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Eco-friendly Recovery and Utilization of Innovative Industrial Adsorbents for Multi-metal Ion Removal in Aqueous Solutions

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ABSTRACT

This study investigates the ability of recovered industrial bentonite waste RIBW as a nano-, low-cost and new adsorbent to remove the ions of Pb, Cd, Cr, Zn, and Ni that have different electrochemical properties in the form of multi-metal ions from aqueous solutions.

The investigated doses of RIBW were (0.1, 0.2, and 0.3) g/100 mL for each system, and the initial concentration of each metal ions in the binary, ternary, quaternary, and quinary system was 50, 33.3, 25, and 20 ppm, respectively, where the total concentration of ions remained constant and equivalent to 100 ppm.

The maximum removal efficiency of Pb, Cd, Cr, Zn, and Ni was in the binary system with a dose of 0.3 gm of RIBW and equaled 99.7, 95.7, 99.5, 99.2, and 97.3. The minimum in the quinary system with an adsorbent mass of 0.1 g equaled 80.8, 51.1, 72.7, 71.1, and 36.6, respectively. The correlation coefficient values of R^2 for all experiments ranged from 0.91 to 1.00.

The adsorption capacity at the level of single- or multi-ion removal decreases with increasing the number of partaken metal ions in an aqueous solution, where the adsorption capacity order of metal ions in binary > ternary > quaternary > quinary system. It was evident that the electrochemical properties, such as atomic weight, ionic radius, hydrated ionic radius, and softness value have essential effects on the adsorption capacity, where the ions metal having high values of these properties was more adsorbed on the surface of RIBW adsorbent.

Keywords: Adsorption, Heavy metals, Binary, Recycling, Water treatment.

INTRODUCTION

Industrial activities release different types and elevated quantities of gases, as well as liquid and solid waste pollutants due to the development of these activities. In particular, the recycling factories of the spent engine oil are part of the industrial activities that release hazardous solid wastes as by-products. Other factories, like mining, plating, and tanning factories, release wastewater with elevated concentrations of heavy metals. All these contaminants contribute to the risks to living beings and the components of the environment (Samaka, 2018). The negative impacts of increasing heavy metals are not only on the water and soil, but they also yield damage to plants, animals, and humans. Heavy metals are hazardous due to their high toxicity even at low concentrations and they are non-biodegradable, persistent, and accumulative (Abdulaziz

& Musayev, 2017). The discharged wastewater of lots of industries has ions of more than one metal, like Pb, Cd, and Cr ions, from the painting industry (Chiamsathit & Netkrut, 2021), Ni, Pb, and Cd ions from the battery industry (Mufakhir et al., 2022), and Zn, Ni, Pb, Cd, Cr ions from the paper and pulp industry (Sharma et al., 2022).

The well-known techniques for removing heavy metals from polluted water are advanced oxidation, chemical precipitation, ion exchange, membrane separation, electrolysis, complexation, and adsorption. Adsorption is the transferring process of substances (which are called adsorbates) from the liquid phase and accumulating them at the solid surface (called adsorbents) due to physical forces, chemical reactions, or both (AL-Shauk, 2020). The adsorption process is one of the best techniques for removing metal ions from an aqueous solution due to its effectiveness, simplicity, economy, less time consumption, and high production capacity (Putro et al., 2017). The well-known adsorbents are activated carbon (Akpomie & Dawodu, 2015), clay minerals (A. Gupta et al., 2021), zeolite (Egashira et al., 2012), and industrial waste (Soliman & Moustafa, 2020). The importance of using bentonite as an adsorbent is due to its natural (Inglezakis et al., 2007), inexpensive, available, and high adsorption resulting from characteristics like high surface charge, small particle size, and high specific surface area (Karapinar & Donat, 2009). The characteristics of heavy metals that have an evident effect on the adsorption capacity are the ion radius, hydrated ions radius, softness value, atomic weight, acidity degree, and electronegativity, varying from one ion to another, where these variations lead to differences in the quantity of the adsorbed ions (Kong et al., 2020).

Numerous researchers used different forms of bentonite as an adsorbent for removing heavy metals like the chemically modified bentonite with a surfactant of (HDTMA-Br) (Castro-Castro et al., 2020), (AEAPS) (Guerra et al., 2014), a quaternary ammonium cation (QAC) (Ouellet-Plamondon et al., 2012), thermally activated bentonite at 500°C (Almeida Neto et al., 2012) and 700◦C (Cao et al., 2017). As well, there are studies conducted on the role of electrochemical characteristics in the removal efficiency of multi-metal ions *via* different adsorbents in an aqueous solution, where Sellaoui et al. (2018) studied the binary adsorption of Cd (II) and Pb (II) ions using the bentonite-chitosan composite. It is apparent that the adsorption capacity was 77.2 mg/g and 63.2 mg/g, at a temperature of 30°C, respectively. Wang et al. (2019) utilized incinerated sewage sludge ash to remove Cd (II), Cu (II), and Zn (II)) ions from contaminated waters in both single- and binary-metal systems.

Al-Zboon (2023) used multi-component adsorption to remove Hg, Cd, and As ions using nanoparticles (FeN) as an adsorbent. He achieved a removal efficiency of Cd ions with an initial concentration of 100 ppm of 59% and 58.3% for single and ternary systems, respectively. Most of the other studies utilized adsorbents that require time and cost in the preparation process, and they don't mix more than three metal ions as adsorbates. Therefore, this study aimed to research simplified materials in the preparation of low-cost, and locally available materials to achieve all assistant factors to make the adsorption process a favorable and sustainable technique. Hence, recycled industrial bentonite waste (RIBW) as an adsorbent to remove multi-heavy metals in an aqueous solution was one of the targets of sustainable development in the conservation of resources and minimization of waste in the environment. In addition, it has a lot of industrial and environmental advantages due to its inexpensiveness, high efficiency, environmental friendliness, and availability, thereby minimizing the negative impacts of hazardous solid wastes, and providing promising alternatives (Soliman & Moustafa, 2020).

