

Enhancement of the Adsorptive Properties of a Desert *Salsola Vermiculata* Species

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ABSTRACT: Heat pretreatment, steam activation and 20% phosphoric acid activation were found to enhance the adsorption capacity of a desert *Salsola Vermiculata* plant species significantly relative to the inactivated plant and powdered wood carbon 600200 EX 694 as obtained from Belgium. The batch adsorption experiments resulted in a maximum adsorption capacity determined from Langmuir models of up to 476 mg/g for acetic acid, 238 mg/g for copper(II) ions and 144 mg/g for nickel(II) ions, whereas for the inactivated plant they attained values of 53 mg/g, 65 mg/g and 18 mg/g, respectively. The results for the wood carbon were 65 mg/g for acetic acid, 45 mg/g for copper(II) ions and 2.4 mg/g for nickel(II) ions. The Freundlich model was found to provide a better description of copper(II) ion adsorption onto the steam- and chemically-activated plant, whereas nickel(II) ion adsorption processes followed Langmuir behaviour. These results showed that the activated plant could prove to be very useful in removing toxic substances from wastewater.

INTRODUCTION

Water contamination by toxic materials such as heavy metals and organic solvents has become a major source of pollution of the environment. Conventional methods for water purification are costly and sometimes not readily adapted to effluent treatment. During the last decade, various cheap natural sorbents have been the subject of intense interest for the removal of contaminants from water. Many plants and fibres, such as water hyacinth, tea leaves, tree barks, ground coffee, wool fibre, mucor rouxi, etc., have been studied for this purpose (Yan and Viraraghavan 2000; Marshall and Champagne 1995; Vasquez *et al.* 1994; De Vasconcelos Teles *et al.* 1992; Ruff *et al.* 1992; Tee and Khan 1988; Randall *et al.* 1975; Balkose and Baltacioglu 1992). The need to use other adsorbents as alternatives to the standard activated carbons is due to the rising production cost of the latter (Quek *et al.* 1998) and also to take advantage of available low-cost natural products.

Copper is used extensively in electrical industries, in fungicides and in anti-fouling paints. It can become toxic to humans when ingested at high concentrations, promoting oxidation and causing gastrointestinal irritation and cancer (Schmuhl *et al.* 2001). Its maximum contaminant level is 1.3 mg/l. Nickel is a metal commonly used in a series of industrial processes such as electroplating, plastics, pigments, nickel/cadmium batteries, etc. It might cause cancer of the upper respiratory tract when associated with other elements, the allowable concentration limit

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being lower than 0.1×10^{-3} mg/l. Acetic acid is a well-known organic substance used extensively in the study of activated carbons.

The use of adsorbents such as activated carbons for the adsorption of acetic acid has been the subject of numerous studies. Adsorption capacities obtained attained values of 112 mg/g for AKOM carbons with specific surface areas of 117 m²/g (Avom *et al.* 2001) and up to 480 mg/g for ZM-1 and ZM-2 Vinylon activated carbons (China) with specific surface areas of 1300 m²/g. Copper(II) ion adsorption capacities onto adsorbents such as chitosan attained values of 288 mg/g (McKay *et al.* 1989). Nickel(II) ion adsorption has also been studied on various adsorbents but the adsorption capacities obtained for this element were generally low with the highest values being for activated cherry and apricot stones with values of 120 mg/g and 90 mg/g, respectively. The iodine number determinations for these activated carbons were 895 mg/g and 905 mg/g, respectively (Petrov *et al.* 2003).

In the present study, attention was focused on a natural desert plant of the *Salsola Vermiculata* species found in southwest Algeria (Béchar province). It is commonly used locally as a cleansing substance. Preliminary studies performed on the inactivated plant revealed interesting adsorptive properties for acetic acid and heavy metal ions such as copper(II) and nickel(II).

EXPERIMENTAL METHODS

Activation methods

The adsorption properties of the inactivated plant leaves towards known elements such as copper, nickel and others were first investigated in a preliminary study. Subsequently, the leaves were subjected to various methods of activation in order to enhance their adsorption potential. In the first method studied, mild atmospheric heat treatment under limited oxygen access at 500°C was applied to 10 g of the plant for 3 h in order to obtain the precursor. In the second method, the plant was exposed simultaneously to steam for the same time length under the same conditions. Finally, the divided dried plant leaves were treated chemically with phosphoric acid (20%) at 170°C for 90 min, then washed 10 times, dried overnight at 95°C and heated at 500°C for 2.5 h. Adsorption measurements were undertaken for each of the activated substances obtained.

