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EVALUATION OF REMOVAL EFFICIENCY OF NI (II) FROM AQUEOUS SOLUTION BY NATURAL LEAVES

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ABSTRACT

Activated carbon was prepared from Behda, Anjan, Chinch and Bakam Neem leaves for the removal of Ni (II) from stimulated waste water. The effects of various parameters such as initial metal concentration, particle size, pH and contact time for the adsorption of Ni (II) on BEAC, AAC, CAC and BNAC were investigated. The amount of adsorbent increased the percentage of metal removal increased accordingly. The optimum pH for the Ni (II) absorption was 7.0 except BEAC. The equilibrium data fit well with both Langmuir and Freundlich models of adsorption. The value of separation factor RL was found to be 0.094, 0.026, 0.030 and 0.025 for BEAC, AAC, CAC and BNAC respectively suggesting the isotherm to be favorable at the concentration studied. The percentage removal of nickel ion on to BEAC, AAC, CAC and BNAC were very significant.

Keywords: Copper ion, Behda, Anjan, Chinch, Bakam Neem, Langmuir, Freundlich isotherms.

INTRODUCTION

Heavy metals are toxic inorganic contaminants that, unlike organic contaminants that can be degraded by microorganisms, must be removed from wastewater before being discharged to the environment.[1] Several environmental and health problems, associated with the metal contamination of the natural systems.[2] Heavy metal contamination may cause changes in the physicochemical composition of the water and finally become unsuitable for human consumption[3] .Nickel is well known heavy metal pollutant. Nickel is extensively used in electroplating, the manufacturing of steel, electronic devices, ceramics and colored glasses. [4]. The nickel presence in natural potable waters is limited up to 0.01 mgL^{-1} since January 01, 2008, according to the WHO recommendation.[5] The higher concentration of Ni causes poisoning effects like headache, dizziness, nausea, tightness of the chest, dry cough, vomiting, and chest pain, shortness of breath, respiration, cyanosis and extreme weakness.[6] Nickel causes more hypersensitivity than any other metal and research conducted by the asthma and allergy centers indicated that 14.2% of the population suffers from nickel hypersensitivity.[7] These harmful effects of Ni (II) necessitate its removal from waste water before release in to streams.[8]Techniques used for removal of heavy metals, like chemical precipitation, lime coagulation, ion exchange, reverse osmosis and solvent extraction are expensive and non-environmental friendly, as compared to adsorption.[9] It is therefore, essential to search agricultural by-products and to transform such materials to adsorbents. Among several chemical and physical methods, the adsorption onto activated carbon has been found to be superior to other techniques because of its capability of adsorbing a broad range of different rapid types of adsorbate efficiently and its simplicity of design. [10] In recent years, special attention has been focused on the use of natural adsorbents as an alternative to replace the conventional adsorbents, based on both the environmental and the economical points of view. Natural materials that are available in large quantities, or certain waste products from industrial or agricultural operations, may have potential as inexpensive sorbents.[11]There are various synthetic and natural waste like Ricinus Communis Paricarp[12], Lewatite S-100, Potato Peels[10], Maize Cob[8], Aniline-Formaldehyde Polymer[13], Neem Leaves[14], Mosambi Fruit Peelings[3] etc. have been used for the removal of Ni (II). Tea factory waste has been used as adsorbent for copper (II) and cadmium (II) by S.C. ay et al[15].

The aim of this work is to study the removal of Ni (II) metal ions by activated carbon prepared from Behda, Anjan, Chinch, Bakam Neem leaves from synthetic waste water and to offer this adsorbent as local replacement for existing commercial adsorbent materials.

EXPERIMENTAL

Materials

The leaves of Behda, Anjan, Chinch and Bakam Neem were collected from local field of Jalgaon District. These waste materials were washed with water, dried in sunlight, then 60 ^{0}C for 24 h in hot air oven. Finally, the dried leaves were ground in clean electric mixer and stored in a clean plastic bag.