The goals of this study are: (1) using the RIBW as a nano-, new, and low-cost adsorbent for removing multi-metal ions Cr (II), Cd (II), Ni (II), Zn (II), and Pb (II) in aqueous solutions in binary, ternary, quaternary and quinary batch systems (2) identifying the effects of adsorbent dose and initial concentration of heavy metals on the removal efficiency by using three quantities of RIBW (0.1, 0.2, and 0.3) gm in 100 mL of solution and four initial concentrations of metal ions (20, 25, 33.3, and 50) ppm, (3) comparing between the targeted metal ions to indicate the extent of the effect of electrochemical characteristics of metal ions on the adsorption capacity.

MATERIALS AND METHODS

Adsorbent and Adsorbates

Spent engine oil from recycling factories releases high quantities of oily saturated industrial solid waste as by-products, as shown in Fig. 1 (right) *via* thermal remediation to eliminate the residual oil and convert this industrial solid waste into safe material. The recycled industrial bentonite waste that results from thermal remediation as an adsorbent material was a reddish-white powder, as shown in Fig. 1 (left), with a high specific surface area (67.17) m^2/g and pore size (8.54)nm, and the particle diameter ranged from 18.08nm to 37.1nm. These features, in addition to chemical characteristics, are assistant factors in the adsorption of heavy metals in aqueous solutions. All details of the chemical and physical properties are shown in Shubber & Kebria (2023). The adsorbates in the experiments were Pb, Cd, Cr, Zn, and Ni. The required weight of metal ions to prepare one-liter stock solutions with a concentration of 1000 ppm from the salts is shown in Col.5 of Table 1, resulting from dividing the molecular weight of salt by the molecular weight of the metal, and then, the required concentrations of metal ions were prepared by diluting the stock solutions with distilled water for conducting the adsorption experiments.

Figure (1): (right) Oily industrial bentonite clay waste, (left) The TRBCW adsorbent in this study

Table 1. Details of preparation of heavy metal ions for the experiments

Metal ion	Mineral salts	M.W. ion (g/mol)	M.W. salt (g/mol)	Weight (g)
Pb	$PbCl_2$	207.2	278.10	1.3422
Cd	$CdCl_2.H_2O$	112.41	201.32	1.7911
Cr	$CrCl_3.6H_2O$	51.99	266.45	5.1250
Zn	$ZnCl_2$	65.38	136.29	2.0846
Ni	$NiCl_2.6H_2O$	58.69	237.69	4.0499

Adsorption Process

The well-known parameters that affect the adsorption capacity are: initial concentration of adsorbate, dose of adsorbent, contact time, temperature, and pH of the solution. The contact time affects the rate of adsorption capacity of metal ions, where the adsorption capacity is high at the first stage, then it decreases due to the decreasing number of available active sites and the rate of metal diffusion (Senberber et al., 2017). In the clay adsorbent, the adsorption is endothermic, in which the adsorption capacity increases with increasing temperature due to increasing the mobility of metal ions toward the adsorbent, then after a certain level of temperature, the adsorption process decreases due to its conversion into a desorption process (Salleh et al., 2011). A high pH value increases the precipitation of metal ions due to the negative state of the surface of the adsorbent and subsequently increases the electrostatic interaction (Héquet et al., 2001). However, a low pH value decreases the adsorption capacity of metal ions due to the protonation of the adsorbent surface, where the produced H_3O^+ ions compete with metal ions and occupy the active binding sites in the adsorbent surface (Soliman & Moustafa, 2020). Increasing the initial concentration of adsorbate increases the adsorption capacity due to elevating the probability of adsorbent collision and the high rate of diffusion of adsorbent onto the surface of the adsorbent (Uysal & Ar, 2007). Increasing the adsorbent dose increases the amount of metal ions adsorbed until a certain level and then, the effect is stopped due to increasing the number of active binding sites and total adsorbent surface area (V. K. Gupta et al., 2003).

Electrochemical characteristics are other factors that affect the preference between metal ions to the adsorbent and lead to differences in the adsorption affinity. These factors, as shown in Table 2, are: electronegativity value (Shaheen et al., 2013), hydrolysis constant (Sun et al., 2019), atomic weight (Ouyang et al., 2019), and softness (Vaca-Paulín et al., 2006) of metal ions. These factors have significant effects on the adsorption process, where increasing the values of these factors increases the preferential retention of metal by the adsorbent. Increasing hydration energies (Al-Essa & Khalili, 2018), the ionic radius (El-Bayaa et al., 2009), on the other hand, decreases the adsorption affinity of metal ions to the adsorbent.

Table 2. Electrochemical characteristics of targeted heavy metals

Ion	Acidity degree	Electronegativity (Pauling scale)	Ionic radius (Å)	Softness value	Hydrolysis constant (pK_b)	Hydration energy (K.j mol^{-1})	Hydrated ionic radius (A^o)	Atomic weight	Solubility product of metal hydroxides (pKso)*
Pb	Borderline	2.33	1.19	3.58	7.8	1481	4.01	207.2	19.9
Cd	Soft	1.69	0.95	3.04	10.1	1807	4.26	112.41	14.4
Cr	Hard	1.66	0.73		4.01*	1934	4.61	51.99	12.4
Zn	Soft	1.65	0.74	2.34	9.0—9.4	2046	4.30	65.38	16.5
Ni	Borderline	1.91	0.69	2.82	9.9	2105	4.04	58.69	

***** (Nguyen et al., 2018).

Adsorption Experiment

Adsorption experiment was conducted for binary, ternary, quaternary, and quinary batch systems for the heavy metals Pb, Cd, Cr, Ni, and Zn, as shown in Table 3 (experimental design). In a separate study from ours, pH, temperature, and contact time were determined; Therefore, the conditions of the experiments were at room temperature, 100 mL size of the aqueous solution, the concentration of 100 ppm for all different ions, the stirring speed was 300 rpm, and 30 minutes was the contact time. Three doses of RIBW were utilized in the experiment; 0.1, 0.2, and 0.3 g for identifying the effect of the dose on the adsorption capacity with heavy metals. The amount of adsorbed individual ions was calculated from Eq (1).

$$q = \frac{(C_i - C_e) * V}{m} \quad (1)$$

$$Removal\ Efficiency\ \% = \frac{C_i - C_e}{C_i} * 100 \quad (2)$$

where q (mg/g) is the adsorption capacity of RIBW for one metal ion, C_i and C_e (ppm) are the initial and equilibrium concentrations of one metal ion, respectively, V (mL) is the size of the aqueous solution, and m (g) is the weight of adsorbent (RIBW).

