

Research Article

Valorization of Wasted Black Tea as a Low-Cost Adsorbent for Nickel and Zinc Removal from Aqueous Solution

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Characteristics and efficiency of wasted black tea (WBT) were investigated as a low-cost sorbent in removal of Ni^{2+} and Zn^{2+} ions from aqueous solution. Initial findings showed WBT potential to be applied as an effective sorbent due to high concentrations of carbon and calcium and high porosity and availability of functional groups. Sorption dynamics were studied with varying pH, contact time, and adsorbent dose. Maximum percentages of metal ions removal were recorded at pH 5, contact time 250 min, and 20 g/L of adsorbent concentration. Binary metal sorption studies showed that Ni^{2+} and Zn^{2+} do not compete with each other for available sorption sites, so the adsorption trend in binary system appears similar to monocomponent metal adsorption. Evaluation of the isotherms confirmed that WBT has high value of adsorption capacity. Sorption data fitted well with both Freundlich and Langmuir models. In the optimum conditions, maximum capacity of WBT could reach up to 90.91 mg-Ni/g adsorbent and 166.67 mg-Zn/g adsorbent. This experiment demonstrated the ability of tea waste as an effective, sustainable, and low-cost adsorbent for removal of the heavy metal ions.

1. Introduction

Nickel (Ni^{2+}) is an essential element which is desired for the human health in small amount. However, at high concentrations or prolonged exposures, it can cause health problems such as allergy, blood and heart disorders, chronic bronchitis, and even cancer [1]. Zinc (Zn^{2+}) is also an essential micronutrient, but too much of zinc intake causes acute and chronic toxicity. Based on the Environmental Protection Agency (EPA) guidelines the maximum concentrations of Ni^{2+} and Zn^{2+} allowed in drinking water are 0.1 and 5 ppm, respectively [2]. Ni^{2+} and Zn^{2+} are among the most common heavy metals in the wastewater of the industries such as electroplating, electronics, batteries, and metal treatment and fabrication. Electroplating industry is one of the major sources of nickel and zinc pollutions. It is reported that the initial Ni^{2+} and Zn^{2+} concentrations in a typical electroplating industry range from 20 to 120 mg/L [3].

Unlike the majority of organic pollutants, trace elements cannot be subjected to degradation into harmless products through biological processes. So, they can be a potential

threat for living organisms due to their bioaccumulation characteristics [4]. Hence, it is required to remove the harmful heavy metals from the contaminated discharges using other methods. Many techniques such as chemical precipitation, electrochemical treatment, ion exchange, filtration, reverse osmosis, and solvent extraction have been employed for treatment of wastewater containing heavy metals [5]. However, most of these conventional techniques are often non-eco-friendly and very costly especially when the heavy metals concentration is in the range of 10–100 mg/L [6, 7]. The adsorption of contaminants is adopted as an efficient and promising technique comparable to the current methods in decrease of heavy metals concentrations to acceptable levels. Adsorption is considered as an economically effective treatment method, if low-cost adsorbents are utilized. Data in Table 1 compare the efficiencies of the sorption technique with some common methods used in removal of Ni^{2+} and Zn^{2+} from wastewater.

The commonly used adsorbents are material containing organic carbon (such as activated carbon, biochar, and plant residues) and many inorganic chemicals and clay [8].

TABLE 1: Efficiencies of the current methods and sorption technique in removal of Ni^{2+} and Zn^{2+} .

Technique	Removal efficiency (%) [*]		Reference
	Ni^{2+}	Zn^{2+}	
Chemical precipitation	98.4	99	[13, 14]
Electrochemical	98	96	[15, 16]
Ion exchange	93.6	100	[17, 18]
Filtration	99.1	98	[19, 20]
Reverse osmosis	99.3	98.9	[21]
Adsorption	82	99.8	[17, 22]

^{*}Initial metals concentrations of less than 200 mg/L.

The necessity of finding inexpensive, renewable, and more effective adsorbents encourages the investigation of different materials. In recent years, a wide range of studies has been conducted to remove heavy metal ions from aqueous environments using sorption methods with different materials. The utilization of agricultural wastes as sorbent can promote the decrease of environmental problems related to green wastes management and their disposal. A vast variety of green wastes have been evaluated for sorption studies such as rice husk [9], wheat bran [10], corn cobs [11], tree leaves and barks [12], and aquatic weeds [7].

