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Removal of Ni(II) from Aqueous Solution by Novel *Lycopersicon esculentum* Peel and *Brassica botrytis* Leaves Adsorbents

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Abstract: The current work reports adsorption of Ni(II) using *Brassica botrytis* leaves (BBL), *Brassica botrytis* leaves-activated carbon (BBL-AC), *Lycopersicon esculentum* peel (LEP) and *Lycopersicon esculentum* peel-activated carbon (LEP-AC). The adsorption of Ni(II) was tested in batch experiments by varying different parameters such as pH, initial metal ion concentration, temperature, adsorbent dosage, and contact time. Thermodynamics and kinetics investigations were performed for Ni removal. The adsorption of Ni(II) was improved by incorporation of activated carbon to the parental *Brassica botrytis* leaves and *Lycopersicon esculentum* peel adsorbents. The studies revealed 40 min of equilibrium time for Ni(II) adsorption by different adsorbents. Adsorption of Ni was drastically declined by temperature with a minimum adsorption of 53% observed for BBL. Similarly, solution pH also played a vital role in Ni(II) adsorption by different adsorbents. A 95% adsorption of Ni was recorded in the case of LEP-AC at pH 7. The study concluded with the application of *Lycopersicon esculentum* peel and *Brassica botrytis* leaves as active adsorbents for Ni(II) adsorption from aqueous solution.

Keywords: *Lycopersicon esculentum* peel; *Brassica botrytis* leaves; adsorption; heavy metals; activated carbon; Ni metal

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1. Introduction

Given the rising proliferation of industry, environmental contamination has recently become a larger worry [1]. The natural environment is threatened by pollutants of all kinds, but heavy metal ions are particularly dangerous because they cannot biodegrade [2–5]. In terms of its use in smelting, mining dyeing processes, and battery manufacture, nickel is regarded as a versatile metal [6]. On the other hand, Ni(II) has been reported to have serious health hazards. Nickel carbonyl is classified as a carcinogen that can easily be absorbed by human skin [7]. A 30 mg/L exposure level of nickel carbonyl for a duration of 30 min is recorded as being lethal [8]. Similarly, higher Ni concentration has been documented in nose, ligaments and bone [9,10]. Likewise, the carcinogenic nature of Ni(II) at high exposure level have also been reported by Amit and Minocha [11]. Keeping in view the above facts, the removal of Ni(II) from aqueous solutions has become a subject of upmost importance. Many techniques, such as chemical precipitation, ion exchange and membrane filtration, have been applied for Ni(II) removal from aqueous solution [12]. However, the adsorption technique, owing to its ease of operation, low cost and the availability of adsorbents, is considered an attractive alternative for Ni(II) removal.

A wide range of adsorbents have been investigated for Ni(II) adsorption from aqueous solutions. Khelifi et al. applied physico-chemically modified sewage sludge for the removal

of Ni from aqueous solution [13]. Their study concluded that activated carbon synthesized from the sewage sludge is a beneficial adsorbent for Ni(II) adsorption. Recently, Jordanian natural zeolite has been used as an adsorbent for Ni removal from water [14]. The adsorption capacity of Ni(II) was shown to be high and well-controlled using a chemisorption process that followed the pseudo-second order kinetic model. The dye removal process is both spontaneous and exothermic. The Freundlich isotherm was chosen as the best model out of the three options presented. Another study conducted by Pahlavanzadeh et al. reported zeolite as a cation-exchange adsorbent for Ni(II) adsorption from aqueous solution [15]. In this study, Zn electrodes were found to successfully remove nickel from wastewater by a simple configuration of the electrocoagulation process.

A comparative study between zeolite and modified zeolite revealed that modified zeolite adsorbed 95% Ni(II) from aqueous solution, which accounts for 20% more than adsorbed by the common zeolite. Likewise, bio char has been reported to adsorb Ni from aqueous solution where the adsorption efficiency was affected by the type of feed stock [15,16]. The results demonstrate that willow wood bio char adsorbs Ni(II) by making a complex with –OH and C=O, while cattle manure bio char was recorded to have a low adsorption profile as compared with its feedstock. *Lycopersicon esculentum* is a globally significant vegetable with an overall production of 126 million tons in 2005. It is a superb wellspring of numerous supplements and auxiliary metabolites that are essential for human wellbeing; mineral matter, vitamins C and E, B-carotene, lycopene, flavonoids, natural acids, phenolics and chlorophyll. *Lycopersicon esculentum* peel (LEP), with a quasi-spherical morphology has been utilized for different applications [17–20]. Similarly, *Brassica botrytis* leaves (BBL) have also been reported in the literature for a variety of applications [21–23]. Keeping in mind their enriched profiles, LEP and BBL have been investigated for Ni(II) adsorption from aqueous solution. Similarly, activated carbon, being a widely used adsorbent, has also been incorporated with these two parent adsorbents to further investigate the performance of adsorbents and elaborate the future potential of these materials. The significance of the current work is application of LEP and BBL alone and their incorporation with activated carbon to study the adsorption of Ni metal.

The aim and objective of the current work is the application of *Lycopersicon esculentum* peel (LEP), *Lycopersicon esculentum* peel doped with activated carbon (LEP-AC), *Brassica botrytis* leaves (BBL) and *Brassica botrytis* leaves doped with activated carbon (BBL-AC) for Ni removal. Furthermore, adsorption studies were undertaken with different reaction parameters to investigate their adsorption capacities with regards to Ni(II) uptake from aqueous solutions.