Preparation of activated carbon from adsorbent

Powder of dried leaves is treated with conc. H_2SO_4 at a weight of ratio 1:1. The resulting black product was kept in an oven maintained at 500^{0}C for 12 h followed by washing with $NaHCO_3$ and water until free from excess acid, the pH of washing becomes 7.0 and dried at 150 $\pm$ 5^{0}C. The resulting products were grounded and sieved into different particle sizes range A) 500-850 micron B) 180-500 micron C) 45-180 micron and stored in a tight lid container for further studies. The physical properties were analyzed by usual standard methodologies.

Preparation of Ni (II) Solution

The stock solution of Ni (II) ion was prepared by dissolving 4.049 gm of $NiCl_2$. $6H_2O$ in 1000 ml volumetric flask and make up to the mark with double distilled water. All the required working solutions were prepared by diluting the stock solution with double distilled water. Batch adsorption studies were performed at room temperature.

Experimental Methods and Measurement

The adsorption experiments were carried out by agitating the carbon with 50. 75. 100 mg/l metal ion solution of desired concentration at pH 7.0 and at room temperature in a mechanical Shaker (120 rpm) after a defined time interval, samples were withdrawn from the shaker, filtered by Whatman filter paper No. 1 and the supernatant solution was analyzed for Ni (II) ion concentration using an Atomic Absorption Spectrometer (Thermo scientific S-series AA Spectrometer) at 232.0 nm.

RESULTS AND DISCUSSION

The four natural easily available adsorbents were selected from local area. The Table-1 shows selected adsorbents from natural waste.

Characteristics of the adsorbent

The physico-chemical properties of the prepared activated carbon were determined by standard methods and presented in Table-2.

Effect of Agitation time on Nickel (II) adsorption

The effect on agitation time on the removal percent of the Ni (II) metal ions by the BEAC, AAC, CAC and BNAC is shown in Figure-1. The effect of agitation time was studied in the range of 15, 30, 45, 60, 75, 90, and 120 min. It reveals that the percent removal of Ni (II) metal ion present in the aqueous solution was in progression with time on all selected adsorbents. The results also showed that, the adsorption was fast at initial stage of contact period and after that near the equilibrium it became slower. Adsorptions reach equilibrium within 120 min for all selected adsorbent. From the result of the adsorption experiment Ni^{2+} ions had the highest percent removal of 98.3118 at the end of 120 min by the BNAC, followed by AAC, CAC and BEAC with 98.1503, 98.2726 and 96.0286, respectively. The percent adsorption vs time plot in Figure-1 was smooth and continuous leading to saturation, suggesting the possible monolayer coverage of metal ions on the surface of the adsorbent.[8]

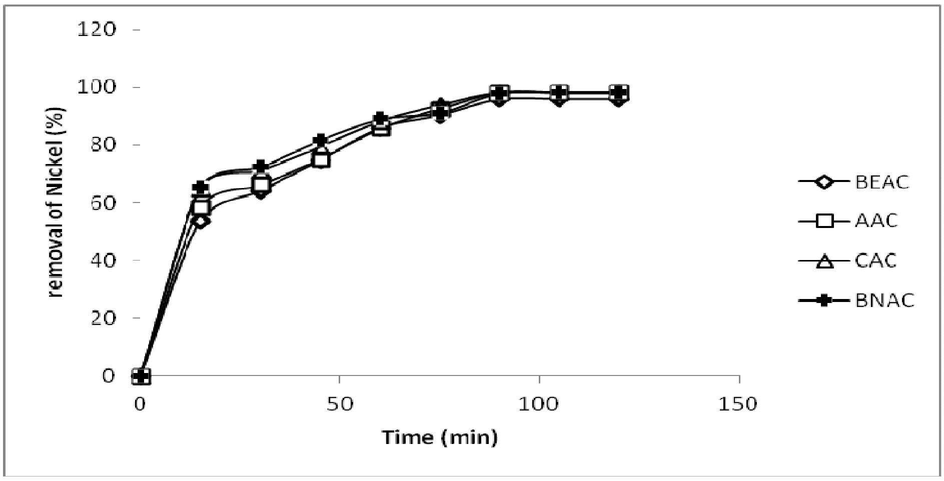

Fig.-1: Effect of contact time on the removal of Nickel by the BEAC, AAC, CAC and BNAC as adsorbent at 50 ppm, pH 7, 0.4 g/100ml adsorbent dose and particle size 45-180 micron.