Material and measurement methods

The *Salsola* plant leaves were washed and dried overnight in an oven at 110°C and finely divided using a Crosschop Viersen grinder rotating at 90 rpm for 4 h to obtain a particle diameter less than 0.071 mm. All reagents used were of analytical grade (Merck, Germany). Glacial acetic acid was further diluted with distilled water produced in the laboratory. Solutions of cupric sulphate, nickel(II) sulphate, sodium hydroxide, sodium thiosulphate, potassium iodide and iodine of known concentrations were prepared according to standard procedures.

Sodium thiosulphate titrations, using a 1% starch end-point indicator (Alexeev 1980), were performed for the estimation of copper(II) ion solutions. Acetic acid determinations were effected using sodium hydroxide with phenolphthalein as the end-point indicator. Nickel(II) ion solutions were prepared using a 0.5 M HCl medium, 2.5% dimethylglyoxime and 10 M NaOH and persulphate solutions. Visible spectrophotometric measurements were performed on the nickel(II) ion solutions at 480 nm using a UV-vis 120-01 Shimadzu spectrophotometer. Solutions were diluted as required so that their absorbance remained within the linear calibration range. Iodine number

determinations were undertaken using the standard 0.02 mol/l iodine equilibrium solution method. All measurements were undertaken in duplicate.

The adsorption of acetic acid, copper(II) and nickel(II) ions was studied using a batchwise process employing 250-ml capped Erlenmeyer flasks at a fixed temperature of 24°C. The starting solutions of the relevant substances were titrated beforehand with the corresponding analytical reagents. For the sorption studies, a 25-ml volume of the relevant sample solution was removed via a micropipette and mixed with 0.1 g of the powdered plant or wood carbon. The resulting suspension was then agitated magnetically for 90 min in the Erlenmeyer flask at a constant speed of 250 rpm. After the adsorption process had occurred, the solutions were centrifuged at 4000 rpm and the supernatants analysed.

RESULTS AND DISCUSSION

Adsorption isotherms

An important physicochemical quantity for evaluating an adsorption process is the equilibrium adsorption which gives the capacity of the adsorbent. The equilibrium relationships between the adsorbent and adsorbate are described by adsorption isotherms determined at a fixed temperature. Two commonly used types of adsorption isotherms are considered here: Langmuir adsorption isotherms and Freundlich adsorption isotherms (Cooney 1999).

The Langmuir relationship describing the adsorption equation may be expressed in its linearised form as:

$$\frac{C_{eq}}{x/m} = \frac{1}{Kb} + \frac{1}{b} C_{eq} \quad (1)$$

where x/m (mg/g) is the adsorption density, C_{eq} (mg/l) is the concentration of adsorbate in solution at equilibrium, b (mg/g) is the maximum adsorption capacity corresponding to complete monolayer coverage and K (l/mg) is the Langmuir constant. The linear plot of $C_{eq}/(x/m)$ versus C_{eq} yields the Langmuir constants b and K . The determination of b allows the active specific surface area of the adsorbent to be calculated, provided that a value is assumed for the surface area covered per molecule. The constant b does not represent a true monolayer capacity but rather the practical limiting capacity for adsorption.

The Freundlich isotherm is an empirical isotherm used to describe heterogeneous surface energies. It assumes that the adsorption heat is an exponential function that decreases with surface coverage and is governed by the following equation:

$$x/m = K_f C_{eq}^{1/n} \quad (2)$$

where K_f and n are constants. The values of K_f and n ($n > 1$) can be determined from the intercept and slope of the plot of $\log(x/m)$ versus $\log C_{eq}$. The intercept is a rough indicator of the sorption capacity and the slope ($1/n$) of the adsorption intensity.

Determination of the adsorption equilibrium times

To determine the equilibrium adsorption times, 25 ml of solutions containing 115 mg/l and 500 mg/l of the three adsorbates investigated were mixed successively with 0.1 g of the inactivated plant, the precursor, the steam-activated and chemically-activated plant, and wood carbon at 24°C.