Thousands of tons of wasted black tea are produced and disposed unutilized in tea shops, restaurants, and houses every day. Wasted black tea is resistant to biodegradation and becomes a matter for disposal as waste. The main constituents of tea leaves are cellulose, hemicelluloses, lignin, tannins, and proteins. The functional groups in these compounds are mainly hydroxyl, oxyl, aromatic carboxylate, amino, sulfonic, and phenolic groups which promote the physicochemical interactions for substances sorption including heavy metals [23]. There are some studies on use of factory tea waste as adsorbent [24–26]. But there are limited reports on the use of wasted black tea after brewing as a low-cost adsorbent and its effects on decreasing the concentrations of metallic substances. Therefore, in this study, after characterization of wasted black tea, its adsorptive performance and efficiency as sorbent for removal of Ni^{2+} , Zn^{2+} , and mixture of Ni^{2+} and Zn^{2+} in a binary system from synthetic wastewater were investigated. The Ni^{2+} and Zn^{2+} concentrations were set to be in range of their actual concentrations in electroplating industry.

2. Materials and Methods

2.1. Preparation of the Adsorbent. Wasted black tea was collected from a local Chinese restaurant located in Kuantan, Malaysia. The samples were black tea (*Camellia sinensis* L.) leaves that have been discarded after the process of making the tea drink in boiling water. The additional coloring, dirt, milk, and sugar were removed by washing the samples with boiled water several times. They were then rinsed with distilled water, oven-dried at 105°C for 24 hours, and passed through a 3.35 mm sieve before the experiment.

2.2. Characteristics Determination. The particle size distribution of the samples was determined using sieve analysis (opening size from 14 to 2 mm). The initial pH was measured by soaking of samples in distilled water (pH 7) for 24 hours and reading the water pH using a pH meter probe (sensION4). The samples were covered with gold sputters using a sputter coater (SC7640) and measured for the surface morphology and elements content. The surface morphology was characterized using Field-Emission Scanning Electron Microscope (FE-SEM, Supra 55 VP, Carl Zeiss, Germany) and elemental analysis was conducted by energy-dispersive X-ray spectroscopy (EDXs, Supra 55 VP, Carl Zeiss, Germany). The functional groups on the surface of adsorbent were determined by Fourier Transform Infrared Spectroscopy (FTIR, Spectrum one, PerkinElmer, US).

2.3. Preparation of Aqueous Solution Containing Heavy Metals. The stock solutions of Ni^{2+} and Zn^{2+} were prepared separately at concentration of 1000 mg/L by dissolving 4.05 g of nickel chloride salt ($\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$) and 4.40 g of zinc sulfate heptahydrate salt ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$) in 1000 mL distilled water. Working solutions were obtained in the range of 1 to 100 ppm by diluting of the standard solutions with distilled water resembling the range of Ni^{2+} and Zn^{2+} concentrations found in effluent released from typical electroplating industry [3]. Medusa-Hydra equilibrium software was used for prediction of different compounds produced in the aqueous solution containing Ni^{2+} and Zn^{2+} . The hydrolysis constants ($\log K$) for each compound were extracted from the software and pH of each component was calculated accordingly.

2.4. Adsorption Behavior. The influences of pH, contact time, and adsorbent dose on adsorption efficiency were studied to determine the optimum values of these factors in sorption mechanism. To conduct these experiments, 250 mL Erlenmeyer flasks containing solutions with known concentrations of Ni^{2+} and Zn^{2+} were kept in a closed environment at ambient temperature (28°C). The pH of the solutions for both Ni^{2+} and Zn^{2+} was varied within the range of 2 to 6. The solutions' pH was adjusted using 0.1N HCl and 0.1N NaOH. This range is justified based on the previous researches which declared that the optimum pH for removal of Ni^{2+} and Zn^{2+} from aqueous solution is in the range of 4 to 5 [23, 24]. The pH was measured by a pH meter probe. The concentration of adsorbent was standardized at 20 g/L and the mixtures were shaken for 60 min. The adsorbent dosage used in the experiment was adjusted to allow for 30–80% of the added adsorbate to be sorbed at equilibrium [27]. To study the effects of contact time on sorption dynamics, the time was set in a range between 0 and 250 min. The concentrations of the heavy metals were measured at 50 min intervals. The agitation speed was set at 150 rotations per min (rpm). An orbital shaker was used to provide adequate contact between adsorbent and adsorbate. The impact of adsorbent dose on sorption was investigated in the range of 2–20 g/L. To measure the concentration of metals ions, the contents of the flasks were filtered through filter paper (Whatman, 0.45 μm) and then analyzed using atomic adsorption spectrophotometer (AAS, AA-6800, Shimadzu,