RESULTS AND DISSECTION

Tests of FE-SEM and FTIR

The FE-SEM (Field Emission Scanning Electron Microscopy) test is significant for the comparison by determining the extent of change in the surface of the adsorbent particles due to the attached adsorbates (heavy metals) and identifying the effect of each one of these heavy metals. Fig. (2) shows the FE-SEM test before and after adsorption of (Pb, Cd, Cr, Zn, or Ni)-containing adsorbent, respectively. It was apparent that most of the pores, openings, cavities, and ravines are filled with metal ions as soon as there are layers of metal ions on the adsorbent particles to be the essential cause in the change of shape and appearance. Also, the adsorption process affects the size of RIBW particles, where there is an evident enlargement in most of the metal ions, and the increase was from 37.17 nm to 38.7, 52.21, 41.51, and 51.74 nm for Pb, Cd, Cr, and Ni, respectively, due to the formation of a layer of metal ions on the external surface, and this is one of the adsorption mechanisms. Depending on the EDS (energy dispersive spectroscopy) analysis, the percentages of Mg and Na ions in the RIBW samples after adsorption were less than before due to the ion-exchange mechanism that plays a significant role in the adsorption process.

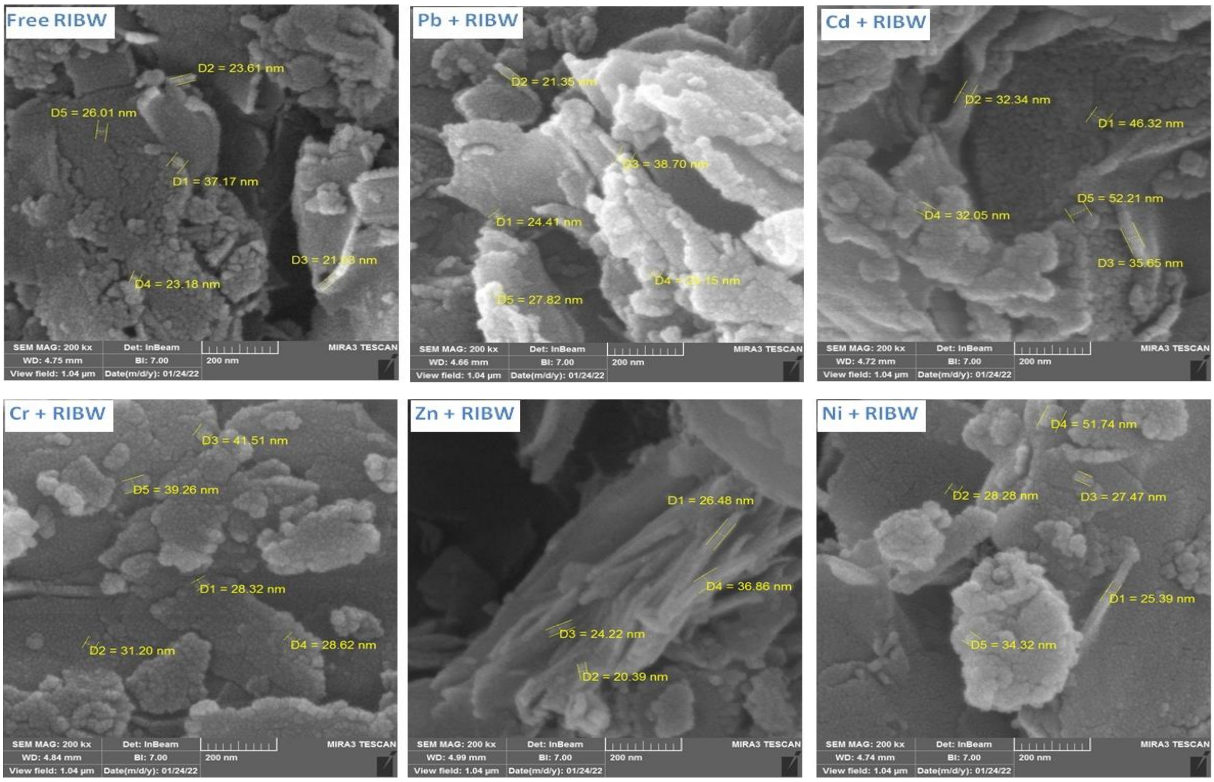

Figure (2): The FE-SEM test for RIBW adsorbent before and after heavy metal adsorption

Table 3. Details of mixture for each system

No.	**Binary** **Ratio (1:1)** **50 mL for one Ion**	**Ternary** **Ratio (1:1:1)** **33.3 mL for one Ion**	**Quaternary** **Ratio (1:1:1:1)** **25 mL for one Ion**	**Quinary** **Ratio (1:1:1:1:1)** **20 mL for one Ion**
1	Cr + Cd	Cr + Ni + Zn	Cr + Cd + Ni + Zn	Cr + Cd + Zn + Pb + Ni
2	Cr + Ni	Cr + Ni + Cd		
3	Cr + Zn	Cr + Ni + Pb	Cr + Cd + Ni + Pb	
4	Cr + Pb	Cr + Zn + Cd		
5	Cd + Ni	Cr + Zn + Pb	Cr + Cd + Zn + Pb	
6	Cd + Zn	Cr + Cd + Pb		
7	Cd + Pb	Cd + Ni + Pb	Cr + Zn + Ni + Pb	
8	Ni + Zn	Cd + Ni + Zn		
9	Ni + Pb	Cd + Zn + Pb	Cd + Ni + Zn + Pb	
10	Zn + Pb	Ni + Zn + Pb		

