IOSR Journal of Dental and Medical Sciences (IOSR-JDMS)
e-ISSN: 2279-0853, p-ISSN: 2279-0861.Volume 17, Issue 1 Ver. XII January. (2018), PP 58-68
www.iosrjournals.org

Adsorption of Methylene Blue on Iraqibentonite ,Narang Leaves And Corn Seeds Powder From Aqueous Solution

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Abstract
***Background**: usethebentonite ,narang leaves and corn seeds as adsorbent surface for the removal of one of dyes from wastewater was studied.*
***Objective:** The adsorption of methylene blue from aqueous solution are investigate on three adsorbents*
***Method** : UV- Spectrophotometric technique to obtained the quantities of adsorption data at*
Different of temperatures (37 , 32 ᵒc) and at pH=1.2in an attempt to simulate the acidity of gastric fluid.
Results: The quantities adsorbed of methylene blue on bentonite ,narang leaves and corn seeds were increased at 37 cᵒ.Adsorption characteristics were described using frundlich isotherm.
***Conclusion:** The adsorption process has been foundthe value of ΔG^oshowed the spontaneous nature of adsorption for three adsorbents and endothermic in nature and thermodynamicParameters, Gibb's free energy (ΔG°), change in enthalpy (ΔH°) and change in Entropy (ΔS°) have been calculated.*
***Keyword** : Adsorption,mathylene blue,bentonite, narinij leaves and corn seeds, thermodynamic parameters.*

Date of Submission: 09-01-2018 Date of acceptance: 26-01-2018

I. Introduction

There are more than 1,00,000 types of dyes commercially available of dyes produced annually, which can be classified according to their structure ascationic and anionic (1).Waste streamcontainingin Dyes is one of the major toxic industrial wastes. Various types of dyes are used in the process industries like textile, paints, pulp and paper, carpet and printing etc. The effluents containing dyes are highly colored and cause water pollution(2,3).Methylene Blue, is one of the dyes,it is a cationic dyeorbasic dye.(4). It is used insynthetic pigmentsand is widely used in textile industry and in several processes such as acrylic, nylon, silk and wool dyeing. Inaddition it's a hydrochloride salt are used in the coloring of paper and leather. (5)Therefore thedisposal of dye into receiving water causes pollution to the environment. In textile processing, a significant proportionof synthetic organic dyestuffs are released annually to wastestreams which eventually enter the environment. (6) Eliminationof dye in the textile dying process is very difficult byConventional wastewater treatment methods. These dyes areStable under the environmental effects, such as light, pH andmicrobial attack.(6)The highest rates of toxicity were found amongst basic and diazo direct dyes. Therefore, it is highly necessary to lowering dye concentration in thewastewater. The elimination methods for treating dyecontainingWastewaters are electrochemical treatment, coagulation and flocculation, chemical oxidation, liquid–liquid extraction and adsorption (7). Among these methods, adsorption process is widely used for its simplicity.There are different types of adsorbents such as kaolin, activated charcoal, attapulgite, talc, magnesium trisilicate and bentonite(8)activated carbon is the most popular for the removal of pollutants from wastewater.(9)in the adsorption study the process effects by different parameters including pH, agitation rate and contact time on the adsorption efficiency(10).These adsorbents or Clays have typical properties (large surface area, high cation exchange capacity, chemical and mechanical stability, and a layered structure) that improved them to be good adsorbents.(11)Among these clays is bentoniteTherefore,thebentonite is considered as a good adsorbent material for heavy metal ions such as Pb^{2+}in aqueous solution due to its typically elevated surface area, high availability and low cost **(12).**Bentonite surface consists of one octahedral alumina sheet lying between two tetrahedral layers of silica. bentoniteit carry negative charge is attributed to the isomorphs replacement of Al^{3+} for Si^{4+} in the tetrahedral layer and Mg^{2+} for Al^{3+} in the octahedral layer(11).Langmuirisotherm: for simple solutes, the adsorption behavioris generally uncomplicated, and can be modeled accurately on the basis of the interactions between the adsorbing adsorbate and the surface of the adsorbent and describes the adsorption behavior to a monolayer level of coverage(13).The Langmuir equation could be expressed as: $C_e/Q_e = 1/K + a/k .C_e$WhereQ_e is the amount of adsorbate (mg/g), C_e is the equilibrium concentration (mg/L), and a, k are constants or sometimes called Langmuir constants.Related to adsorption capacity and energy of adsorption(8).