2. Experimental

2.1. Sample Collection and Adsorption Studies

Samples were collected as solid wastes. The collected material were washed by tap and distilled water several times to remove heavy metal contents. These collected materials were converted into small, round pieces of about 1–2 cm and were then dried in sunlight and then in an oven at 383 K until they reached a constant weight, which was obtained in 48 h. Then, the dried material was ground to a fine powder and was sieved through 600 μ size. Adsorbents were dried and digested in acids before their application. All four adsorbents were tested for their adsorption of Ni from water solutions of 50, 100, 150, 200 and 250 ppm solutions of Ni at different reaction parameters. The Ni concentrations were measured by an inductively coupled plasma–optical emission spectrometry (ICP-OES) technique. In the current work, a Thermo Scientific iCAP 7000 series ICP spectrometer with model 7400 was used.

2.2. Instruments and Materials

HCl purchased from MERCK Germany and HNO_3 purchased from Riedel-DeHaan were used in the current work. Nickel sulfide purchased from Riedel-DeHaan was utilized as nickel salt. The 10% HNO_3 solution was used for washing the glassware, which was

followed by washing with distilled water. The two adsorbents, LEP and BBL, were washed with distilled water. Then, they were converted to round pieces of 1–2 cm in size, dried in the open air followed by drying in an oven at 383 K [24,25]. The process of drying was continued for about 48 h to obtain a stable weight. The dried adsorbents were fine ground in order to pass through sieved mesh of 600 μ size.

The dried adsorbents were placed in a crucible and heated in a furnace for 4 h at 823 K. Then, they were cooled in a desiccator to attain room temperature. The adsorbent samples were dissolved in 6 M HNO_3 solution and diluted with distilled water.

The adsorbents were placed in a 250 mL flask and mixed with 8 mL of concentrated HCL and 3 mL concentrated HNO_3. The mixture was tightly enclosed for 24 h. Afterward, the tightly closed mixture was heated to reduce the total volume content to 1 mL. The mixture was then filtered into a volumetric flask and diluted to the desired volume.

To incorporate the activated carbon, the known amount of adsorbent was mixed with activated carbon and the same procedure was repeated as mentioned above. Finally, the parent adsorbents and modified adsorbents with activated carbons were investigated for their Ni(II) adsorption.

2.3. Equilibrium Adsorption Isotherms

The link between the adsorbate and adsorbent's equilibrium was best explained using sorption isotherms. The relationship between the mass of adsorbents and the mass of the components of the adsorbate was described using biosorption isotherms. The analysis of the environment of the adsorption process was carried out by Freundlich isotherms. This kind of isotherm model involves surfaces with diverse binding sites and interactions between sorbed species. The Freundlich equation makes it obvious that the sorption energy will decrease after the sorption center of the adsorbents is complete. The Freundlich equilibrium constants are often calculated from the plots of log Qeq versus log Ceq. The value of n indicates the degree of nonlinearity: if n 1, adsorption is a chemical process; if n = 1, adsorption is linear. Adsorption is a physical process if n is greater than 1.

$$qe = Kf\ Ce\ 1/n$$

where;
qe: amount adsorbed per unit weight of adsorbent at equilibrium (mg/g), (mol/g).
Ce: equilibrium concentration of adsorbate in solution after adsorption (mg/g), (mol/L).
Kf: empirical Freundlich constant or capacity factor (mg/g), (mol/g).
1/n: Freundlich exponent.
$\log qe = \log Kf + 1/n \times \log Ce$.

Such a plot yields a straight line with a slope equal to 1/n and an intercept equal to log Kf.

2.4. Biosorption Kinetics

Predicting biosorption rate is crucial since it provides details for developing batch biosorption systems. The sorption kinetics are crucial to the process of purifying wastewater because they provide important insights into the reaction's routes and the most effective mechanisms. Ho's pseudo second-order kinetics were carried out.

The pseudo second order kinetic model as proposed by Ho and McKay is expressed as follows:

$$t/qt = 1/k2qe2 + 1/qe \times t$$

where;
qe: amount of metal adsorbed at equilibrium time (mg/g), (mol/g)
qt: amount of metal adsorbed at any time (mg/g), (mol/g)
k2: pseudo second order rate constant (g/mg.min), (g/mol.min).

2.5. Adsorption Studies

Batch experiments were performed to study the Ni(II) adsorption by different adsorbents. The adsorption process was studied at different initial metal ion concentrations namely, 50, 100, 150, 200 and 250 ppm with different reaction parameters such as adsorbent dosage, pH, temperature, and contact time. The adsorption of Ni was evaluated by varying one parameter while keeping all other parameters constant. The details of adsorption parameters are included in the obtained graphs during the adsorption studies. The amount of metal removed from the solution by the adsorbent was calculated by using the following formula;

$$Q = V\,(Ci - Cf)/S \quad (1)$$

Q = Metal ion uptake capacity, Ci = initial metal ion concentration of the solution before the sorption analysis (mg/L), Cf = final metal ion concentration of the solution after the sorption analysis (mg/L), S = dry weight of adsorbents in grams and V = volume of the solution.

3. Result and Discussion

3.1. Adsorption Studies

Ni(II) ion adsorbs onto *Lycopersicon esculentum* peel and its activated carbons, as well as *Brassica botrytis* leaves and its activated carbon, according to a Freundlich adsorption isotherm (Figure 1) associated with data in Table 1. Reem et al. studied removal of Ni(II) ions from aqueous solutions by using analcime and found the Freundlich adsorption isotherm to be the best fitted adsorption isotherm for Ni removal.

