	Co^{2+}	*	86.1	9.8	3-12	30-70	[21]
	Alluvial soil	Pb^{2+}, Cd^{2+}	*	*	100-2000	2-8	25	[22]
C	Bark	Cd^{2+}	5.32	*	25-300	2-5	20-50	[23]
	Sawdust	Zn^{2+}, Cu^{2+}, Cd^{2+}, Fe^{2+}, Ni^{2+}, Mn^{2+}	*	*	5-200	5	25	[24]
	Coconut shell	Pb^{2+}, Cd^{2+}	*	*	100-2000	2-8	25	[22]
	Coir pith	Cd^{2+}	5.5	181	25-300	2-8	30	[25]
	Peanut shell	Hg^{2+}, Cd^{2+}	*	*	1-300	1-7	25	[26]
	Neem leaves	Cr^{6+}	*	*	40-700	1-11	30	[27]
	Bamboo	As^{3+}, As^{5+}	5	*	2-38	2-9	25	[28]
	Oat hulls	As^{5+}	*	*	0.025-0.2	5-9	24	[29]
	Wheat straw	As^{3+}, As^{5+}	*	*	1-28	3-11	30	[30]

*Data not available

Table 3: Summary of data analyses in adsorption studies

Category of adsorbent	Adsorbent	Target metals	Used model (isotherm study)	Used model (kinetic study)	Mechanism of adsorption	Thermodynamic parameters	Ref.
Construc-tion materi-als, CDW materials and recycle materials	Brick	As^{5+}	L, F	*	diffusion	*	[8]
		Cu^{2+}	L, F	FO,SO,ABT	film-diffusion	(-)ΔH, (+)ΔS,(-)ΔG	[9]
		Zn^{2+}	L, F, DR, T, G, RP, KC	FO, SO, E	*	*	[10]
	Concrete	Cu^{2+}, Zn^{2+} Pb^{2+}	*	*	*	*	[11]
	Grout	Pb^{2+} ,Cd^{2+}	L, F	*	*	*	[12]
	Wood	Cr^{6+}	L, F	FO, SO	chemisorption	(+)ΔH,(+)ΔS,(-)ΔG	[13]
	Clay tile	Zn^{2+}	L, F, DR, T, G, RP, KC	FO, SO, E	*	*	[10]
	Marble	Cd^{2+}	L, F, DR	BE, MWE	ion-exchange	(+)ΔH,(+)ΔS,(-)ΔG	[14]
		Pb^{2+}	L, F, DR	BE, MWE	ion-exchange	(+)ΔH,(+)ΔS,(-)ΔG	[15]
		As^{5+}	L, F	*	*	*	[8]
	Cement	Cd^{2+},Zn^{2+}, Cu^{2+}, Pb^{2+}	L	FO	chemisorption (Cu^{2+})	*	[16]
		As^{5+}	L, F	*	diffusion	*	[8]
Natural and industrial geomateri-als	Bentonite	Cd^{2+}, Pb^{2+}	L, F	FO,SO, IPD	ion exchange and intraparticle dif-fusion	(-)ΔH, (-)ΔS,(-) ΔG for Cd^{2+}, (+) ΔH, (+)ΔS,(-) ΔG for Pb^{2+}	[17]
	Kaolinite	Ni^{2+}, Mn^{2+}	L, F, T, DR	FO, SO, E, IPD	intraparticle dif-fusion	(+)ΔH, (+)ΔS,(-)ΔG	[18]
	Zeolite	Pb^{2+},Cu^{2+}, Zn^{2+}, Cd^{2+}	L, F, DKR	*	ion-exchange	*	[19]
	Sepiolites	Pb^{2+}	*	FO, SO	*	(-)ΔH, (-)ΔS, (+)ΔG	[20]
	Montmoril-lonite	Co^{2+}	L, F, DR	FO, SO	outer-sphere sur-face complexa-tion	(+)ΔH ,(+)ΔS,(-)ΔG	[21]
	Alluvial soil	Pb^{2+}, Cd^{2+}	L, F	FO, SO, IPD	*	*	[22]
Bio-sorbants	Bark	Cd^{2+}	L, F	FO, SO	*	(-)G	[23]
	Sawdust	Zn^{2+},Cu^{2+}, Cd^{2+},Fe^{2+}, Ni^{2+},Mn^{2+}	L, F	SO	ion exchange	*	[24]
	Coconut shell	Pb^{2+}, Cd^{2+}	L, F	FO, SO, IPD	*	*	[22]
	Coir pith	Cd^{2+}	L	FO	*	*	[25]
	Peanut shell	Hg^{2+},Cd^{2+}	L, F	SO	*	*	[26]
	Neem leaves	Cr^{6+}	L	FO, SO	film diffusion	*	[27]
	Bamboo	As^{3+}, As^{5+}	L, F, T	FO, SO, E, IPD	chemisorption	*	[28]
	Oat hulls	As^{5+}	L, F	FO, LDF	surface diffusion	*	[29]
	Wheat straw	As^{3+}, As^{5+}	L, F, T	*	inner-sphere complexation	*	[30]