FTIR (Fourier-Transform Infrared Spectroscopy) test is a significant method to determine the change in the chemical function of the RIBW adsorbent after adsorption of a single metal ion, where it is apparent in Fig. (3) that the transmittance before adsorption is less than after adsorption due to the presence of the heavy metals on the surface of the RIBW adsorbent. Moreover, there are variations in the transmittance between the Pb, Cd, Cr, Zn, and Ni-loaded RIBW adsorbent, where the maximum transmittance is for the Pb-loaded RIBW and the minimum one is for Ni-loaded RIBW, which is compatible with adsorption capacity by the RIBW adsorbent, where the ions of Pb are the most adsorbing, while the Ni ions are the less adsorbing, compared to another metal ions, and then, the transmittance value depends on the adsorbed quantity of a metal ions by the RIBW adsorbent. Also, from Table 4, there are some of the absorption peaks of RIBW that were shifted upwards and downwards when adsorbing one of the targeted metal ions by the hydroxyl and Si-O groups of RIBW as mentioned in (Samaka, 2018; Shubber & Kebria, 2023), including the peaks at (3407, 1505, 1050, 794, and 569) cm^{-1}. However, there are no shifts in the absorption peaks of RIBW for the peaks at (1959, 1737, 1652, 1558, 1538, 1456,1404, and 1404) cm^{-1}. The interpretations of the absorbing peaks in Table 4 depend on the results reported in (Karapinar & Donat, 2009; Mohammed et al., 2018; Fatiha & Belkacem, 2016; Nwosu et al., 2018; Ma et al., 2016; Ritz et al., 2011; Aguilar et al., 2020; Paluszkiewicz et al., 2008; Amari et al., 2018; Eloussaief et al., 2011).

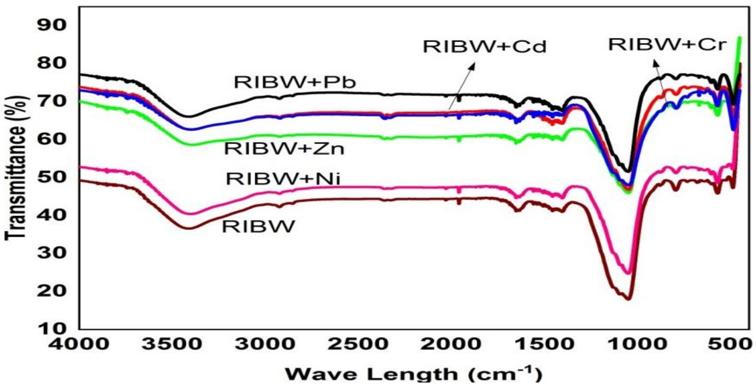

Figure (3): The results of the FTIR test for RIBW adsorbent before and after adsorbing the heavy metals

Table 4. the absorption peaks of FTIR test for RIBW before and after adsorption of heavy metals

Wave number (cm^{-1})						Functional groups
RIBW Before adsorption	RIBW After adsorption for ions of					
	Pb	Cd	Zn	Cr	Ni	
3407	3401	3398	3392	3396	3401	Stretching vibration of H-O-H
1959	1959	1959	1959	1959	1959	OH-bonding
1733	1733	1733	1733	1733	1733	C=O bonding
1652	1652	1652	1652	1652	1652	OH− deformation or H-OH bending vibration
1558	1558	1558	1558	1558	1558	the stretching of C−N, out-of-plane vibration
1456	1456	1456	1456	1456	1456	alkyl CH deforming
1404	1404	1404	1404	1418	1404	carbonate's presence as impurities
1050	1049	1047	1047	1047	1050	Stretching vibrations Si-O-Si
794	793	791	794	789	794	Si-O stretching due to the presence of quartz and kaolin
678	----	668	668	668	----	combined Al-O and Si-O bending vibration
569	569	569	568	571	568	deformation vibrations of Si-O-Al
486	484	484	488	484	487	deformation vibrations of Si-O-Si or Si-O-Mg

Binary Adsorption

Three doses of RIBW were utilized (0.1, 0.2, and 0.3 g) for removing binary metal ions with a mixing volume ratio of 50:50 mL per sample with an initial concentration of 100 mg/L of the aqueous solution, as shown in Table 3. Table 5 contains the removal efficiency and the maximum adsorption capacity for each metal ions of the binary mixing, where it was apparent that the high removal efficiency was for the dose of 0.3g, while the maximum adsorption capacity was for the RIBW adsorbent dose of 0.1g, and the high removal efficiency was for the Pb ions, ranging from (79.1 to 88.6)% and from (99.5 to 99.9)% for the dose of 0.1 and 0.3g, respectively, based on the type of the partaken metal ions, while the minimum removal efficiency was for Ni ions, ranging from (53.8 to 65.3)% and (93.4 to 100)% for the dose of 0.1g and 0.3 g, respectively, where the maximum removal efficiency was when mixing with Pb ions. The maximum total adsorption capacity was from mixing the metal ions of Pb and Cd at 80.8 mg/g, indicating that these metal ions have more affinity to RIBW adsorbent, while the minimum adsorption capacity was from mixing (Cr and Ni) ions at 61.6 mg/g.

From Fig.4 (right), it is apparent that the increase of RIBW dose from 0.1 to 0.3 g increases the average removal efficiency due to a rise in the number of active sites in the adsorbent of RIBW, where the removal efficiency of Pb, Cd, Cr, Zn, and Ni increased from (85.23 to 99.68)%, (72.79 to 95.69)%, (64.90 to 99.53)%, (65.33 to 99.20)%, and (59.21 to 97.32), respectively. Also, it was noticeable that the average removal efficiency values were close to each other, except that the average removal efficiency value for lead (Pb) ions is very high for all dose value. The maximum adsorption capacity qm of metal ions in binary mixture alters according to the type of the partaken metal, where it is apparent in Fig. 4 (left) that the ions are more adsorbed when mixing with the lead Pb due to the lower effect of competition. The results were compatible with other studies that showed the affinity of Pb more than Cd to the adsorbent of the *Carpobrotus edulis* plant (Chiban et al., 2011). Further, the uptake of incinerated sewage sludge ash for Cd is more than for Zn (Wang et al., 2019). In another study conducted by Etci et al. (2010) using beidellite clay to remove Pb and Cd ions in binary adsorption, it was evident that the adsorption capacity of Pb and Cd was 50mg/g and 30mg/g, respectively, and it was apparent that the metal ions have less affinity to the adsorbent compared with a single state. From Fig. 4, it was apparent that the adsorption capacity of metal ions was high when mixed with Pb. The difference in the obtained adsorption capacity was due to the variation of the electrochemical characteristics between targeted metal ions, as previously explained.