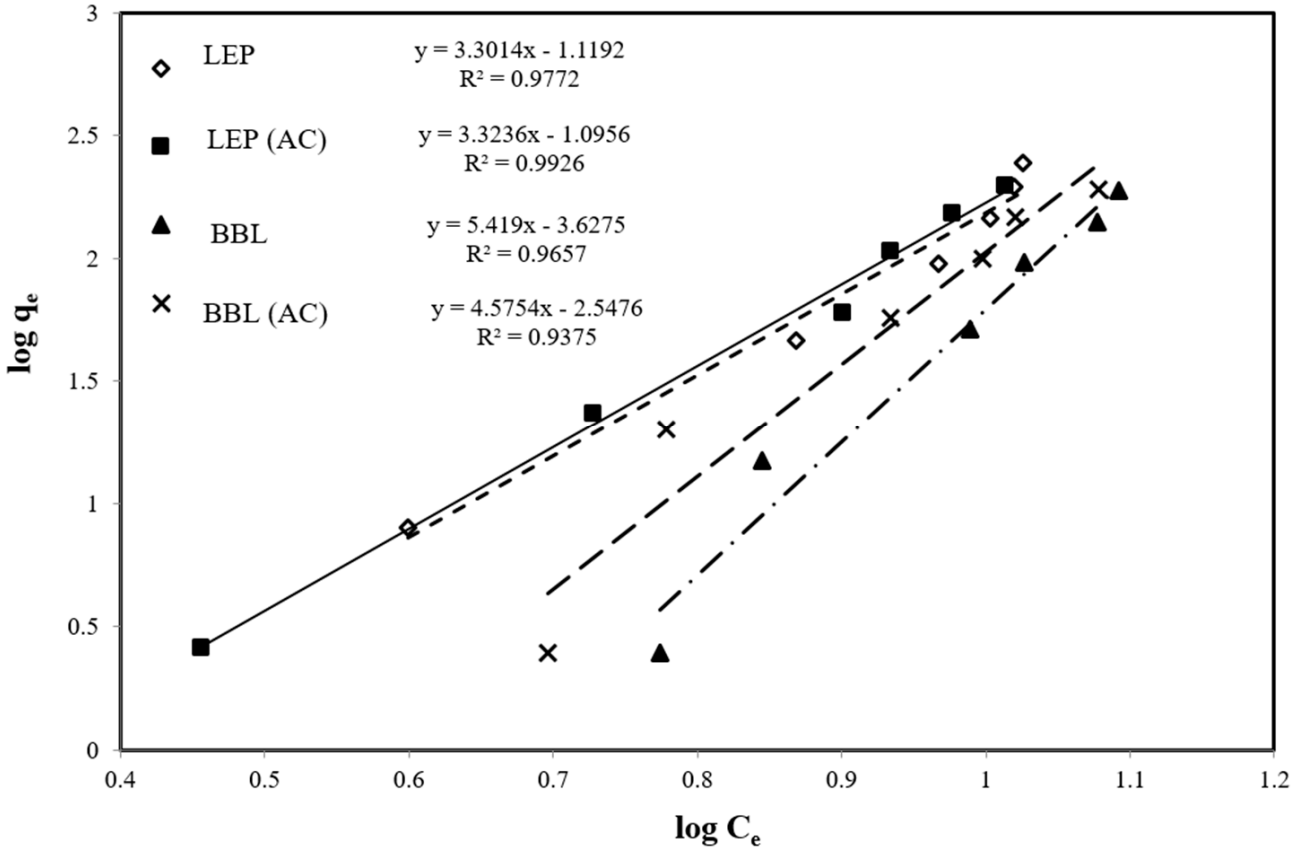


Figure 1. Freundlich adsorption isotherm for adsorption of Ni(II).

Table 1. Freundlich parameters for the adsorption of Ni(II) ion on different adsorbents.

Adsorbents	K_f (mg/g)	1/n	n	R^2
LYCOPERSICON ESCULENTUM PEEL	0.048	3.301	0.302	0.977
LYCOPERSICON ESCULENTUM PEEL (AC)	0.336	2.296	0.435	0.992
BRASSICA BOTRYTIS LEAVES	0.260	5.419	0.198	0.965
BRASSICA BOTRYTIS LEAVES(AC)	0.310	4.575	0.248	0.937

3.2. Kinetics Studies

Ho's pseudo second-order kinetics were carried out to investigate the kinetics of Ni removal. As depicted by Figure 2, the removal of Ni was governed by Ho's pseudo second-order kinetics. Similar observations were also recorded by R.K. Shah et al. [26].