Freundlich Isotherm is one of the most important isotherms that found with the adsorption process at solid-liquid interface. Most of surfaces areheterogeneous, so the change in potential energy is regular, and the adsorption sites are not equivalent in energy hence the multilayer formation is highly expected.
Freundlich equation could be written as follows:-$Q = k_F\ Ce1/n$ ……..….(2)
$\log Q = \log k_F + 1/n \log C_e$ …….(3)
Where Q is the adsorbate quantity (mg/g), C_e is the concentration of adsorbate at equilibrium (mg/L), k_F, and n is the adsorption capacity and an empirical parameter, respectively and also called Freundlich constants **[8]**.

II. Methods

The instruments used were UV - VIS Spectrophotometer (UV-1800) Shimadzu, thermostatic Shaker bath/GFL (D-3006), Germany, pH Meter/HM -73, TDA Electronics Ltd., Centrifuge /eppendorf 5804 R, electronic Balance/Sartorius Lab. BP 3015. The material used is NaOH (Emscope laboratories Ltd). The dye used in this studymethylene blue that is obtained from chemical and biochemical department.The chemical formula and molecular weight for methylene blue are$C_{16}H_{18}N_3SCl \cdot 4H_2O$ and 320 g mol-1.(14)Acid, basic, reactive and direct and soluble dyes, while dispersed pigments and oxidized dyes are insoluble in water.[5]

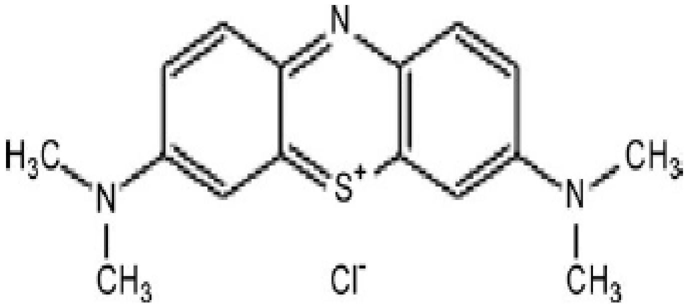

Fig (1): structure of methylene blue. [15]

A stock of (500ml) aqueous solution of methylene blue(0.003 mg L^{-1}) was prepared and its (λmax) was determined. The maximum absorbance (λmax) was (495) nm. As shown at figure (2).

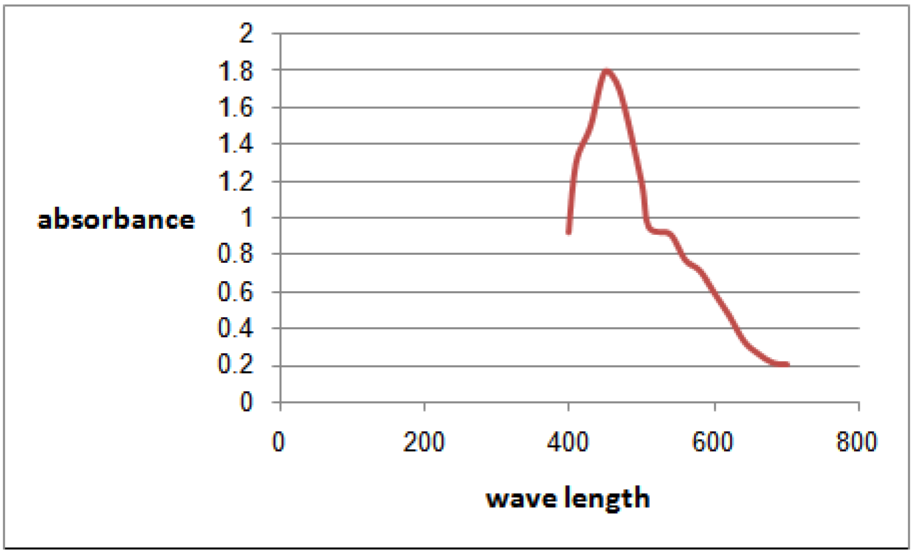

Fig (2): Spectra of aqueous solution of methylene blue at temperature 37°c.