TABLE 1. Equilibrium Times for the Adsorption of Acetic Acid, Copper(II) and Nickel(II) Ions^a

System	Initial concentration = 115 mg/l			Initial concentration = 550 mg/l		
	Acetic acid	Copper ions	Nickel ions	Acetic acid	Copper ions	Nickel ions
Inactivated plant	40	45	58	50	55	45
Precursor	35	40	45	46	49	51
Steam-activated	38	42	50	42	50	55
Chemically-activated	32	45	52	39	51	53
Wood carbon	29	30	31	30	29	32

^aEquilibrium times quoted in min.

The resulting suspensions were analysed at 10-min time intervals. Table 1 shows the equilibrium concentration times (in min) for acetic acid, copper(II) ions and nickel(II) ions, respectively. Rapid equilibrium was attained for all three adsorbates with the treated plant and carbon, with the maximum equilibrium time observed being less than 60 min. As a consequence, subsequent adsorption studies were all performed for 90 min which was assumed to be ample for all the experiments.

Acetic acid adsorption

Acetic acid was studied on the basis that it represented a standard adsorbate used in activated carbon studies. The surface area of an adsorbed acetic acid molecule was taken as ca. $21 \times 10^{-20} \text{ m}^2$ in subsequent calculations of the specific surface area of the adsorbent under consideration (Shoemaker *et al.* 1981).

Figure 1 depicts the results obtained for the adsorption of acetic acid on the inactivated, the precursor, the heat-activated and chemically-activated Saharan plant as compared to 600200 EX 694

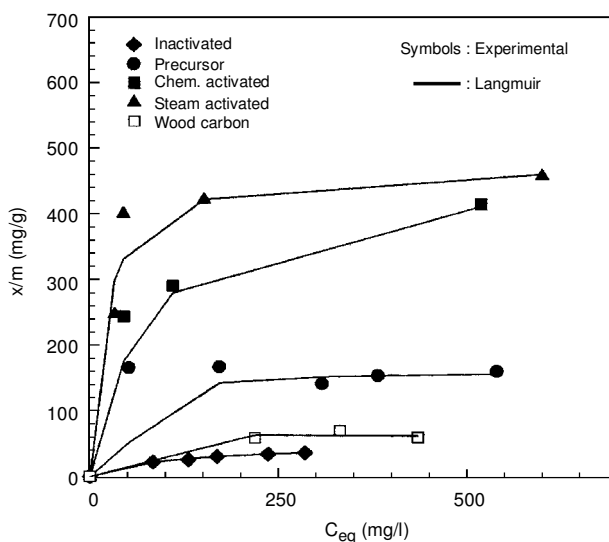


Figure 1. Adsorption of acetic acid onto *Salsola Vermiculata* plant and wood carbon.

wood carbon. The pH values during the adsorption process ranged from 3.94 to 3.41. It can be seen that the highest values for the adsorbed concentration were obtained for the steam- and chemically-activated adsorbents, while the lowest values were those for the natural inactivated plant. The wood carbon values ranked just above the latter. Hence, a significant enhancement was attained with both activation processes.

This is more clearly seen in Figure 2, which shows that the Langmuir isotherm fitted the data obtained for all five systems studied. Table 2 lists the equations of the fitted straight lines and the corresponding Langmuir constants. The high correlation coefficients for the five linear fitted curves ranged from 0.93 (chemical activation) to 0.99 (steam activation), thereby indicating that these systems were all well described by the Langmuir isotherms. The slope of the line is related to the adsorption capacity such that a low value for the slope corresponds to a high capacity. Lower slopes (ca. 0.0021) were observed for adsorbents obtained via steam and chemical activation. The slope obtained for the precursor system was 0.006, which was far lower than that for the natural inactivated system (0.0187). The powdered wood carbon ranked just above the inactivated plant with a slope of 0.0155.