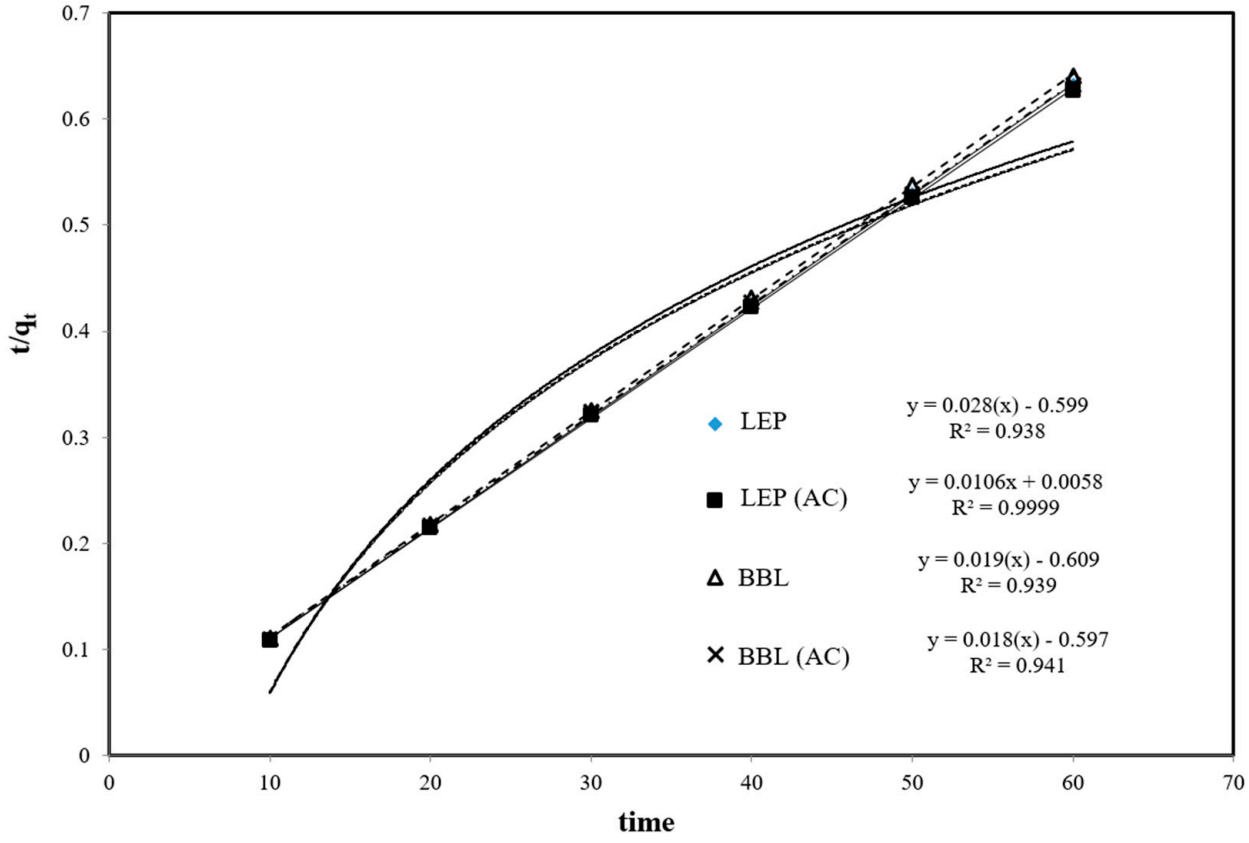


Figure 2. Ho's pseudo second-order kinetics of Ni removal.

The kinetic parameters for the adsorption of Ni(II) ion on different adsorbents are included in Table 2. The tabulated data indicate that the Ni adsorption follows pseudo second-order kinetics as confirmed by R^2 values.

3.3. Effect of Initial Metal Ion Concentration

The percentage of adsorption was affected by the alteration in metal concentrations. As depicted by Figure 3, the percent adsorption was decreased by increasing the metal concentration [27]. This is understandable as more active adsorption sites are available for metal at lower concentration. By increasing the metal concentrations, the active adsorbent sites are saturated and, consequently, the percent adsorption is decreased. The comparative study of the LEP and BBL absorbents for Ni adoption revealed that BBL is a more active

adsorbent as compared with the LEP counterpart. Furthermore, as is evident from the figure, the incorporation of activated carbon to both parent LEP and BBL adsorbents enhanced the percentage uptake of Ni(II).

Table 2. Kinetic parameters for the adsorption of Ni(II) ion on different adsorbents.

Adsorbents	K_e (mg/g)	K_2 g.mg−1.min−1	R^2
LYCOPERSICON ESCULENTUM PEEL	19.585	0.002	0.938
LYCOPERSICON ESCULENTUM PEEL (AC)	18.575	0.013	0.999
BRASSICA BOTRYTIS LEAVES	18.756	0.021	0.939
BRASSICA BOTRYTIS LEAVES(AC)	19.944	0.024	0.941