Various drug solutions with different concentrations were prepared by diluting the stock solution with distilled water (0.001, 0.0008, 0.0006, 0.0004, 0.0002 mgL^{-1}).In order to obtain the calibration curve for aqueous solutions of methylene blue at pH =1.2 the absorbance values of these drug solutions were measured at the specific (λmax) using UV-Vis double beam Spectrophotometer and plotted versus the concentrations of these drug solutions (in figure 3).

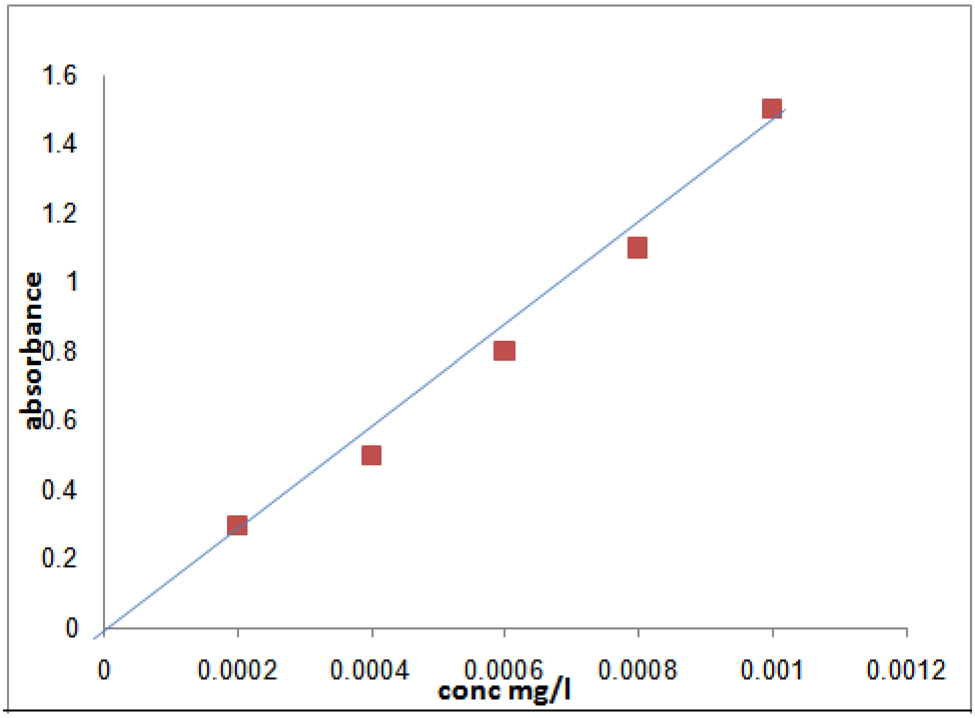

Fig (3) : Calibration curve for aqueous solutions of methylene blue at temperature 37ºc

Adsorbents

BentoniteThe bentonite clay size 75 µm used in this study was supplied by Geological scanning company and has the following composition (bypercentage weight):
SiO_2 (56.77%), Al_2O_3 (15.67%), Fe_2O_3 (5%), CaO (4.48%)
MgO (3.42%), K_2O (0.60%) ,Na_2O (1.11%), L.O.I (12.49%)