The maximum adsorption capacity b values for acetic acid adsorption could be deduced from the inverse of the slopes ($1/b$). It should be noted from Table 2 that both the steam and chemical activation processes improved the sorptive properties of the *Salsola* plant significantly, from 53 mg/g for the inactivated plant up to ca. 476 mg/g for the activated plant. The maximum adsorption capacity attained for the wood carbon was 65 mg/g. Table 2 also shows the values of the iodine number (mg/g) and the estimated specific surface areas (m^2/g) calculated according to the treatment method. The specific surface areas attained were $113 m^2/g$ for the natural inactivated plant, $375 m^2/g$ for the precursor plant and ca. $1000 m^2/g$ for the steam- and chemically-activated plant. In comparison, the specific surface area of the powdered wood carbon was ca. $136 m^2/g$. The values of b obtained demonstrate the relatively greater practical adsorption capacity of the activated plant with respect to the wood carbon. The iodine number values showed the same pattern which is not unexpected since this number can be roughly related to the specific surface area (Noszko *et al.* 1984).

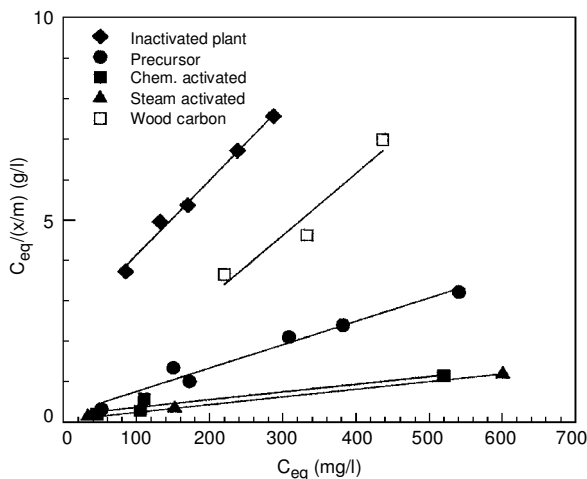


Figure 2. Langmuir isotherms for the adsorption of acetic acid onto *Salsola Vermiculata* plant and wood carbon.

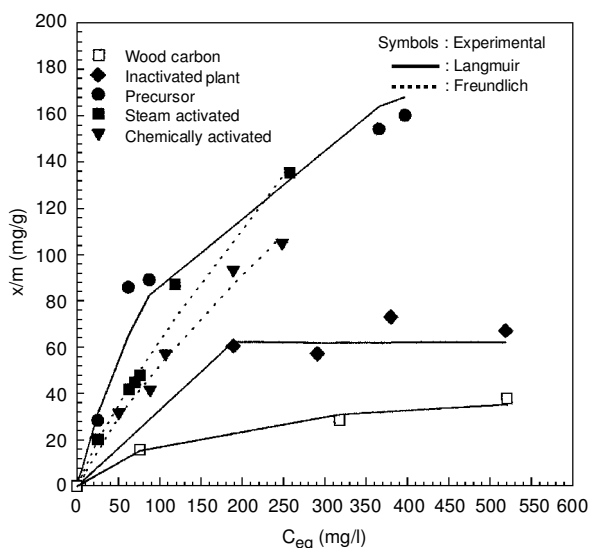
TABLE 2. Langmuir Equations and Constants for Acetic Acid Adsorption, Specific Surface Area and Iodine Number Values for Salsola Plant and Wood Carbon

System	Langmuir equation	K (l/mg)	b (mg/g)	Specific surface area (m ² /g)	Iodine number (mg/g)
Inactivated plant	$C_{eq}/(x/m) = 2.300 + 0.0187C_{eq}$	0.008	53.48	133	209
Precursor	$C_{eq}/(x/m) = 0.169 + 0.006C_{eq}$	0.036	166.66	375	496
Steam-activated	$C_{eq}/(x/m) = 0.041 + 0.0021C_{eq}$	0.051	476.19	1000	1100
Chemically-activated	$C_{eq}/(x/m) = 0.165 + 0.0021C_{eq}$	0.013	476.19	1000	1050
Wood carbon	$C_{eq}/(x/m) = 0.0268 + 0.0155C_{eq}$	0.578	64.52	136	272

Copper(II) ion adsorption

Figure 3 shows the adsorption isotherms obtained for copper(II) ions. No attempt was made to maintain the pH value constant in all the equilibrium adsorption experiments conducted, and for this reason the pH values ranged from 5.1 to 6.4. This range corresponds to a maximum copper(II) and nickel(II) ion uptake with the various adsorbents studied (Quek *et al.* 1998; Gardea-Torresdey *et al.* 1998).