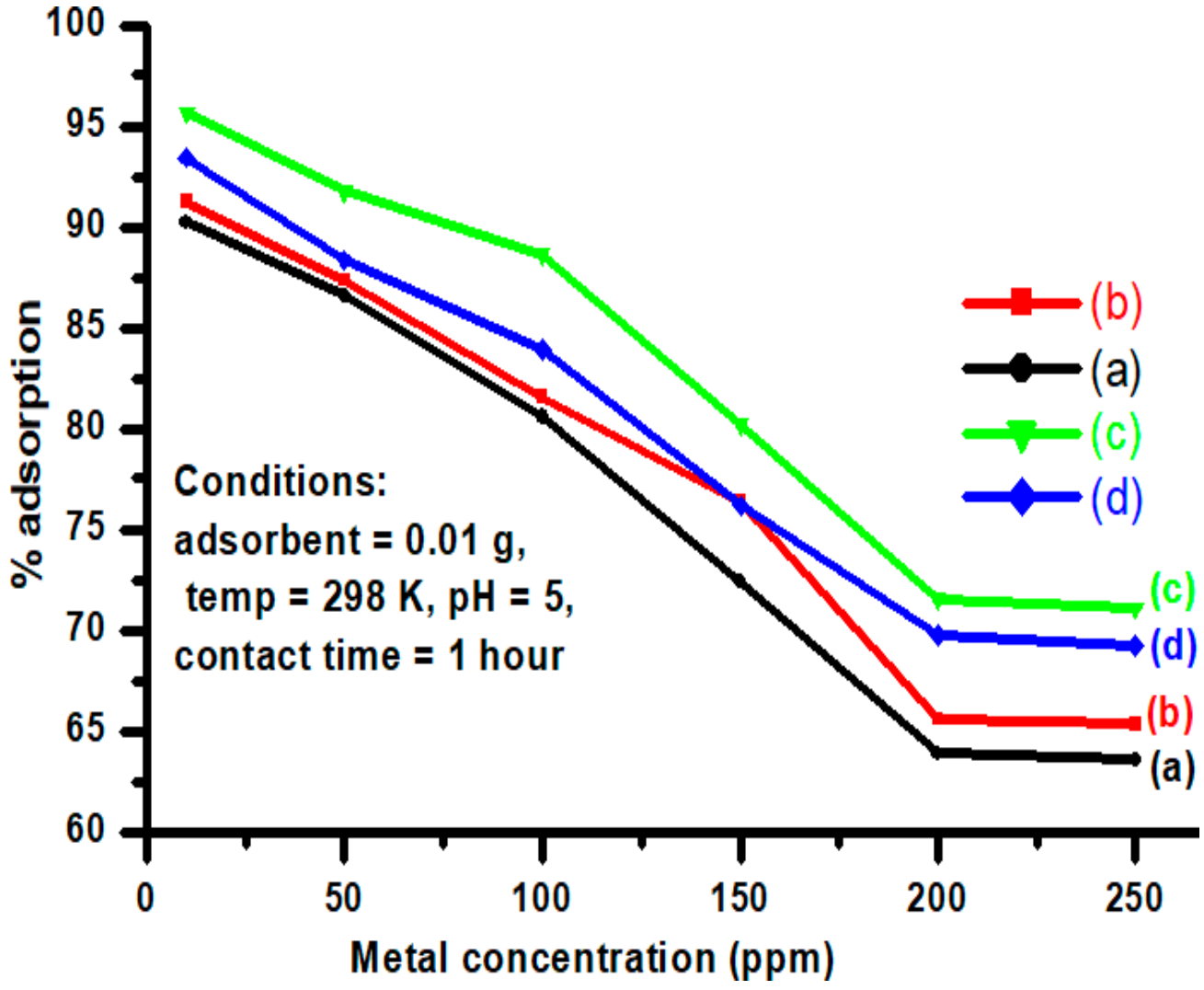


Figure 3. Percentage adsorption of Ni(II) by (a) LEP, (b) BBL, (c) LEP-AC and (d) BBL-AC vs. metal concentration.

This indicates the promoting role of activated carbon in Ni(II) adsorption from the aqueous solution. Such observations have also been recorded in the literature [28,29].

3.4. Effect of Adsorbent Dosage

The variation in adsorbent dosage also affected the percent adsorption in the current case. As shown by Figure 4, irrespective of the adsorbent, when the adsorbent dosage was increased the percent adsorption was increased in all cases. The increase in percent adsorption as a consequence of increasing adsorbent dosage can be justified by the fact that total number of active adsorption sites are increased by increasing adsorbent dosage. Therefore, the resultant pattern in increasing adsorption percentage is justified by adsorbent dosage.

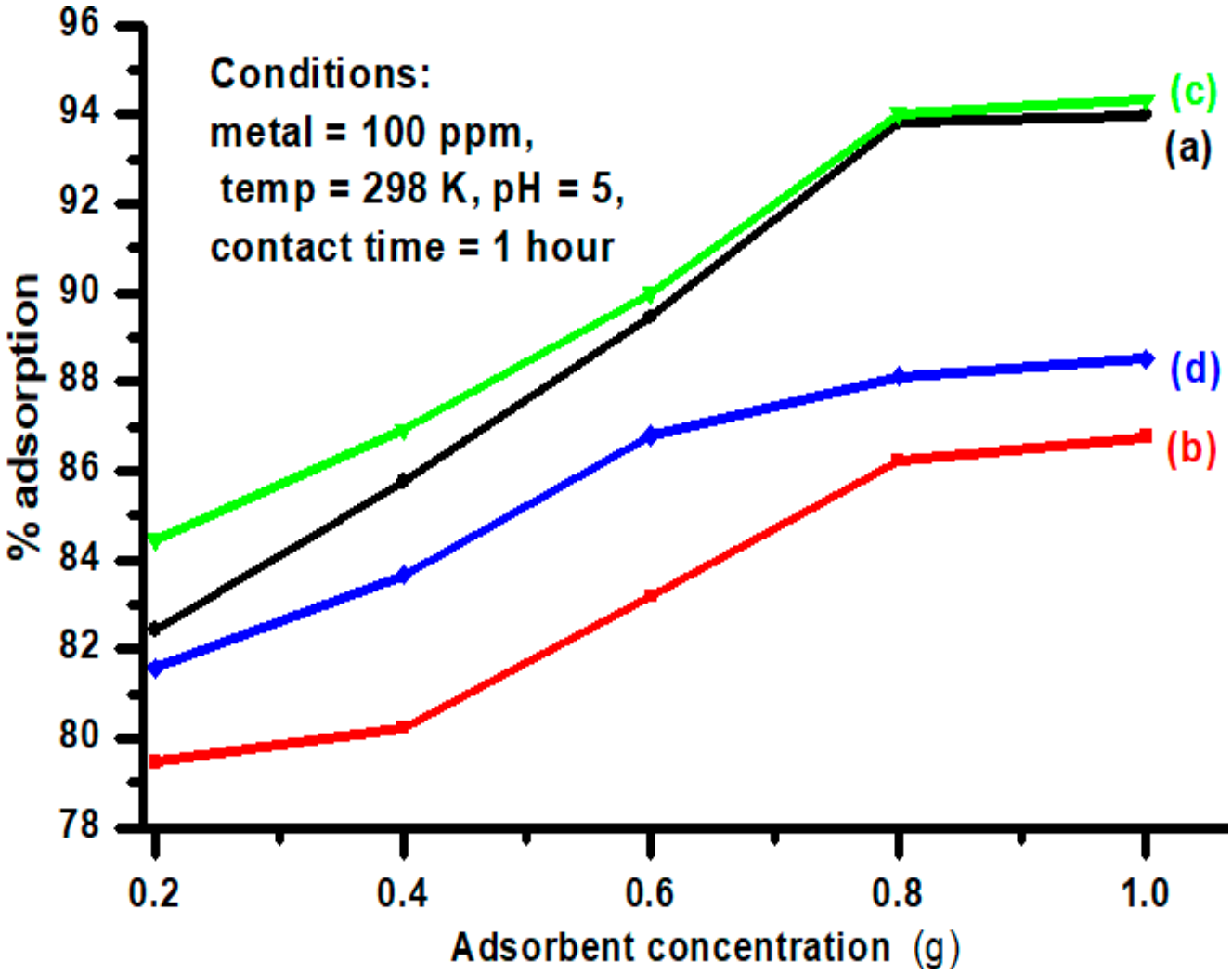


Figure 4. Percentage adsorption of Ni(II) by (a) LEP, (b) BBL, (c) LEP-AC and (d) BBL-AC vs. adsorbent dosage.