Japan). The removal percentage of the ions was calculated using

$$\text{Adsorption uptake (\%)} = \frac{(C_o - C_t)}{C_o} \times 100, \quad (1)$$

where C_o is the initial concentration of the ions and C_t is the concentration at time t .

2.5. Binary Metal Adsorption. 50 mL of each metal solution containing 100 ppm of Ni^{2+} and Zn^{2+} was used for this study. 20 g/L of the sample was added and the pH was adjusted in the range of 2 to 6. Samples were shaken and the dose of heavy metals was determined after 60 min.

2.6. Sorption Isotherms. To determine the sorption isotherms of adsorbate on the surface of adsorbent, the batch equilibration procedure was applied. 20 g/L of the adsorbent was added separately to the aqueous solutions containing a range of Ni^{2+} and Zn^{2+} concentrations between 1 and 70 mg/L and then agitated at 150 rpm for 60 min. The pH of all solutions was standardized at pH 5. The experiment was conducted at room temperature (28°C). After equilibration, the concentration of each metal ion in the solutions was measured using AAS in triplicate. Any decrease in mass of the ions in the solutions was assumed to be due to adsorption onto the adsorbent and in this way the sorbed concentrations were calculated. A relationship was then established between the amount of ions adsorbed per unit weight of adsorbent and the ions concentration in the solution at equilibrium. The most frequently used equation for substances sorption is Freundlich and Langmuir adsorption equation which can be described by (2).

Freundlich model is as follows:

$$\frac{x}{m} = K_f C_e^{1/n}, \quad (2)$$

where x is the mass of the sorbed adsorbate (mg), m is the unit mass of adsorbent (g), K_f is the Freundlich capacity factor, C_e is the adsorbate concentration in the solution at equilibrium (mg/L), and $1/n$ is the Freundlich intensity parameter.

Langmuir model is as follows:

$$\frac{x}{m} = \frac{Q_o b C_e}{1 + b C_e}, \quad (3)$$

where Q_o is the maximum adsorption of the substrate to the adsorbent and b is Langmuir constant. The empirical constants in the Freundlich and Langmuir isotherms were determined by plotting graphs using (4) and (5).

Freundlich isotherm is

$$\log\left(\frac{x}{m}\right) \text{ versus } \log C_e. \quad (4)$$

Langmuir isotherm is

$$\frac{C_e}{(x/m)} \text{ versus } C_e. \quad (5)$$

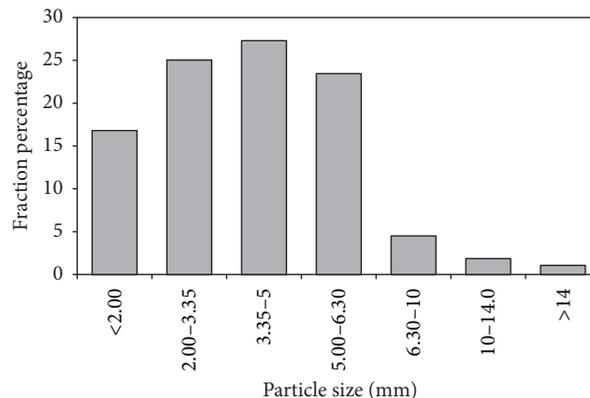


FIGURE 1: Particle size distribution of the wasted black tea.