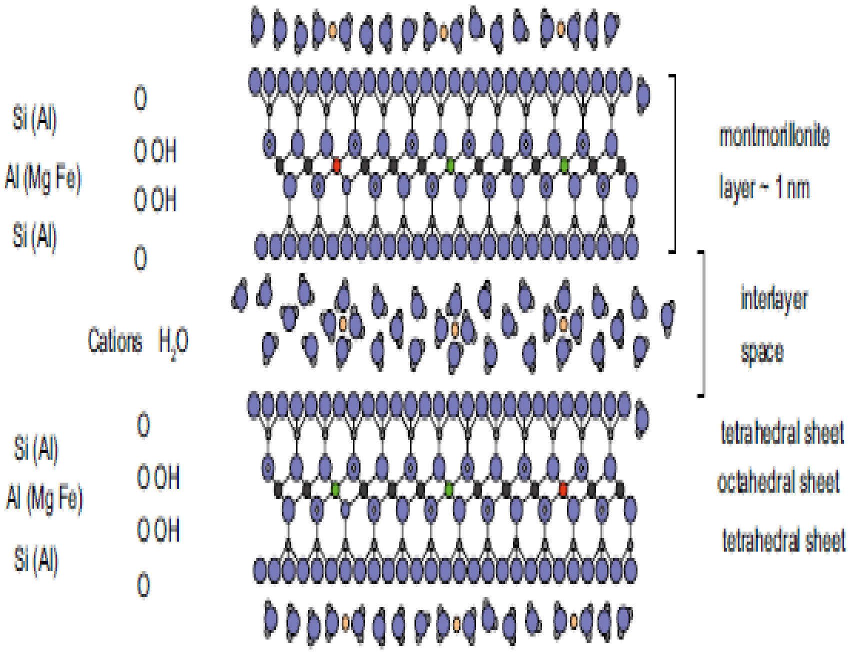

Figure 4. The Structural formula of bentonite(16)

Narang leaves , corn seeds and bentonite adsorbents were in powder form.Also for narang leaves powder and corn seeds powder size 75 µm used in this study, the function groups were supplied by Nano center in university of technology. As shown in figure (5) and (6).

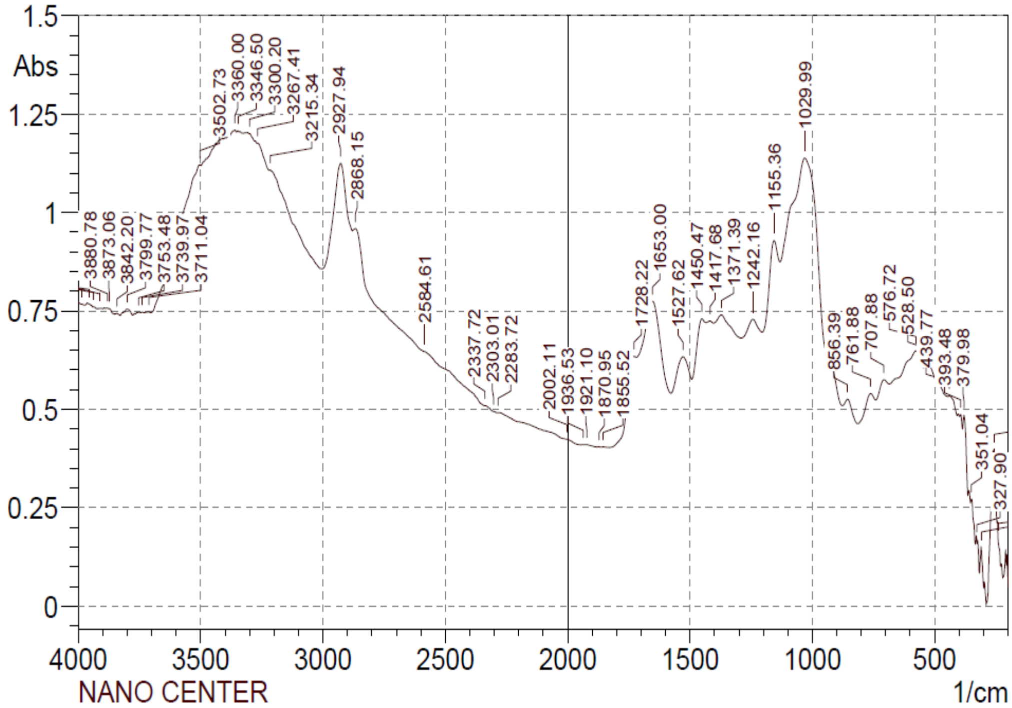

Figure (5):FT-IR spectrumfor corn seeds powder.