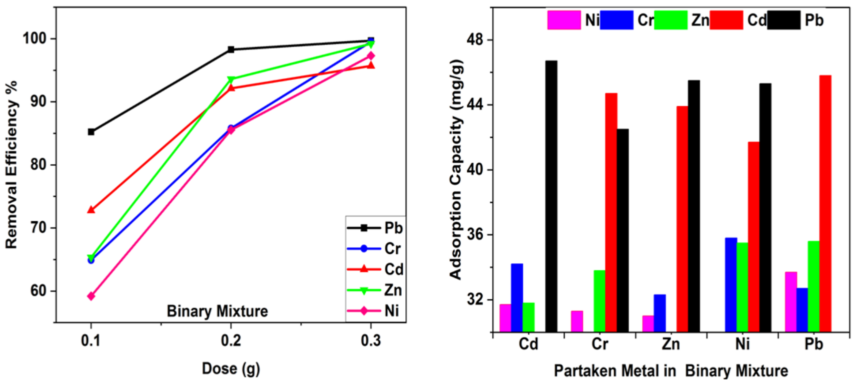

Figure (4): The removal efficiency and adsorption capacity of metal ions in binary mixture

Table 5. Binary Adsorption (50 ppm Ion1: 50 ppm Ion2)

Ion 1						Ion 2						Total
Dose (g)	**0.1**	**0.2**	**0.3**	**q_m (mg/g)**	**R^2**	**Dose (g)**	**0.1**	**0.2**	**0.3**	**q_m (mg/g)**	**R^2**	**q_m (mg/g)**
Ion1	**Removal Ratio (R) %**					**Ion2**	**Removal Ratio (R) %**					
Cr	65.6	87.0	99.7	32.8	0.953	Cd	67.3	90.0	93.9	33.6	0.998	66.4
Cr	69.3	86.6	99.8	34.7	0.914	Ni	53.8	79.3	93.4	26.9	0.980	61.6
Cr	61.4	83.0	98.8	30.7	0.944	Zn	64.6	93.1	99.3	32.3	0.996	63
Cr	63.2	86.6	99.9	31.6	0.964	Pb	79.1	96.3	99.5	39.6	0.991	71.2
Cd	70.2	91.1	97.0	35.1	0.990	Ni	61.2	90.3	99.6	30.6	0.992	65.7
Cd	63.5	87.8	91.9	31.7	0.990	Zn	60.9	93.3	98.7	30.5	0.999	62.2
Cd	90.2	99.7	100.0	45.1	1.000	Pb	87.5	98.2	99.8	43.7	0.991	88.8
Ni	56.6	79.7	96.3	28.3	0.955	Zn	66.9	93.7	98.8	33.5	0.997	61.8
Ni	65.3	92.8	100.0	32.7	0.992	Pb	85.7	98.9	99.6	42.9	1.000	75.6
Zn	68.8	94.3	100.0	34.4	0.992	Pb	88.6	99.6	99.9	44.3	1.000	78.7

Ternary Adsorption

In ternary adsorption, three metal ions are used with a mixing volume ratio of 33.3: 33.3: 33.3 mL per sample as shown in Table 3, with the initial concentration of 100mg/L of the aqueous solution for three doses of 0.1, 0.2, and 0.3 g. Table 6 includes the removal efficiency for the RIBW adsorbent dose of 0.1 g to be the difference between the removal efficiency results noticeable for the targeted metal ions and the ease of the comparison between them, where it was apparent that the removal efficiency for the dose of 0.1g ranged from (65.1 to 96.3)%, (56.9 to 64.0)%, (69.1 to 72.5)%, (62.5

to 74.9)%, and (41.1 to 60.2)% and the adsorption capacity ranged from (21.7 to 32.1), (18.9 to 21.3), (23.0 to 24.5), (20.8 to 24.9), and (13.7 to 20.1) mg/g for Pb, Cd, Cr, Zn, and Ni, respectively, and these ranges in the removal efficiency are due to the types of the partaken metal ions, causing the increase or decrease in the removal efficiency for the targeted metal ions. Also, it was apparent that the higher removal efficiency was for the Pb ions and the lower for the Ni ions due to the electrochemical characteristics of the metal ions as a result of the reaction between the mixed metal ions. The maximum total adsorption capacity was from mixing the metal ions of (Pb, Cd, and Zn) at 73.4 mg/g and these metal ions have more affinity to the RIBW adsorbent, while the minimum adsorption capacity was from mixing (Cr, Ni, and Cd) ions at 59 mg/g. These results are compatible with the results of the binary system.

Table 6. Ternary adsorption (33.3 ppm ion 1: 33.3 ppm ion 2: 33.3 ppm ions 3)

		Ion 1				Ion 2				Ion 3		Total
Ion1	R %	q_m (mg/g)	R^2	Ion2	R %	q_m (mg/g)	R^2	Ion3	R %	q_m (mg/g)	R^2	q_m (mg/g)
Cr	72.5	24.1	0.996	Ni	41.1	13.7	0.995	Zn	65.8	21.9	0.998	59.7
Cr	71.8	23.9	1.000	Ni	43.9	14.6	0.992	Cd	61.6	20.5	0.993	59.0
Cr	73.5	24.5	1.000	Ni	44.4	14.8	0.995	Pb	76.3	25.4	1.000	64.7
Cr	70.1	23.3	0.981	Zn	65.6	21.8	0.993	Cd	61.4	20.4	0.991	65.5
Cr	69.1	23.0	1.000	Zn	62.5	20.8	0.993	Pb	65.1	21.7	1.000	65.5
Cr	71.4	23.8	0.999	Cd	61.4	20.4	0.991	Pb	72.7	24.2	1.000	68.4
Cd	64.0	21.3	0.991	Ni	60.2	20.1	0.993	Pb	93.6	31.2	0.998	72.6
Cd	56.9	18.9	0.996	Ni	42.3	14.1	0.991	Zn	63.1	21.0	0.997	54.0
Cd	59.4	19.8	0.992	Zn	65.8	21.9	0.993	Pb	95.3	31.7	0.998	73.4
Ni	45.8	15.3	0.992	Zn	74.9	24.9	0.994	Pb	96.3	32.1	0.998	72.3