The plots depicted show that the Salsola plant exhibited good adsorption properties towards copper(II) ions even when inactivated. However, the various activation methods applied to the plant resulted in a somewhat different behaviour relative to acetic acid. Although the chemical and the steam activation processes improved the adsorption capacity towards copper(II) ions, the precursor seemed to exhibit better adsorption properties towards this ion. The wood carbon exhibited the lowest adsorption potential towards copper(II) ions.

**Figure 3.** Adsorption of copper(II) ions onto Salsola Vermiculata plant and wood carbon.

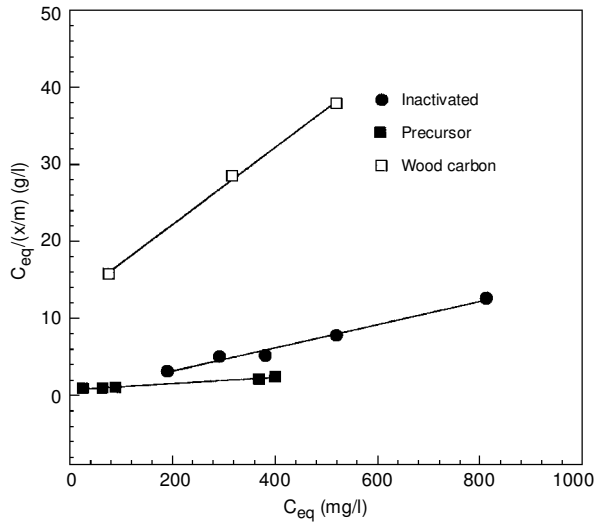


Figure 4. Langmuir isotherms for copper(II) ion adsorption onto inactivated plant, precursor and wood carbon.

Figure 4 depicts the Langmuir-fitted curves for the inactivated plant, precursor and wood carbon. The linear regression correlation coefficients $R^2 > 0.98$ indicated a good linear fit for the three curves. However, the data obtained for the chemical and steam activated adsorbents did not follow the Langmuir equation, suggesting that more heterogeneous systems were involved. Plots of the data for the steam- and chemically-activated substances fitted to the Freundlich equation are shown in Figure 5, with correlation coefficients for both types of adsorbent being 0.97.

Table 3 lists the Langmuir equations and corresponding b values. It will be noted that the value of b for the inactivated plant was 65 mg/g whereas a value of b equal to 238 mg/g was obtained

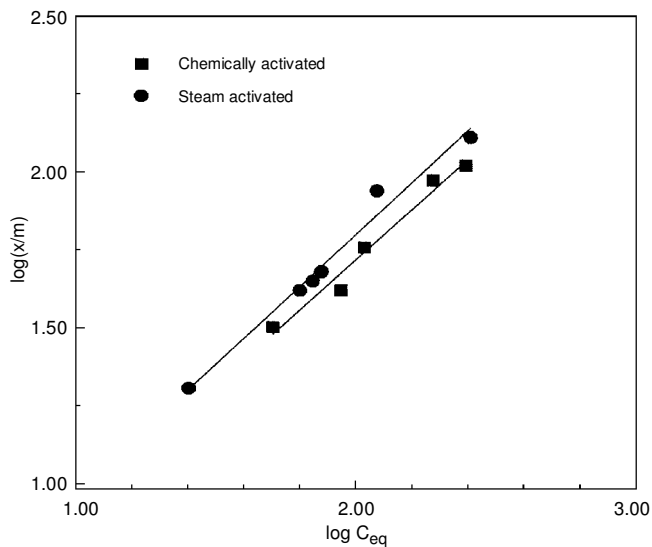


Figure 5. Freundlich isotherms for copper(II) adsorption onto steam- and chemically-activated *Salsola* plant.

TABLE 3. Langmuir and Freundlich Equations and Constants for Copper(II) Adsorption onto Salsola Plant and Wood Carbon

System	Langmuir	Freundlich		R ²			
	Equation	K	b		Equation	K _f	1/n
Inactivated plant	$C_{eq} = 0.08 + 0.015C_{eq}$	0.191	65.36				0.99
Precursor	$C_{eq} = 0.70 + 0.004C_{eq}$	0.006	238.1				0.98
Steam-activated	$C_{eq} = 12.1 + 0.050C_{eq}$	0.004	20.0				0.99
Chemically-activated				$\log(x/m) = 0.137 + 0.829 \log C_{eq}$	1.37	0.83	0.97
Wood carbon				$\log(x/m) = 0.109 + 0.803 \log C_{eq}$	1.29	0.80	0.97

for the precursor, i.e. an improvement in the adsorption capacity by a factor of 3.66. Similar values (289 mg/g) were obtained for copper(II) ion adsorption onto chitosan (McKay *et al.* 1989). In comparison, only 20 mg copper(II) ions were adsorbed per gram of wood carbon. The equations of the Freundlich-fitted linear plots are also reported in Table 3.