Effect of particle size

To study the effect of particle size, adsorbent particles of sizes A= 500 -850, B= 180-500, C= 45-180 micron were used. In each study 0.4g of adsorbent in 100 ml of 50 mgL-1 of Ni (II) solution was agitated to equilibrium time of 120 min, the adsorbent was separated and the supernatant solution was analyzed for Nickel concentration and the results are shown in figure-2.

It is evident from figure-2 that increase in particle size decreased the percentage of removal. At a fixed adsorbent dosage, the decrease in particle size increases the metal uptake. The increase in the uptake by smaller particles was due to the greater accessibility to pores and to the greater surface area for bulk adsorption per unit mass of the adsorbent. Similar trend had been observed on removal of Ni (II) by using Powder of Mbonambi Fruit Peelings.[8]

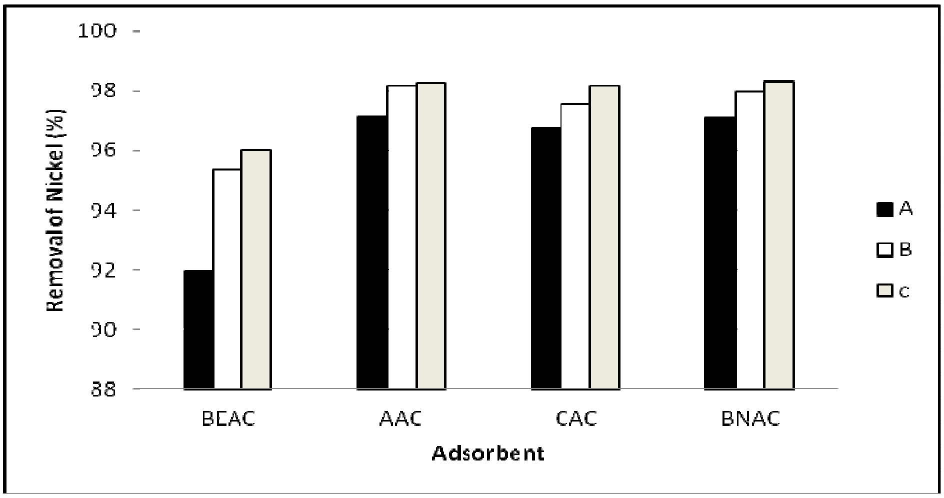

Fig.-2: Effect of particle size on the adsorption of Ni (II) ions (Initial concentration: 50 mg/ l, contact time: 120 min, pH: 7.0, temperature: 301 ± 1 K, adsorbent dosage: 0.4 g/100 ml, Particle size: A= 500 -850, B= 180-500, C= 45-180 micron)

Effect of pH

The pH of solution played an important role for adsorption of metal ions. To determine the pH effect on adsorption capacity of Ni (II) ions, solutions were prepared at different pH levels from (2-9) the results are given in Fig.-3. The adsorption values were increased with increasing pH values for both adsorbent due to lesser number of H+ and greater number of surface ligands with negative charges. The optimum pH for removal of Ni^{2+} ions is fixed as 7.0 for AAC, CAC and BNAC except BEAC (pH= 6). The neutral pH is found to be favorable .[14]