To compare the capability of Freundlich equation with Langmuir model in prediction of adsorption, average absolute deviation (AAD%) was calculated using

$$\text{AAD\%} = 100 \times \frac{\sum \sqrt{(Y_{\text{Actual}} - Y_{\text{Model}})^2 / Y_{\text{Actual}}^2}}{\text{Number of tests}}. \quad (6)$$

3. Results and Discussion

3.1. Adsorbent Characteristics. Particle size distribution of the wasted black tea sample is expressed in Figure 1. Particle size distribution determines the surface area of a sorbent. Smaller particle size accounts for larger surface area available for adsorption. Based on the outcomes, the size of particles was mostly 3.35 mm and thus the adsorbent with the sizes 3.35 mm and below was used in this study.

Based on the results, the initial pH was 4.76 which is almost the optimum pH for removal of Ni^{2+} and Zn^{2+} [24] and therefore minute preparations were needed to use the wasted black tea for adsorption. Suitability of adsorbent pH is a considerable advantage and it reduces the application cost further as adjustment of pH requires addition of chemicals and usage of extra energy and labor. Figure 2 indicates the concentration of determined elements in the sample using EDX spectroscopy. Based on the results, wasted black tea is rich in elements such as carbon and calcium similar to the activated carbon. The high concentrations of these elements allow the adsorbent to exhibit high sorption capacity [28].

The surface morphology of adsorbent obtained by SEM and the results of FTIR spectra are presented in Figure 3. Based on the photograph, there are a large volume of voids on the surface which result in great surface area, abundant adsorption sites, and therefore a high sorption capacity. This allows the adsorbate to efficiently adhere to the surface of the adsorbent. The FTIR peaks represent various chemical bonds in the sample which can be effective in sorption activity. The broad peak at 3421 cm^{-1} is assigned to $-\text{OH}$ polar groups. The bands at $2920-2851$ and $1451-1319 \text{ cm}^{-1}$ are assigned to aliphatic $-\text{CH}_2$ units. The sorption peaks at $1637-1542 \text{ cm}^{-1}$ are assigned to aromatic $\text{C}=\text{O}$ and $\text{C}=\text{C}$ vibration indicating presence of hard-carbon components. This confirms the EDX

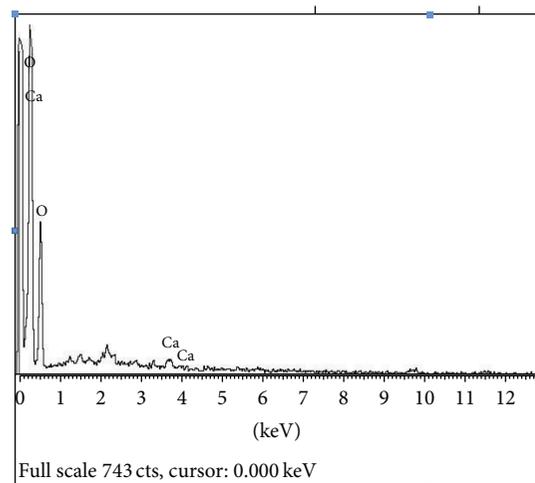


FIGURE 2: EDS curve of tea waste.

results specifying the high carbon content measured in the sample. An adsorption peak at 1240 cm^{-1} is attributed to aromatic CO- and phenolic -OH bond. The peaks around $1157\text{--}1034\text{ cm}^{-1}$ correspond to aliphatic C-O-C and alcohol -OH representing oxygenated functional groups of cellulosic and ligneous components. The oxygen-containing functional groups have been reported that play important roles in the capacity of organic sorbents in adsorption of heavy metals [29]. Therefore, it can be concluded that the physical sorption of Ni^{2+} and Zn^{2+} by tea waste was related to the coulombic forces which are electrostatic energy of interaction between the metal ions and the adsorbent's oxygenated functional groups [22].