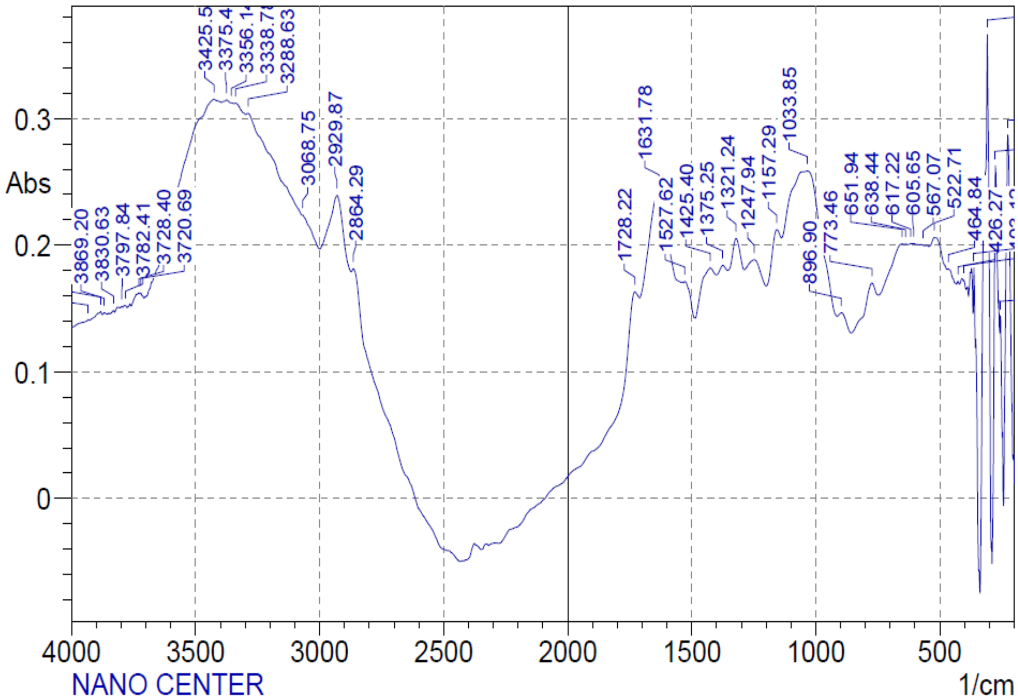

Figure (6) :FT-IR spectrumfor narang leaves powder.

Each of them was washed several times with excessive amounts of distilled water then dried at (150 ^{o}C) in the oven for three hours and kept in airtight containers. Each adsorbent was ground and sieved using Retch test sieve150μm. The time to reach equilibrium state, that is required for full saturation of adsorbent surface at 37 ^{o}C by the adsorbate has been determined by the following procedure: 500 ml initial concentration (0.003 mg/L) of adsorbate solution was shaken with (0.5 g) of each adsorbent. The absorbance of adsorbate solutions were measured by UV/Visible spectrophotometer at different intervals 10, 20, 30, 60 …minutes until reaching equilibrium (no further uptake of adsorbate by adsorbent as the time proceeds).A systematic procedure was followed to determine the adsorption isotherms for each pair of adsorbent -adsorbate systems. A volume of (50ml) of five different concentrations of drug solution (0.001, 0.0008, 0.0006, 0.0004, 0.0002 mg/L) was shaken with (0.5 g) of adsorbent at a certain temperature in a thermostatic shaker. The speed of shaking was 60 cycles per minute. After the equilibrium time (15 min) elapsed, the mixtures were allowed to settle and the clear liquids were either centrifuged at 3500 round per minute (rpm) for 10 minutes. The absorbencies of the filtrate solutions were measured at (λmax). The equilibrium concentrations of the prepared solutions can be determined from the calibration curve using their absorbencies. Adsorbed amount of the drug was calculated at certain conditions from the concentration of solution before and after adsorption according to equation (1):

$X_m = (C_o - C_e)\ V / m$……………..(1), where C_o and C_e are the initial and equilibrium concentrations of drug solution (mg/L) respectively, V is the volume of solution in liter, X_m= the maximum quantity of adsorbate (in mg) that is adsorbed on the adsorbent at certain value of C_e that was fixed for all temperatures used in the study, (m) is the weight of adsorbent in grams.
X_m can be determined from equation (2): .

$Q_e = X_m / m$……………………(2), where Q_e= is the quantity of adsorbate (in mg) held by (0.5 g) of adsorbent.
The equilibrium constant (k) for the adsorption process at each temperature is calculated from equation (3):
$K = (Q_e)\ (0.5\ g)/(C_e)\ (0.05\ L)$……….(4), where (0.5 g) represents the weight of the clay that has been used, (0.05 liter) represents the volume of the drug solution used in the adsorption process.
The change in free energy (ΔG) could be determined from equation (4):
$\Delta G = -\ R\ T\ \ln k$…………….(4), where R is the gas constant (8.314 J/mol. deg) and T is the absolute temperature.
The heat of adsorption (ΔH) may be obtained from equation (5):
In Xm = -ΔH/RT + constant ……….(5)
The change in entropy (ΔS) can be determined from equation (6):
$\Delta G = \Delta H - T\Delta S$………(6)

III. Results

Temperature effects and thermodynamic parameters:-

The general shapes of the adsorption of methylene blue on bentonite, narang leaves and corn seeds at different temperatures (37 and 32°c) are given in figures (7, 8 and 9). Figures show that the adsorption of methylene blue increases at 37 °c temperature .