Although the percentage adsorption was enhanced in all cases, the results could be improved further by incorporating activated carbon adsorbents as compared with their parent adsorbents. This trend has also been observed in the literature [30].

3.5. Effect of Temperature

The adsorption process was altered by variation in temperatures in the studied zone of 293–343 K. As displayed in Figure 5, the percent adsorption was adversely affected by magnification in adsorption temperature in line with the literature [31]. This observation confirms the exothermic nature of the Ni (II) adsorption over all studied adsorbents.

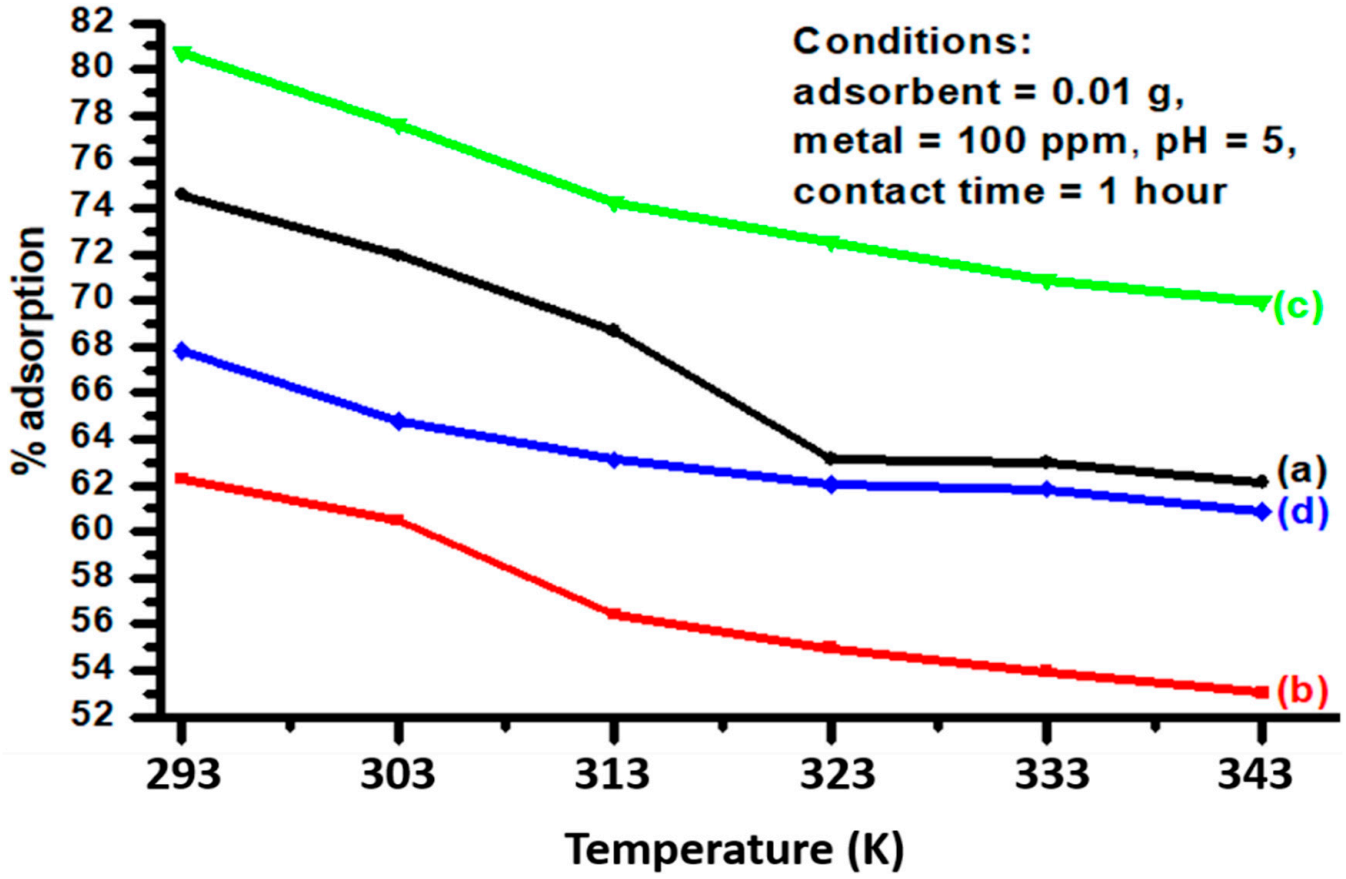


Figure 5. Percentage adsorption of Ni(II) by (a) LEP, (b) BBL, (c) LEP-AC and (d) BBL-AC vs. temperature.

Furthermore, the influence of adsorption temperature investigations also demonstrated the promoting role of activated carbon by showing an almost 10% increase in Ni(II) percent adsorption over the adsorbent incorporating activated carbon as compared with parent adsorbents. The enhanced performance of activated carbon-incorporated adsorbent could be justified by the potentially greater surface area associated with activated carbon.