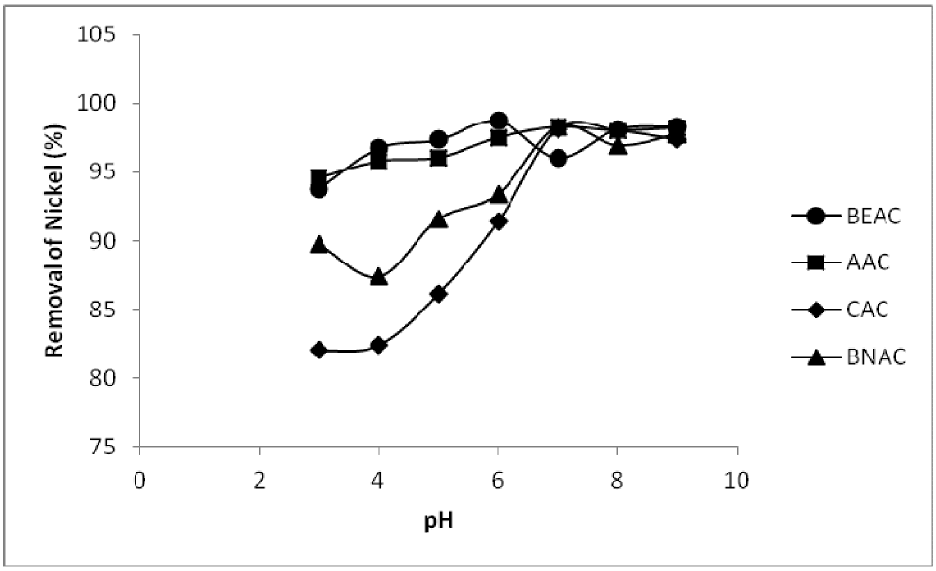

Fig.-3: Effect of pH on Nickel adsorption by BEAC, AAC, CAC and BNAC leaves at an initial concentration of 50 ppm, 120 min, and 0.4 g/100 ml adsorbent dose.

Effect of Initial Concentration of Ni (II)

In batch adsorption processes, the initial metal ion concentration of metal ions in the solution plays a key role as a driving force to overcome the mass transfer resistance between the solution and solid phase. The effect of initial metal ion concentration ranging from 50-100 mg/1 on BEAC, AAC, CAC and BNAC was studied by taking different concentrations of Ni (II) solutions at pH 7, while keeping the dosage of the adsorbent 0.4g/100 ml constant and temperature at 28 ^{0}C. The results are tabulated in Table-3.

It is indicated from the Table-3 that the percent adsorption decreases with the increase of initial metal ion concentration. As a result of the above observations, it is indicated that the adsorption process of Ni (II) ions on BEAC, AAC, CAC and BNAC has to be dependent on concentration of the metal ion solution up to some extent.

Adsorption Isotherms

The adsorption data was analyzed with the help of the Freundlich and Langmuir isotherms.

The Langmuir isotherm is represented by the following equation-

$$(Ce/qe) = (1/ b\, Q_0) + (Ce/Q_0) \qquad (1)$$

Where,

qe = amount of metal ion adsorbed at equilibrium (mg/g),

Ce = equilibrium concentration of metal ion (mg/L),

Q_0 = Langmuir Constant related to adsorption efficiency (mg/g) and

b = Langmuir Constant related to energy of adsorption (L/mg) [4].

The linear plots of Ce/qe versus Ce suggest the applicability of the Langmuir isotherms (Fig.-4 a, b, c, d) for BEAC, AAC, CAC and BNAC respectively. The values of Qo and b were determined from the slope and intercept of the plots (Table 4). The high value of correlation coefficient r^2 from Table 8 indicates that the adsorption of Ni (II) ion by BEAC, AAC, CAC and BNAC follows Langmuir isotherm model. The Table-4 also gives information about the adsorption capacity (Qo) and energy of adsorption b of BEAC, AAC, CAC and BNAC adsorbents.