The removal efficiency elevated with increasing the dose of the adsorbent RIBW, where with the increase in the RIBW doses from 0.1 to 0.3g, as shown in Fig.5 (right), the removal efficiency raised from (77.3 to 99.7) %, (60.5 to 97.8)%, (71.0 to 99.9)%, (67.4 to 99.7)%, and (48.2 to 99.2)% for Pb, Cd, Cr, Zn, and Ni, respectively, due to increasing the active sites for adsorbing metal ions. It was apparent that the RIBW is more adsorbing to Pb ions and less adsorbing to Ni ions, while Cr, Cd, and Zn ions are close to one another.

Also, it was notable that the range of adsorption capacity of Cr and Pb produced from the reaction with other metal ions was high, as shown in Fig.5 (left). This difference in the adsorption behavior for self-metal may belong to the electrochemical characteristics of the partaken metals. Many researchers investigated the ternary adsorption of heavy metals, and some of them explained that the affinity order was Cu (II) > Cd (II) > Ni (II) on the boron nitride (Han et al., 2020), while the affinity order to the tourmaline was Cd (II) > Zn (II) > Ni (II) (Liu et al., 2013), where the results of both studies were compatible with the results of this study.

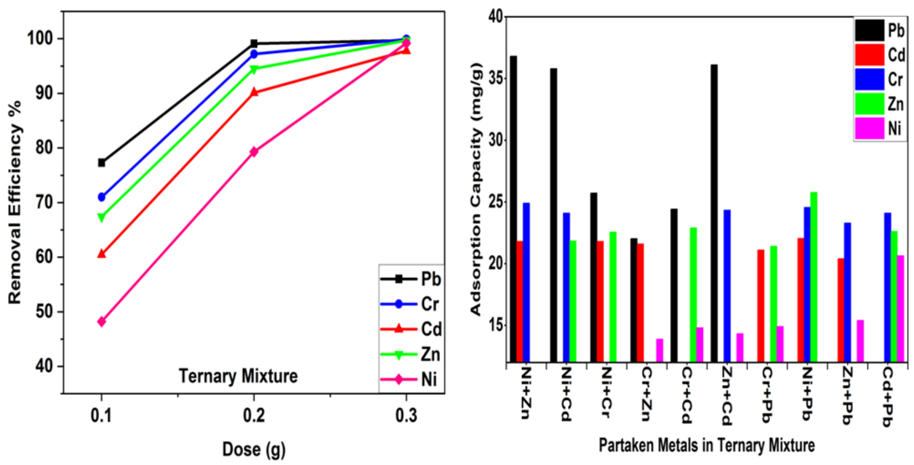

Figure (5): The removal efficiency and adsorption capacity of metal ions in ternary mixture

Quaternary Adsorption

Quaternary adsorption involves mixing a four-volume ratio of metal ions (25 mL per metal ion), as shown in Table 3. The initial concentration of each metal ions was 100 ppm using three RIBW doses of 0.1, 0.2, and 0.2 g. Table 7 shows the obtained results of the removal efficiency and the maximum adsorption capacity, where the removal efficiency result from using the adsorbent dose of 0.1g ranged from (76.0 to 86.9)%, (53.7 to 56.4)%, (69.7 to 72.7)%, (64.6 to 80.9)%, and (39.1 to 40.6)% and the adsorption capacity ranged from (19.0 to 21.7), (13.4 to 14.1), (17.4 to 18.2), (16.1 to 20.2), and (9.8 to 10.2) mg/g for Pb, Cd, Cr, Zn, and Ni, respectively, and this variation in the removal efficiency and adsorption capacity is due to the type of the partaken metal ions, where the electrochemical characteristics have an evident effect on the adsorption process, because of the nature and strength of the competition between the metal ions. The variation between the maximum and minimum adsorption capacity resulting from mixing different metal ions in a quaternary system is slight, where the maximum is 68.1 mg/g resulting from the mixing of (Pb, Cr, Zn, and Ni) and the minimum is 61.6 mg/g resulting from the mixing of (Cd, Zn, Cr, and Ni).

Table 7. Quaternary adsorption for RIBW dose of (0.1 g) (Ion 1: ion 2: ion 3: ion 4 with (25: 25: 25: 25 ppm)

Ion 1				Ion 2				Ion 3				Ion 4				Total
Ion 1	R%	q_m mg/g	R^2	Ion2	R%	q_m mg/g	R^2	Ion 3	R%	q_m mg/g	R^2	Ion 3	R%	q_m mg/g	R^2	qm mg/g
Cr	69.7	17.4	1.000	Cd	55.2	13.8	0.996	Ni	40.6	10.2	0.996	Zn	80.9	20.2	0.994	61.6
Cr	72.7	18.2	0.998	Cd	56.4	14.1	0.993	Ni	40.6	10.1	0.996	Pb	86.8	21.7	1.000	64.1
Cr	70.3	17.6	0.999	Cd	53.7	13.4	0.996	Zn	64.6	16.1	0.993	Pb	76.0	19.0	0.999	66.1
Cr	72.3	18.1	1.000	Zn	80.8	20.2	0.998	Ni	40.1	10.0	0.995	Pb	79.1	19.8	1.000	68.1
Cd	54.2	13.6	0.995	Ni	39.1	9.8	0.995	Zn	72.9	18.2	0.997	Pb	86.9	21.7	0.999	63.3

From Fig. 6 (right), it is apparent that the average removal efficiency increased with increasing the dose of RIBW adsorbent, so raising the RIBW dose from 0.1 g to 0.3 g improved the removal efficiency from (82.2 to 99.2)%, (54.9 to 98.0)%, (71.3 to 99.9)%, (74.8 to 97.3)%, and (40.1 to 97.3)% for Pb, Cd, Cr, Zn, and Ni, respectively, due to the increase in the mass of the dose, which increases the adsorbing active site and then elevates the opportunity of the removal. The adsorption capacity for each metal ions and the effect of the partaken metal ions in the quaternary system are reported in Fig. 6 (left), which shows an affinity order of Pb (II) > Zn (II) > Cr (II) > Cd (II) > Ni (II). The affinity order in the quaternary adsorption was compatible with other studies, which approved that Pb (II) ions in the quaternary mixture have more affinity than other metal ions to perlite (Vijayaraghavan et al., 2009), while other researchers utilized the natural and activated clay and approved the affinity order of Pb (II)> Cu (II) > Zn (II) > Cd (II) (Khalfa et al., 2020).