3.6. Effect of pH

The effect of pH on Ni(II) adsorption by different adsorbents can be visualized by Figure 6. As evident from the figure, percent adsorption was incremented by increasing the magnitude of pH. This trend of increasing adsorption by increasing pH could be apprehended by fact that the concentration of H+ ion is higher at lower pH, with the adsorbent surface positively charged. This way, the interaction between positively charged metal and positively charged adsorbent is minimized. However, when the pH of the solution is increased the concentration of H+ decreases, making the surface negative charged and consequently favoring the interaction of positively charged metal and negatively charged adsorbent. Therefore, the percentage adsorption was increased by increasing pH and the optimum magnitude of pH was recorded for Ni adsorption over all the studied adsorbents.

However, a further rise in pH the percentage decreased the adsorption. This decline in percentage adsorption through increasing pH could be justified by the precipitation phenomenon due to the relevant hydroxides. The comparative studies of the Ni adsorption at different pH also revealed the promoting role of activated carbon when it is incorporated with the parent adsorbents.

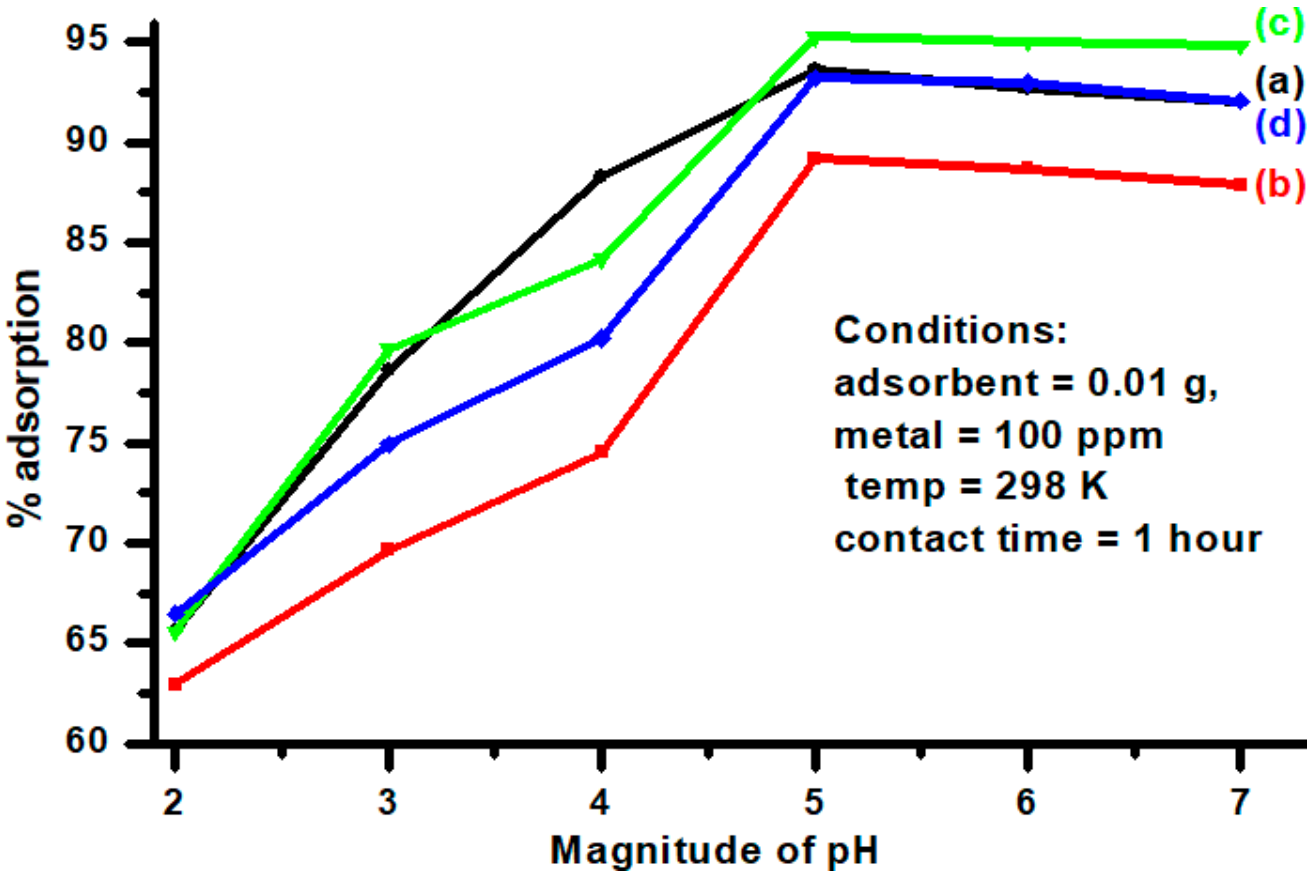


Figure 6. Percentage adsorption of Ni(II) by (a) LEP, (b) BBL, (c) LEP-AC and (d) BBL-AC vs. pH.

3.7. Effect of Contact Time

As with all other parameters, the adsorption was also influenced by variation in contact time. As displayed by Figure 7, the percentage adsorption was found to be linearly increased by increasing the contact time. A sharp increase in percentage adsorption was recorded from the beginning of the contact time until around 40 min of contact time. Nevertheless, the percentage adsorption was observed to be almost constant with a further increase in contact time. This trend could be justified by the fact that the rate of adsorption is faster in the beginning of the adsorption process, as more active adsorption sites are available. However, with the passage of time, since the number of active adsorption sites are decreased, the rate of adsorption is decreased. In the current case, the percent adsorption was increased until an equilibrium time of 40 min was reached for Ni(II) adsorption. The slight variation was recorded in terms of equilibrium time for each adsorbent as demonstrated by the figure. The observed trend of increasing adsorption with increasing contact time is in line with the literature [32].