3.2. Effect of Solution pH on Adsorption. Figure 4 shows the removal percentage of Ni^{2+} and Zn^{2+} in pH ranging from 2 to 6. The amount of metal ions sorption enhanced as the pH increased. The sorption of Ni^{2+} showed a gentle increasing trend and reached its maximum (about 60%) at pH 6. This result was similar to that of Soares et al. [30] who evaluated the effects of soil pH (ranged from 3 to 5) on Ni^{2+} adsorption in Brazilian soils. Based on their results, the metal sorption increased (20–90%) when the pH was raised from 4 to 6. As it is presented in Figure 4, the highest percentage (about 80%) of Zn^{2+} sorption occurred at pH 5 and stayed unchanged while the pH reached 6. Several studies have reported that the pH ranging from 4 to 5 is the optimum values for removal of Zn^{2+} from aqueous solution. According to Malkoc and Nuhoglu [24], the maximum sorption of Ni^{2+} was recorded at pH 4.0. Wasewar et al. [25] investigated a wide range of pH from 2 to 12 and reported that the best Zn^{2+} removal happened at pH 4.2. It has been shown that, at lower pH value, the H^+ ions compete with metal cations for the electrostatic surface charges in the system decreasing the percentage of sorption. It is well known that the sorption of Ni^{2+} and Zn^{2+} is affected by the species of the ions in aqueous solutions which are strongly controlled by pH values. The reduction in adsorption at higher pH can be attributed

TABLE 2: Hydrolysis constants ($\log K$) and pH of Ni^{2+} and Zn^{2+} species.

Zn^{2+} species	$\log K$	pH	Ni^{2+} species	$\log K$	pH
$\text{Zn}(\text{OH})_2$	-16.4	8.3	$\text{Ni}(\text{OH})_2$	-20.01	7.1
$\text{Zn}(\text{OH})_4^{-2}$	-41.3	5.5	$\text{Ni}(\text{OH})_4^{-2}$	-45.0	4.5
$\text{Zn}_2(\text{OH})_6^{-2}$	-54.3	6.5	$\text{Ni}_2\text{OH}^{+3}$	-9.8	10.5
$\text{Zn}_2\text{OH}^{+3}$	-9.0	10.8	$\text{Ni}_4(\text{OH})_4^{+4}$	-27.9	9.9
$\text{Zn}_4(\text{OH})_4^{+4}$	-27.0	10.3	NiOH^+	-9.5	5.3
ZnOH^+	-7.5	10.2			

to the precipitation of metal hydroxides of these ions [24]. Table 2 shows the overall hydrolysis constants ($\log K$) of the Ni^{2+} and Zn^{2+} hydroxyl species that can be used to calculate the relative distribution of the ions species in different pH values. Overall, the pH of both Ni^{2+} and Zn^{2+} compounds in solution remained below the pH that sedimentation of nickel and zinc compounds exhibited.

3.3. Effect of Contact Time on Adsorption. The variation in Ni^{2+} and Zn^{2+} sorption with varying contact time is illustrated in Figure 5. Based on the results, the amount of sorption showed an increasing tendency as the contact time increased, but it was more significant for zinc. Removal percentage of Zn^{2+} was just below 60% in contact time of 50 min and then reached up to 86.7% in time of 250 min, while for Ni^{2+} the changes in sorption percentage were just 5% during the range of agitation times. These results are similar to those obtained by Malkoc and Nuhoglu [24] that showed that the increase in agitation rate improves efficiency of metal ions removal. Longer period of contact between the adsorbent and adsorbate increases the amount of sorption; however the adsorption rate reduces as the contact time increases [25]. This is due to the decrease of adsorption sites on the adsorbent while it is being saturated by the heavy metals. Based on the results, the optimum contact time for sorption was around 50–60 min.

3.4. Effect of Adsorbent Dose on Adsorption. Figure 6 illustrates the percentage of Ni^{2+} and Zn^{2+} sorption with increasing the adsorbent dose. Removal of the metal ions showed similar increasing pattern as the adsorbent concentrations rose from 2 to 20 g/L. Sorption percentage of nickel and zinc increased by 41% and 47%, respectively. The increase of adsorbent concentration means the increase of surface area and more available sorption sites which increases the amount of metal ions removal [24].