The steam-activated adsorbent gave values of 0.83 and 1.37 for the $1/n$ and K_f constants whereas these quantities were equal to 0.80 and 1.29, respectively, for the chemically-activated system. Hence, within experimental error, the values of the adsorption intensity ($1/n$) for these two systems were close. This suggests that the forces between the adsorbed copper(II) ions and the treated plant surface were of equal strength. This kind of behaviour has also been observed with other adsorbents for which corresponding $1/n$ values of 0.18 (Marshall and Champagne 1996) and 0.98 (McKay *et al.* 1989) were obtained.

The K_f value for the steam-activated plant was slightly higher than that for the chemically-activated plant, suggesting a slightly better affinity between the adsorbed copper(II) ions and the chemically-activated surface. This value is in agreement with the literature value (1.53) for the adsorption of copper(II) ions in slow sand filters, where the adsorption capacity was found to be 719 mg copper(II) ions per gram of sand (Muhammad *et al.* 1998).

Nickel(II) ion adsorption

Figure 6 shows the adsorption isotherms for nickel(II) ion adsorption for the four plant systems and powdered wood carbon. The highest adsorption values were obtained for the steam-activated system followed by the chemically-activated one, whereas those for the natural and precursor systems were close. As shown in Figure 7, all the adsorption isotherms followed Langmuir behaviour thereby suggesting a limiting adsorption capacity for nickel(II) ions. Curve-fitting yielded straight lines with correlation coefficients of 0.99 for the precursor, steam and wood carbon systems. For the inactivated and chemically-activated plant systems, the correlation coefficients were 0.96 and

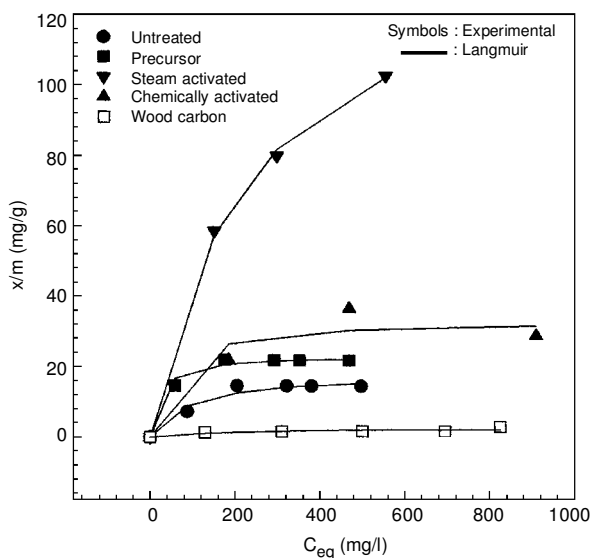


Figure 6. Adsorption isotherms for nickel(II) adsorption onto *Salsola* plant and wood carbon.

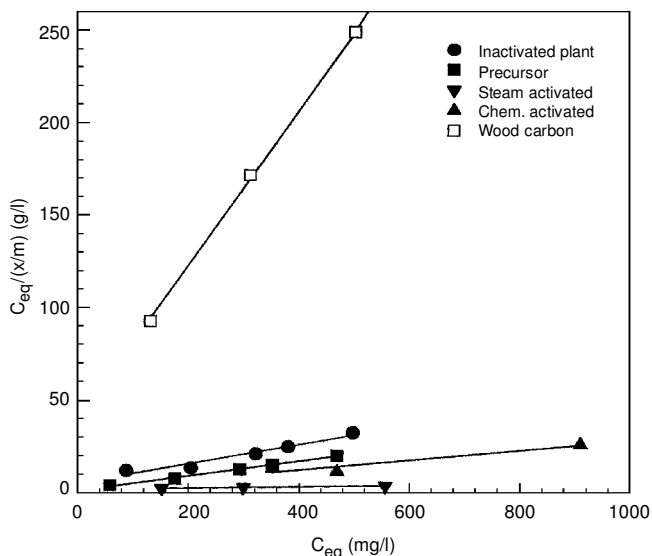


Figure 7. Langmuir isotherms for nickel(II) adsorption onto Salsola plant and wood carbon.