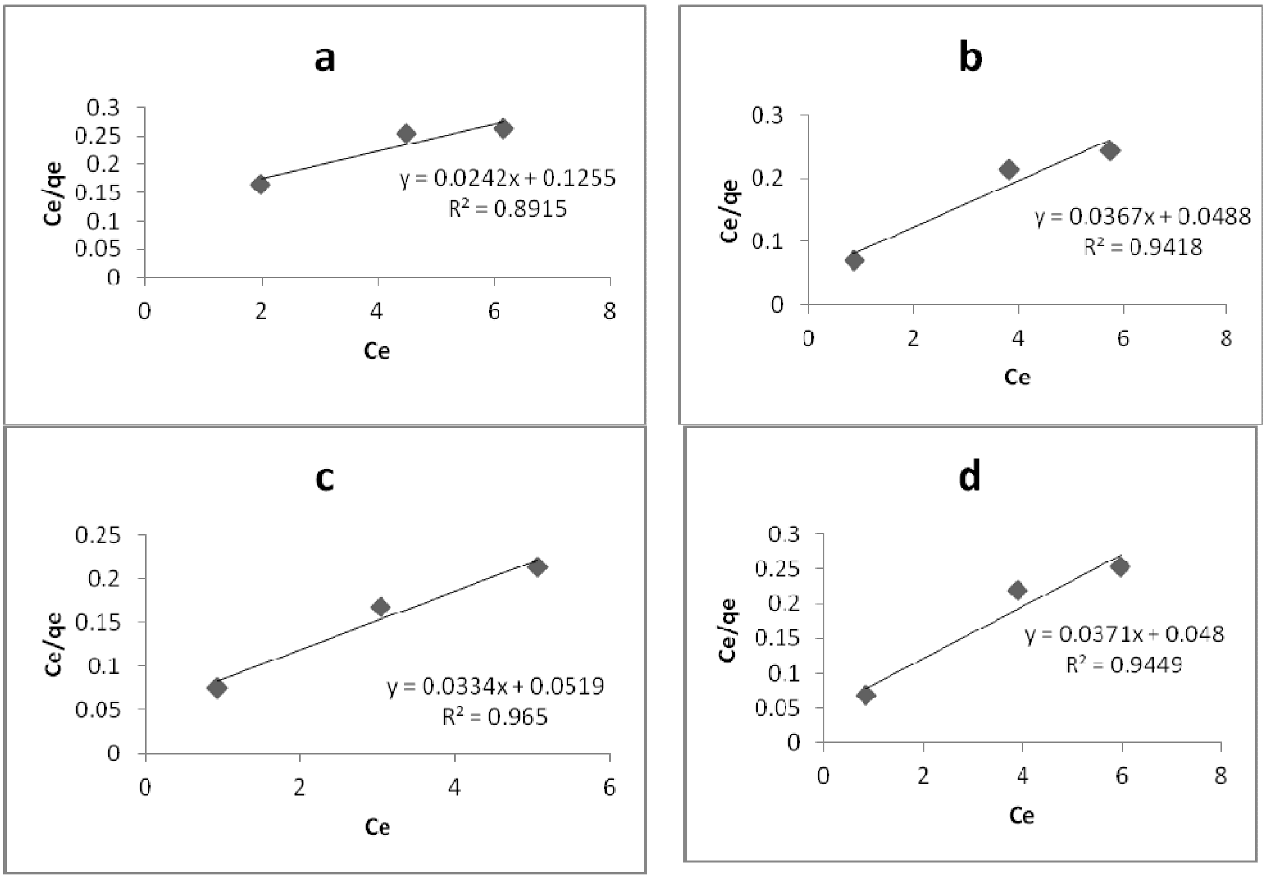

Fig.-4: Langmuir isotherm for the removal of Ni (II) by adsorption on (a) BEAC. (b) AAC. (c) CAC (d) BNAC.

To confirm favorability of the adsorption process, the separation factor (R_L) is calculated and presented in Table 4. The values are found to be between 0 and 1 and confirmed that the ongoing adsorption process is favorable. The similar result of RL was obtained by S. Madhavakrishan.[12]

$R_L = 1/ (1+ bCi)$ (2)

Here, b is the Langmuir constant and Ci is the initial concentration of Ni (II).