L-Langmuir, F-Freundlich, T-Temkin, DR-Dubinin-Radushkevic, DKR-Dubinin-Kaganer-Radushkevich, RP-Redlich-Peterson, KC-Koble-Corrigan, G-Generalized model, FO-pseudo first order, SO-pseudo second order, E-Elovich , IPD-intra-particle diffusion, ABT-Adam-Bohart-Thomas, BE-Bangham equation, MWE- Morris-Weber equation, LDF-linear driving force model, ΔH-enthalpy, ΔS-entropy, ΔG-Gibbs free energy, (+) positive energy, (-) negative energy

The reported maximum adsorption capacity (Q_{max}) illustrated in Figs. 2 and 3 for selected adsorbents and all adsorbent shows a potential for adsorbing heavy metals from wastewater. It is clear that less consideration has been given to CDW and recycled materials and limited consideration for few heavy metals such as Pb, Cd, and Cu (see Fig. 1). Furthermore, brick and marble materials show more than 100 mg/g Q_{max} for Cu and Pb respectively [9,15], implying that CDW and recycled materials have great potential to adsorb heavy metals as geo- and biosorbents.

4. CONCLUSION

According to all the parameters discussed in this paper, it was clear that CDW and recycled materials have the same potential as geo- and biosorbents to adsorb heavy metals from wastewater, which is a marginalized material currently. Therefore, researchers and scientists need to focus their attention on those research gaps to effectively use abundantly available CDW materials for wastewater treatment processes in the future.

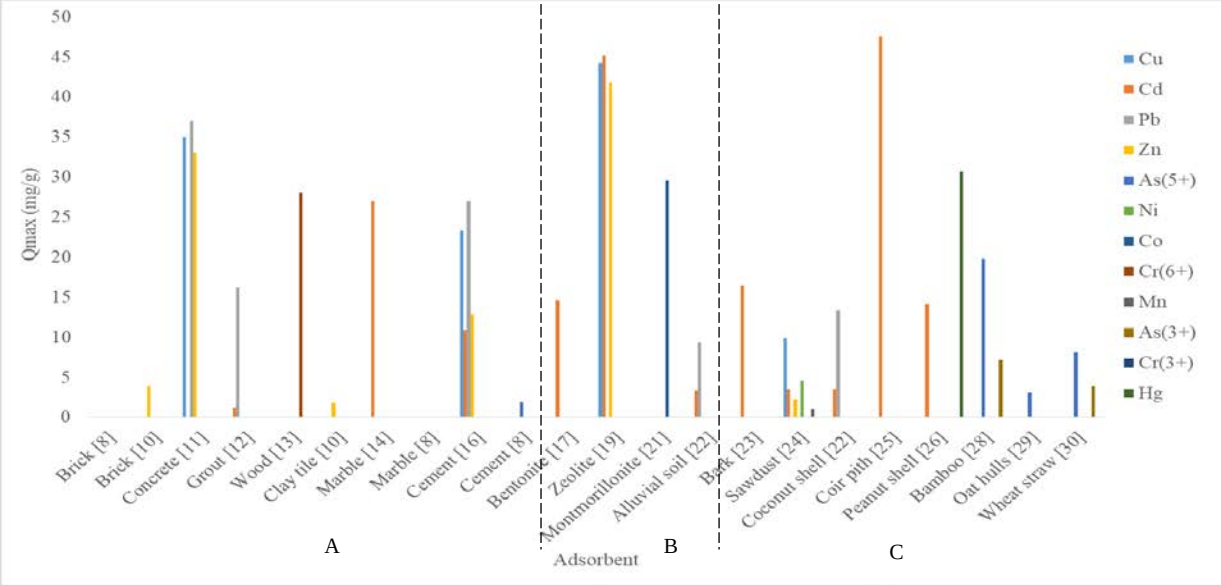

Figure 2: Reported adsorption capacities in selected adsorption studies

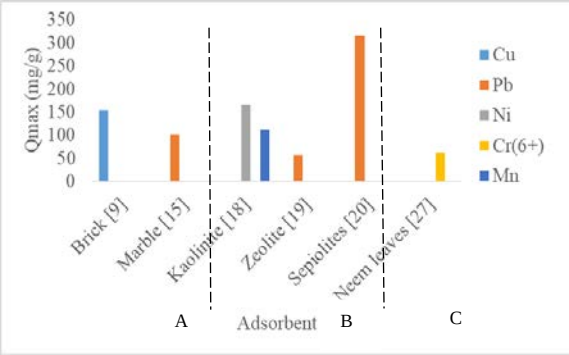

Figure 3: Reported adsorption capacities in selected adsorption studies

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