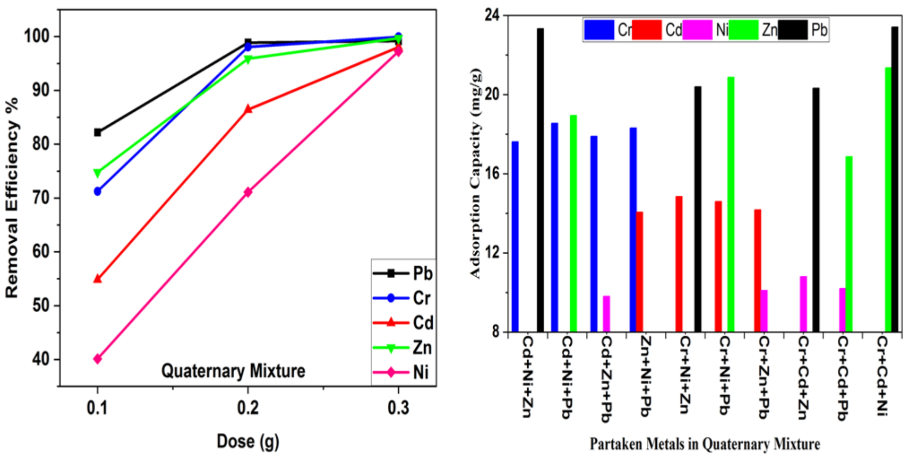

Figure (6): The removal efficiency and adsorption capacity of metal ions in quaternary mixture

Quinary Adsorption

Quinary adsorption involves mixing all the targeted metal ions of Pb, Cr, Cd, Zn, and Ni in one sample with a five-volume ratio (20 ml per each metal ions), and then, the total summation of volume was 100 mL, as shown in Table 3, where the initial concentration of each one was 100 ppm. From the results in Table 8 and Fig.7 (right), the removal efficiency was increased by raising the adsorbent dose from 0.1g to 0.3g, which improves the removal efficiency from (80.8 to 99.2) %, (72.7 to 99.8)%, (71.1 to 99.9)%, (51.1 to 98.2)%, and (36.6 to 97.4)% and decreases the adsorption capacity from (16.2 to 6.6), (14.5 to 6.7), (14.2 to 6.7), (10.2 to 6.5), and (7.3 to 6.5) mg/g for Pb, Cr, Zn, Cd, and Ni, respectively. According to the adsorption results, the affinity order of the metal ions to RIBW adsorbent was Pb > Cr > Zn > Cd > Ni, as shown in Table 8 and Fig 7 (left). These results are consistent with those of other studies, like (Unuabonah et al., 2016) that utilized modified kaolinite with geothite-humic acid as an adsorbent to remove metal ions in the quinary system with the affinity order Pb > Cu > Zn > Cd > Ni. Also, the adsorbent of the abies bornmulleriana was investigated in the quinary adsorption system to remove Pb, Cu, Cd, Ni, and Co ions, and the result of the selectivity order was Pb > Cu > Cd > Ni > Co, amounting to 8.5, 5.6, 3.5, 2.2, and 1.9 mg/g, respectively (Oguz, 2020).

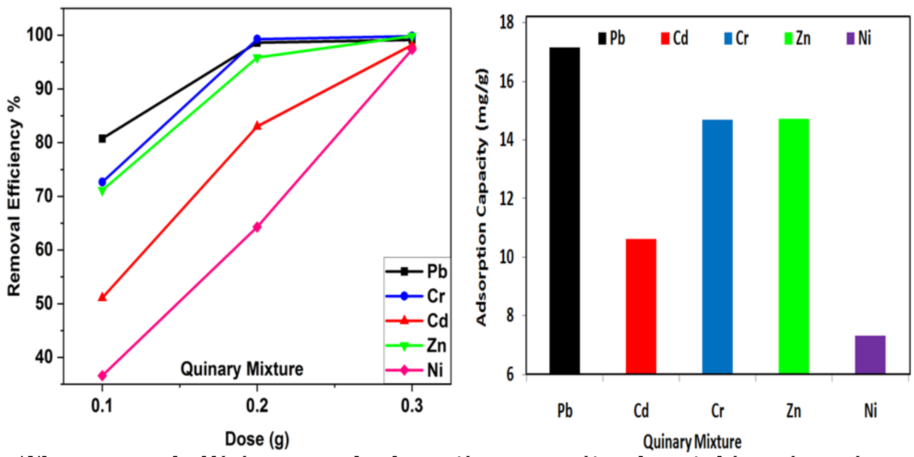

Figure (7): The removal efficiency and adsorption capacity of metal ions in quinary mixture

Table 8. Quinary adsorption (Ion 1: ion 2: ion 3: ion 4: ion 5 = (20:20:20:20:20 ppm)

Ions	Adsorption Capacity q (mg/g)			R^2	Removal Efficiency %		
	Dose =0.1 g	Dose =0.2 g	Dose =0.3 g		Dose =0.1 g	Dose =0.2 g	Dose =0.3 g
Pb	16.2	9.9	6.6	1.000	80.8	98.6	99.2
Cr	14.5	9.9	6.7	1.000	72.7	99.3	99.8
Cd	10.2	8.3	6.5	0.994	51.1	83.0	98.2
Zn	14.2	9.6	6.7	0.996	71.1	95.9	99.9
Ni	7.3	6.4	6.5	0.992	36.6	64.3	97.4
Total	62.4	44.1	33.0				