0.93, respectively. The lowest slope value observed for the adsorbent obtained via steam activation suggests that this method of preparation would be most likely to give the best activation results for nickel(II) ion adsorption.

Table 4 reports the Langmuir equations and the values of the constants K (l/mg) and b (mg/g) as obtained from the intercepts and slopes of the plots depicted in Figure 7. The close K values, ranging from 0.011 for the inactivated plant and powdered wood carbon to ca. 0.04 for the precursor and chemically-activated systems to finally 0.004 for the steam-activated system, suggest that the energy factors for nickel(II) ion adsorption onto these systems were similar. The b value of 18 mg/g for the inactivated biomass plant indicates that this material already exhibited a good potential for the adsorption of nickel(II) ions compared, for example, to inactivated creosote bush leaves where a literature value of 5.90 mg/g has been reported (Muhammed *et al.* 1998). The precursor showed an improvement of ca. 16% (from 18 mg/g to 21 mg/g). However, the materials obtained after chemical and steam treatment that exhibited adsorption capacities of 31 and 144 mg/g towards nickel(II) ions, i.e. a significant enhancement, demonstrated the tremendous application possibilities for this plant. In comparison, activated cherry stones attained

TABLE 4. Langmuir Constants for Nickel(II) Ion Adsorption onto Salsola Plant and Wood Carbon

System	Langmuir equation	K (l/mg)	b (mg/g)	R^2
Inactivated plant	$C_{eq}/(x/m) = 5.10 + 0.054C_{eq}$	0.011	18.52	0.96
Precursor	$C_{eq}/(x/m) = 0.99 + 0.043C_{eq}$	0.043	23.26	0.99
Steam-activated	$C_{eq}/(x/m) = 1.58 + 0.0069C_{eq}$	0.004	144.93	0.99
Chemically-activated	$C_{eq}/(x/m) = 0.7 + 0.029C_{eq}$	0.041	34.48	0.93
Wood carbon	$C_{eq}/(x/m) = 39.73 + 0.42C_{eq}$	0.011	2.38	0.99

an adsorption capacity of 120 mg/g towards nickel(II) ions (Petrov *et al.* 2003). The wood carbon investigated in this study exhibited a poor adsorption capacity towards nickel(II) ions with only 2.38 mg/g carbon being adsorbed.

CONCLUSIONS

This study showed that the low-cost natural *Salsola Vermiculata* was capable of adsorbing acetic acid, nickel(II) and copper(II) ions from synthetic aqueous solutions. Its adsorption potential towards these substances was greatly enhanced by activation. From the data obtained from the acetic acid system which were well described by the Langmuir isotherm, the steam and chemical activation processes gave adsorbents with the highest uptake value (476 mg/g) at pH values within the range 3.41–3.94. The specific surface area available for acetic acid increased from 113 m²/g for the natural plant to ca. 1000 m²/g for the steam- and chemically-activated plant. Encouraging results were also obtained from iodine number determinations.

Relative to the adsorption capacity of the inactivated plant (65 mg/g), the precursor exhibited the best adsorption capacity towards the copper(II) ion system (238 mg/g) over the pH range 5.1–6.4. Freundlich isotherms best described copper(II) ion adsorption on the steam- and chemically-activated materials, with 1/n and K_f values of ca. 0.80 and 1.3, respectively. Although many adsorbents show only small adsorption capacities towards the nickel(II) ion, this study showed that the nickel(II) ion adsorption capacity (which was well described by the Langmuir equation) increased from 18 mg/g to 144 mg/g as a result of steam activation. In comparison with the wood carbon investigated, the *Salsola Vermiculata* plant activated in this way exhibited a much higher adsorption capacity overall. For this reason, this material may be very useful for the treatment of wastewater and industrial effluents which nowadays constitute a major source of the contamination of soil and natural waters by toxic heavy metals.

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