3.5. Binary Metal Adsorption. The sorption percentage of Ni^{2+} and Zn^{2+} in the binary system within the pH range of 2 to 6 is presented in Figure 7. The results of the simultaneous adsorption showed that the sorption capacity for both heavy metals in binary metal sorption was still very similar to single metal ions sorption. So, the binary adsorption exhibits the noninteraction behavior where the mixture of metal ions has no effect on the adsorption of the ions in the mixture. This can be attributed to similar charge intensity and ionic radius

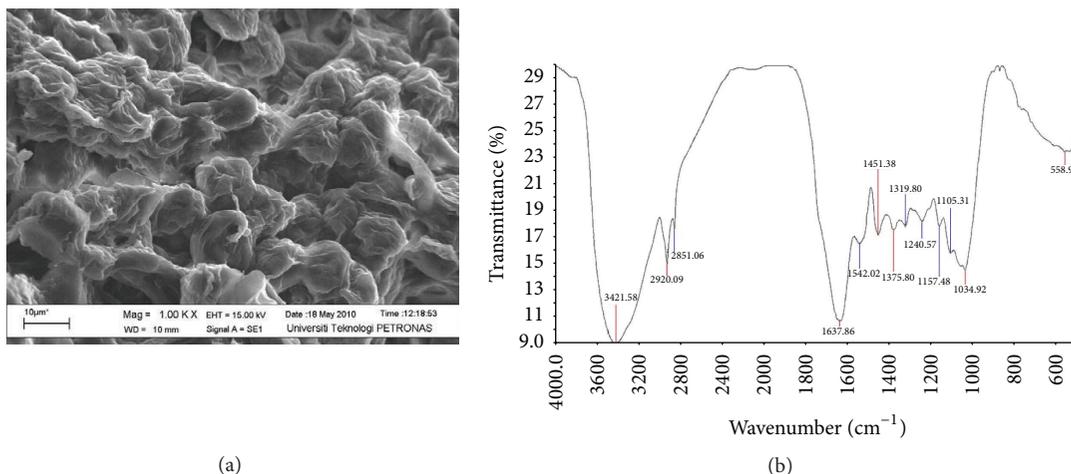


FIGURE 3: SEM photograph of wasted black tea at 1000x magnification (a) and FTIR spectra of the wasted black tea (b).

TABLE 3: The parameters of Freundlich and Langmuir adsorption equations.

Adsorbate	Freundlich constants			AAD%	Langmuir constants			AAD%
	K_f (mg/g)	$1/n$	R^2		Q_o (mg/g)	b	R^2	
Ni ²⁺	20.80	0.24	0.976	3.49	90.91	0.224	0.949	3.37
Zn ²⁺	16.22	0.36	0.994	2.92	166.67	0.133	0.917	3.53

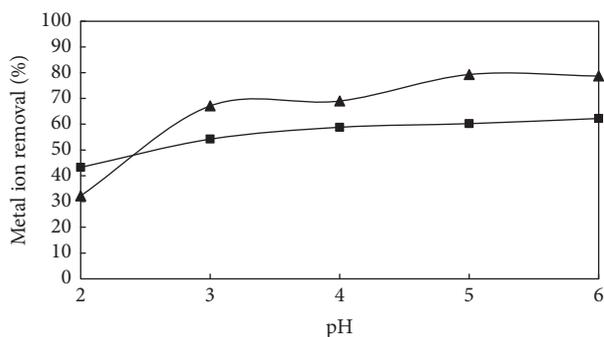


FIGURE 4: Percentage of Ni²⁺ (■) and Zn²⁺ (▲) ions removal with varying pH.

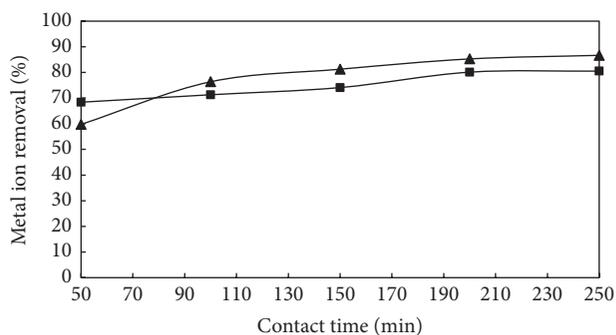


FIGURE 5: Percentage of Ni²⁺ (■) and Zn²⁺ (▲) ions removal with varying contact time.