Effect of Initial Single Concentration

The concentration of each metal ions in binary, ternary, quaternary, and quinary systems is 50, 33.3, 25, and 20 ppm, respectively, where the final total weight of different metal ions was constant, amounting to 100 ppm for each system. Although the total concentration is equal for all systems, the adsorption capacity of ions lessened with increasing the number of metal ions in an aqueous solution. It was apparent that increasing the number of the partaken metal ions decreases the total maximum adsorption capacity (qt), where the (qt) of the binary, ternary, quaternary, and quinary systems is equal to 88.8, 73.4, 68.1, and 62.4, respectively. Increasing the number of ions in the aqueous solution increases the competition, noting that elevating the electrostatic repulsion between them decreases the adsorption capacity for these ions (Khalfa et al., 2020). Moreover, the adsorption capacity of the alone metal ions decreased with increasing the number of the competing metal ions, as shown in Fig.8, where the average adsorption capacity in binary, ternary, quaternary, and quinary systems for Cr was 33.8, 24.2, 18.1, and 14.7, for Cd was 44, 21.5, 14.4, and 10.6 mg/g, for Zn was 34.2, 22.8, 19.5, and 14.7 mg/g, for Ni was 31.9, 15.7, 10.2, and 7.3 mg/g, and for Pb was 45, 30.1, 21.9, and 17.2 mg/g, respectively. Also, it was apparent that the affinity order varied according to the number of metal ions, where the affinity of Cd in the binary system was high and reduced in the quinary mixture due to the electrochemical characteristics, such as electronegativity, ionic radius, softness, hydrolysis constant, hydration energy, and hydrated ionic radius.

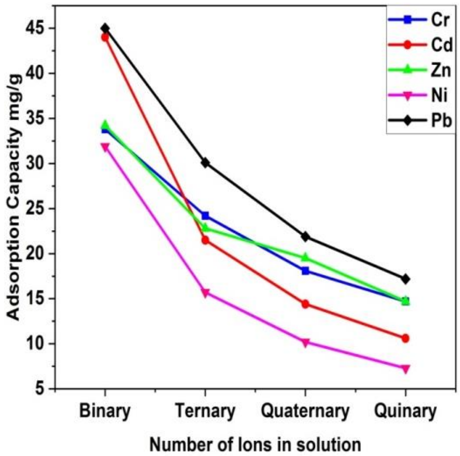

Figure (8): The effect of the number of the partaken metal ions on the adsorption capacity

Effect of Electrochemical Characteristics

From the adsorption experiment data, it was apparent that the ions having the highest values of atomic weight, ionic radius, hydration energy, and softness value are more adsorbing on the adsorbent RIBW surface, where Pb was more adsorbing due to it having the highest value of these electrochemical characteristics. Also, it was apparent that the hard ions, like chromium and nickel ions, had a lower adsorbing capacity than soft ions, where maximum adsorbing of the Cr ions in the binary system occurs when mixing with Ni, while minimum adsorbing occurs for the Ni ions when mixing with Cr. In the ternary system, it was apparent that the third ion (Cd, Zn, or Cr) partaken with (Ni+Pb) is more adsorbing due to ions of Pb and Ni having extremely opposite electrochemical characteristics, then yielding less competition to the partaken ions. In contrast, the third ion is lower adsorbing when mixed with (Cr + Zn) due to the similarity of these ions in characteristics, like atomic weight, ionic radius, and hydration energy, then creating high competition for the third ion on the RIBW surface. In the quaternary system, the variation in the adsorption capacity of the same ions was low due to the increase in the number of partaken ions that led to less difference in properties, where the ranking of the ions' adsorption capacity was similar to the ranking of the ions in the quinary system.

CONCLUSIONS

The recycling factories of spent engine oil produce a high amount of oil-saturated bentonite clay as a by-product. The final production of thermal remediation for this oily industrial bentonite clay waste was a nano-, reddish-white powder called in this study "recycled industrial bentonite waste (RIBW)" to be a new, green, efficient, low-cost, available, and eco-friendly adsorbent. Significant conclusions from the experimental results in the batch adsorption system for multi-component aqueous solution were:

- Metal ions with high values of electrochemical properties, such as atomic weight, ionic radius, hydrated ionic radius, and softness, were more adsorbed on the surface of RIBW adsorbent, like Pb ions and opposite as Ni ions.
- -RIBW was a cheap, new, and effective adsorbent of multi-component removal from aqueous solutions.
- -The removal efficiency decreases with increasing the number of metal ions in an aqueous solution, where the affinity order of ions is in binary > ternary > quaternary > quinary systems due to the complicated interaction and the competition between the partaken ions.
- Increasing the RIBW adsorbent dose increases the removal efficiency and decreases the adsorption capacity (mg/g) due to the steadiness of the adsorbates and rising the active sites of the RIBW adsorbent.
- Pb ions were the most removed, and Ni ions were the least removed, due to the electrochemical characteristics of the ions, while the uptake of cadmium Cd was varied, where its uptake decreases with increasing the number of partaken ions in the aqueous solution compared with other metal ions.

Recommendations

- Studying the capability of using prepared TRBW to remove other heavy-metal ions and organic pollutants, such as phenolic compounds, dyes, and furfural from aqueous solutions in batch and continuous-flow systems.

- Extending this work to study the competitive adsorption in fixed beds in binary and ternary systems.
- Studying the adsorption of heavy metal ions onto TRBW in a fluidized bed reactor.
- Studying removal of multi-metal ions from an aqueous solution using a new, green adsorbent resulting from recycling the industrial bentonite clay waste.
- Studying the adsorption process *via* different metal ions with multiple systems, such as binary, ternary, quaternary, and quinary systems.
- Studying the effect of electrochemical characteristics, like atomic weight, ionic radius, hydration energy, and softness value, on the adsorption capacity.

Conflict of Interests

The authors declare that they have no known or potential competing financial interests or personal relationships that could have influenced the work reported in this paper.

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