The Freundlich isotherm is also employed for the adsorption of Ni (II) on the adsorbent. The Freundlich isotherm is represented by the following equation-

$\log qe = \log kf + (1/n) \log Ce$ (3)

Here, qe is the amount of Ni (II) adsorbed (mg/l), Ce is the equilibrium concentration of Ni (II) in the solution (mg/l) and kf and n are constant incorporating all factors affecting the adsorption capacity and intensity of adsorption, respectively. The plot of log qe versus log Ce (Fig.-5 a, b, c and d) suggest that applicability of Freundlich isotherm for BEAC, AAC, CAC and BNAC. The values of kf and n were determined from the slope and intercept of the plots (Table-4).

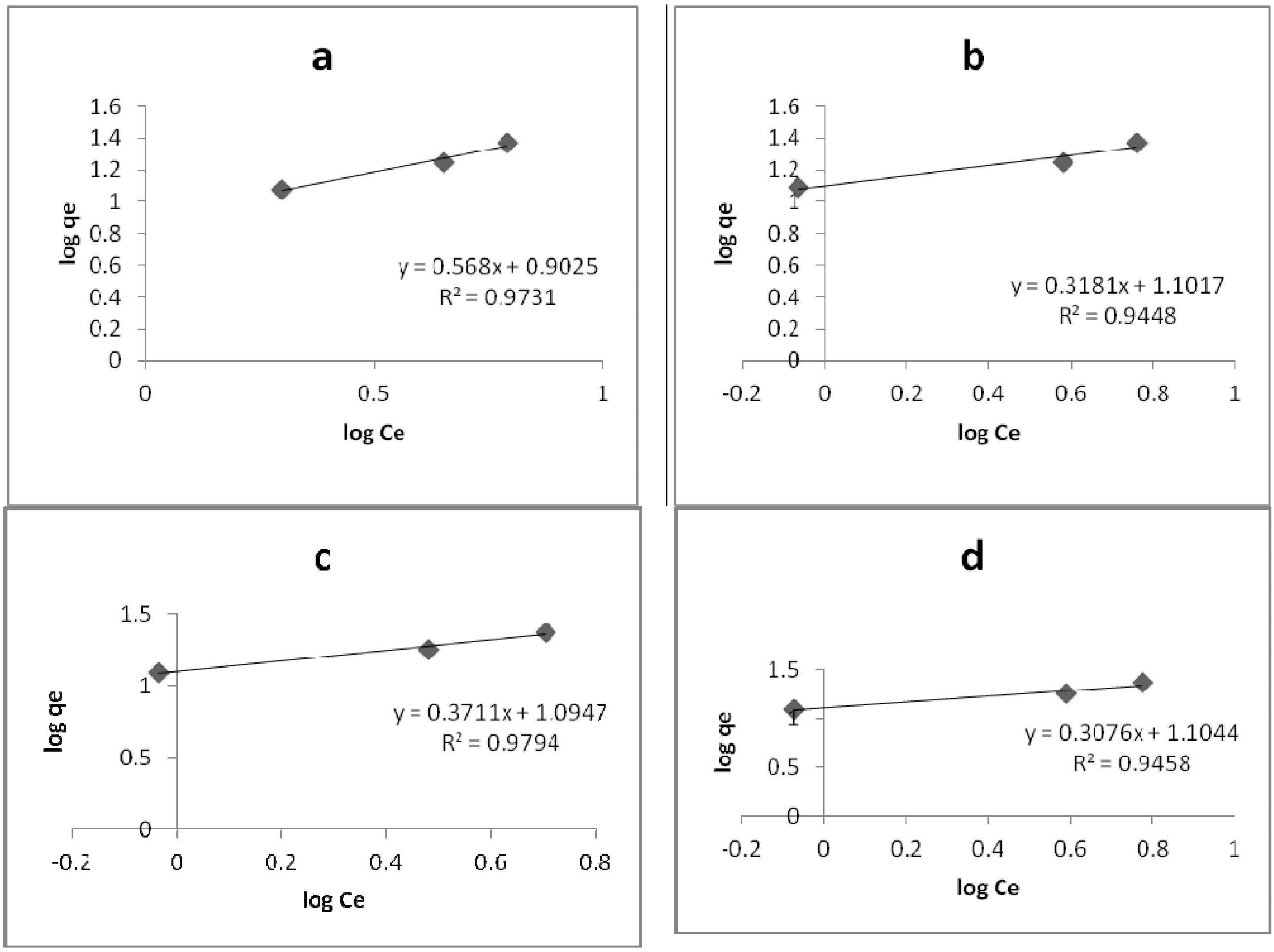

Fig.-5: Freundlich isotherm for the removal of Ni by adsorption on (a) BEAC. (b) AAC. (c) CAC (d) BNAC.

Table-1: Selected adsorbents from natural waste

S.No.	Adsorbent Name	Botanical name	Abbreviation
1	Behda	Terminallia bellerica	BEAC
2	Anjan	Hardwickia binata	AAC
3	Chinch	Tarmirindus indica	CAC
4	Bakam Neem	Melia azedarach	BNAC

Table-2**:** Characteristics of adsorbents

Properties	BEAC	AAC	CAC	BNAC
Density (g/cm^3)	0.2961	0.3559	0.2550	0.3781
pH	6.5	6.5	7.0	6.3
Moisture content (%)	16.00	42.40	25.60	11.73
Ash Content (%)	7.60	7.27	10.40	16.03

Table-3: Effect of initial metal concentration on the adsorption of Ni (II) ions by BEAC, AAC, CAC and BNAC (contact time = 120 min, pH 7.0, temperature = 28^0C and adsorbent dosage = 0.4 g/100 ml, V =100 ml. W= 400 mg)

Ci	Ce				Qe				% age of Copper removed			
	BEAC	AAC	CAC	BNAC	BEAC	AAC	CAC	BNAC	BEAC	AAC	CAC	BNAC
50	1.987	0.864	0.924	0.844	12.00	12.284	12.269	12.289	96.028	98.272	98.153	98.312
75	4.490	3.821	3.029	3.895	17.628	17.795	17.993	17.776	91.019	92.358	93.942	92.210
100	6.161	5.757	5.066	5.966	23.460	23.561	23.734	23.509	87.678	88.485	89.868	88.067