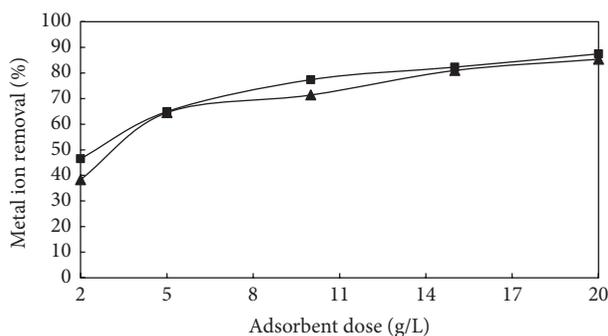
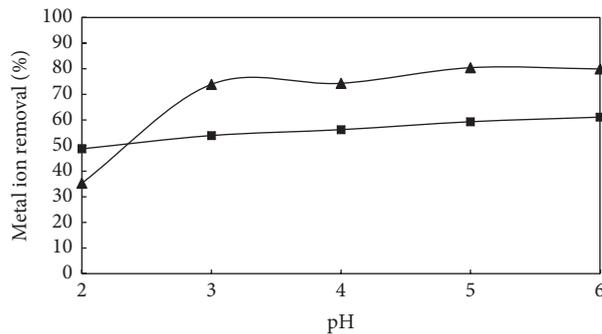
of the metal ions. The ionic radii values of Ni²⁺ and Zn²⁺ are 0.69 and 0.68 Å, respectively [43].

3.6. Adsorption Isotherms. The outcomes of evaluation of sorption isotherms of Ni²⁺ and Zn²⁺ on the wasted black tea are presented in Figure 8. Both Freundlich and Langmuir adsorption equations were investigated. The data fitted well with both models. However, correlation coefficients (R^2) showed higher values in Freundlich equation compared to Langmuir model. The AAD% for Freundlich and Langmuir equations indicated that both models were almost similar and precise in prediction of Ni²⁺ and Zn²⁺ sorption. The related parameters are listed in Table 3. Magnitude of the sorption coefficients (Q_o and K_f , y -intercepts) in both equations signifies the high sorption affinity of wasted black tea for nickel and zinc.

Table 4 presents comparison of the adsorption capacity of wasted black tea used in this study with other studies aimed at removal of Ni²⁺ or Zn²⁺. The sorption capacity (Q_o value) of wasted black tea showed a magnitude of 90.91 mg-Ni/g adsorbent, while that for powdered activated carbon was reported to be 31.08 mg/g [34]. Although the adsorbent dose in the present study was 1.7 times more, the sorption performance of tea waste was still higher than the potential of activated carbon. Q_o value for wasted black tea was 166.67 mg-Zn/g adsorbent. In spite of higher applied adsorbent dose (2%), the sorption capacity of wasted black tea was comparable with the capacity of bagasse-based activated carbon in sorption of Zn²⁺ ion (54 mg/g) [37]. The high sorption potential of wasted black tea was achieved without any activation or additional treatment of the biomass. Therefore, wasted black tea can be still considered as a cost-effective and sustainable

TABLE 4: The capacity of different adsorbents for Ni²⁺ and Zn²⁺ removal.

Adsorbent	Adsorbent mass to solution volume ratio	Sorption capacity (mg/g)	Reference
Ni ²⁺ removal			
Sphagnum moss peat	0.4 : 100	9.18	[31]
Baker's yeast	0.1 : 100	11.40	[32]
<i>Chlorella sorokiniana</i> (FBCS)	0.1 : 100	48.08	[33]
Powdered activated carbon	1.2 : 100	31.08	[34]
Waste tea	1.5 : 100	18.42	[24]
Waste tea (formaldehyde-treated)	0.2 : 100	120.50	[35]
Black wasted tea	2 : 100	90.91	This study
Zn ²⁺ removal			
Olive oil mill residues	0.4 : 100	52.91	[36]
Bagasse-based activated carbon	0.6 : 100	54.00	[37]
Date pits-based activated carbon	0.02 : 100	120.5	[38]
Chitosan	0.25 : 100	58.83	[39]
Waste tea leaves	0.15 : 100	11.76	[40]
Black tea waste	1 : 100	12.24	[41]
Tea factory waste	0.4 : 100	8.9	[25]
Tea leaves (hydrazine monohydrate-exhausted)	0.125 : 100	79.76	[42]
Wasted black tea	2 : 100	166.67	This study

FIGURE 6: Percentage of Ni²⁺ (■) and Zn²⁺ (▲) ions removal with varying adsorbent dose.FIGURE 7: Percentage of Ni²⁺ (■) and Zn²⁺ (▲) ions removal with varying pH in binary metal sorption.

adsorbent compared to other adsorbents for remediation of aquatic systems.