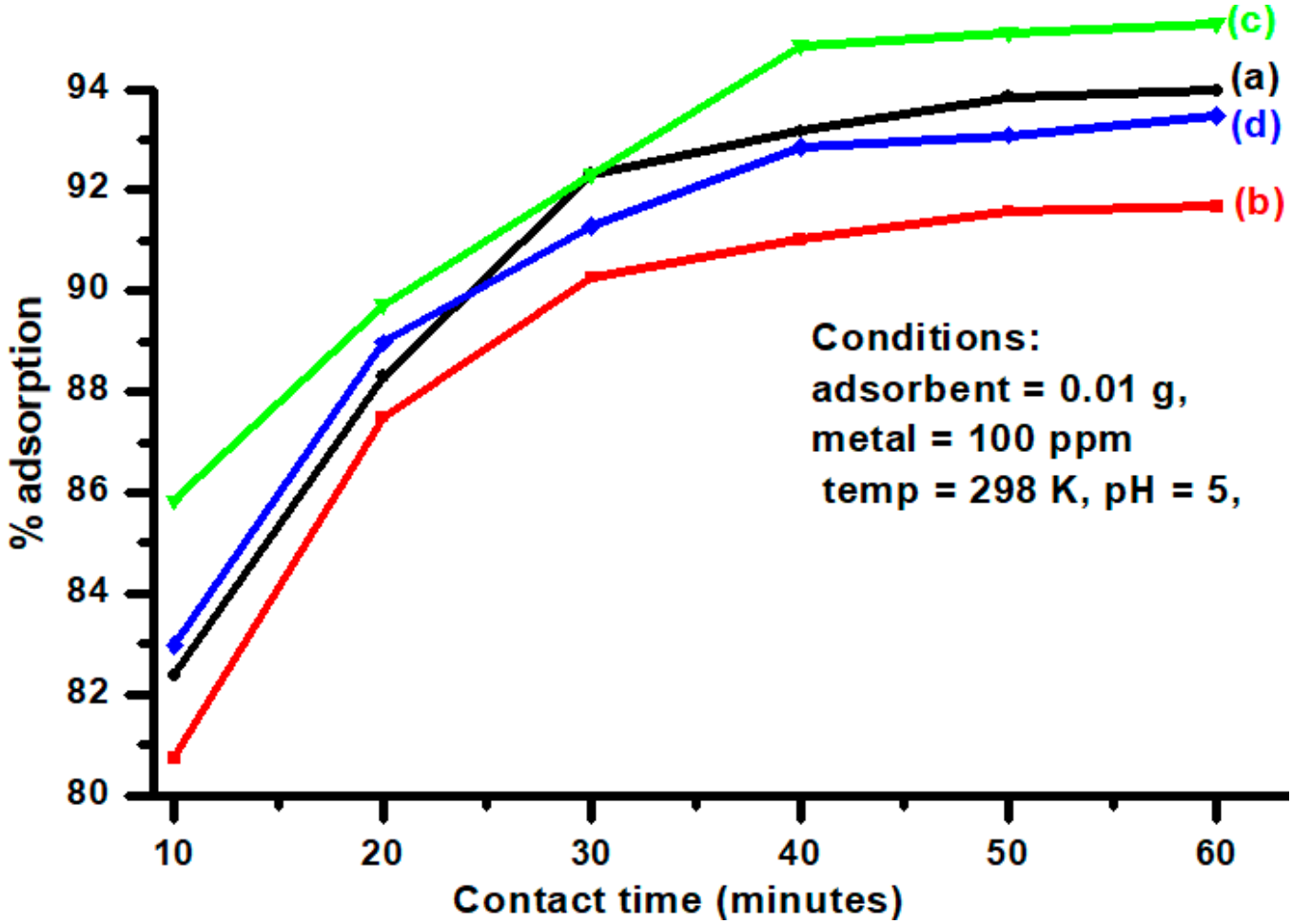


Figure 7. Percentage adsorption of Ni(II) by (a) LEP, (b) BBL, (c) LEP-AC and (d) BBL-AC vs. contact time.

4. Conclusions

The parent and activated carbon incorporated *Lycopersicon esculentum* peel and *Brassica botrytis* leaves were investigated for their ability to remove Ni(II) from aqueous solution. The overall performance determined the promoting role of activated carbon incorporation as compared with their parent counterpart. Adsorption efficiency declined with increasing Ni concentration in water and reaction temperature. However, this was incremented by adsorbent concentration, reaction pH and contact time. The work concluded that adsorption of Ni(II) was influenced by different reaction parameters. However, in each case, adsorbent that incorporated activated carbon showed better results for NI(II) adsorption. Thermodynamics and kinetics investigations revealed that adsorption studies were governed by Freundlich adsorption isotherm and second-order kinetics, respectively.

Author Contributions: Conceptualization, I.U.D., Q.U.S., S.T., A.N., A.I.A. and M.A.A.; methodology, I.U.D., Q.U.S., S.T. and A.N.; validation, I.U.D., Q.U.S., S.T., A.N., A.I.A. and M.A.A., data curation, Q.U.S. and S.T.; writing—Q.U.S., S.T., A.N.; writing—review and editing, I.U.D., A.I.A. and M.A.A.; project administration, I.U.D., Q.U.S. and S.T.; funding acquisition, I.U.D. and M.A.A. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

Research Highlights:

✓ *Lycopersicon esculentum* peel and *Brassica botrytis* leaves were used as adsorbents.
✓ Activated carbon was incorporated in *Lycopersicon esculentum* peel and *Brassica botrytis* leaves.
✓ Ni adsorption was improved by AC incorporation.
✓ Ni adsorption was effected by reaction parameters.

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