From the Table-4, the Freundlich constant, 1/n denoting the intensity of adsorption indicates a favorable adsorption since 1/n < 1, the adsorptions of Ni (II) ions onto BEAC, AAC, CAC and BNAC adsorbents has been comparable, among them BEAC shows highest value of adsorption capacity.

Table-4 : Langmuir and Freunlich parameters of adsorption isotherms.

Adsorbents	Langmuir isotherm results				Freundlich isotherm results		
	Q_0	b	r^2	R_L	Kf	1/n	r^2
BEAC	41.66	0.192	0.891	0.09433	0.568	0.902	0.973
AAC	27.77	0.7502	0.941	0.02596	0.318	1.101	0.944
CAC	30.3	0.6471	0.965	0.02998	0.371	1.094	0.979
BNAC	27.027	0.7708	0.944	0.02529	0.307	1.104	0.945

CONCLUSION

In this study the heavy metal Nickel was selected for the removal from aqueous solutions using adsorption techniques. Batch experiments were conducted to study the impacts of particle size, pH, agitation time and initial metal ion concentrations. A particle size of 45-180 micron observed to be highly efficient for Nickel (II). A decrease in the size of the particle increased the adsorption, as a decrease in size of the particle increase the availability of the number of active sites. The increase of pH showed a negative impact on the adsorbent of Ni (II). Maximum adsorption was recorded at pH 7.0 for Nickel (II). The adsorption decreased with increase of initial metal ion concentrations. The adsorbents selected for the study proved to be good adsorbent which was evident with the adsorption data obeying the Langmuir and Freundlich isotherms. Taking into consideration of the above results, it can be concluded that the BEAC, AAC, CAC and BNAC were a suitable adsorbents for the removal of Ni (II) ions from aqueous solution in terms of low cost, natural and abundant availability.

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