The same results were obtained by Ahluwalia and Goyal [44] who showed the high sorption capacity of tea leaves for heavy metals with Q_0 values of 515.03 mg/g and 785.5 mg/g for Ni²⁺ and Zn²⁺, respectively (adsorbent dose of 0.5%).

4. Conclusion

The findings in this study indicate that wasted black tea is effective adsorbent for the removal of nickel and zinc from wastewater due to its unique properties including the high content of carbon, high porosity, and reactive functional

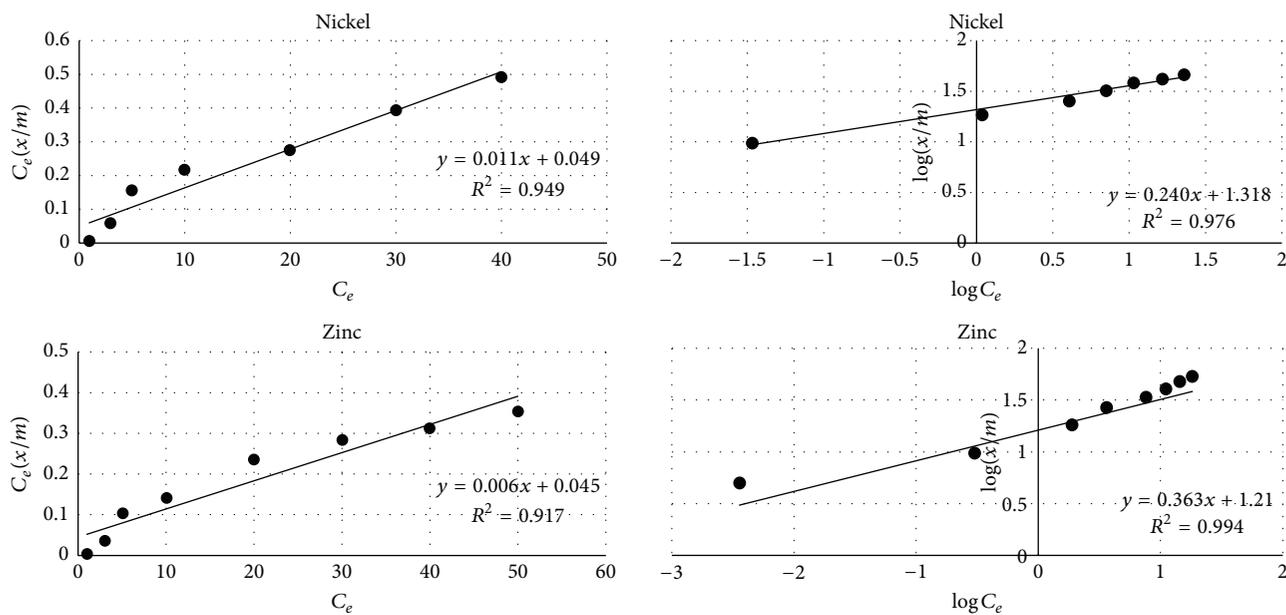


FIGURE 8: Langmuir and Freundlich isotherms for the adsorption of Ni^{2+} and Zn^{2+} .

sites. Wasted black tea as a salvaged material has a very low economic value and since it was found that its original pH is proper for sorption of nickel and zinc, its utilization in industries having nickel and zinc in their discharges seems to be possible. Wasted black tea had shown good performance in removal of nickel and zinc from aqueous solution in both monocomponent metal and binary systems. Maximum adsorptions of 90.91 and 166.67 mg/g of nickel and zinc, respectively, indicate that wasted black tea is a competitive adsorbent for industrial applications. However, a feasibility study is recommended to take place when the wasted black tea is in plan to be used for industrial discharge treatment. Some factors including collection, transportation, and preparation costs of wasted black tea should be considered in the feasibility study before its applications at large scales.

Competing Interests

The authors declare that they have no competing interests.

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