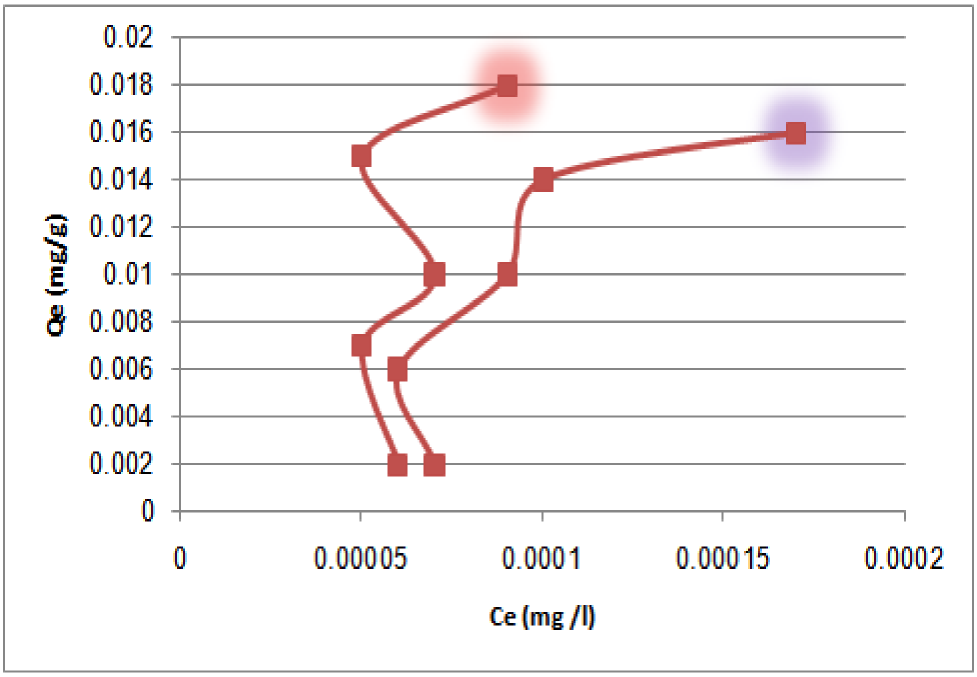

Fig (7): adsorption isotherm of methylene blue on bentonite at different temperatures (37 and 32°c) and at pH=1.2.

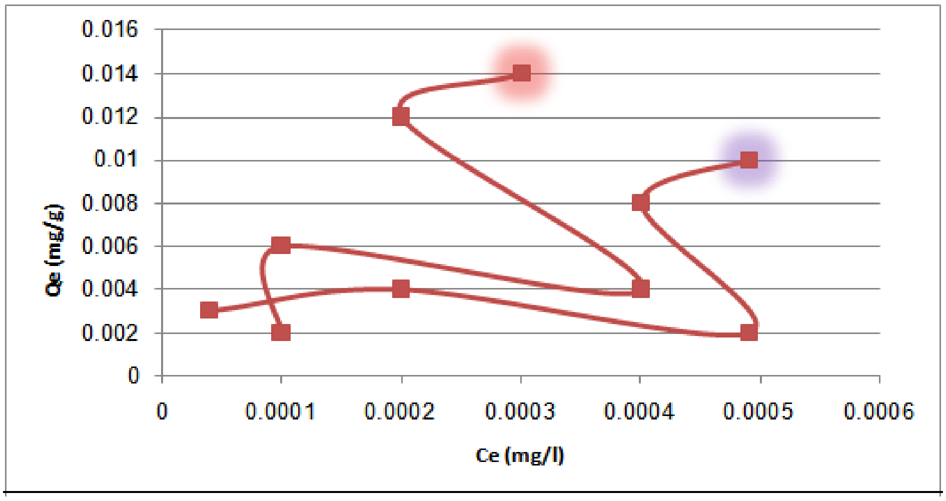

Fig (8): adsorption isotherm of methylene blue on narangleavse at different temperatures (37 and 32ºc) and at pH=1.2.

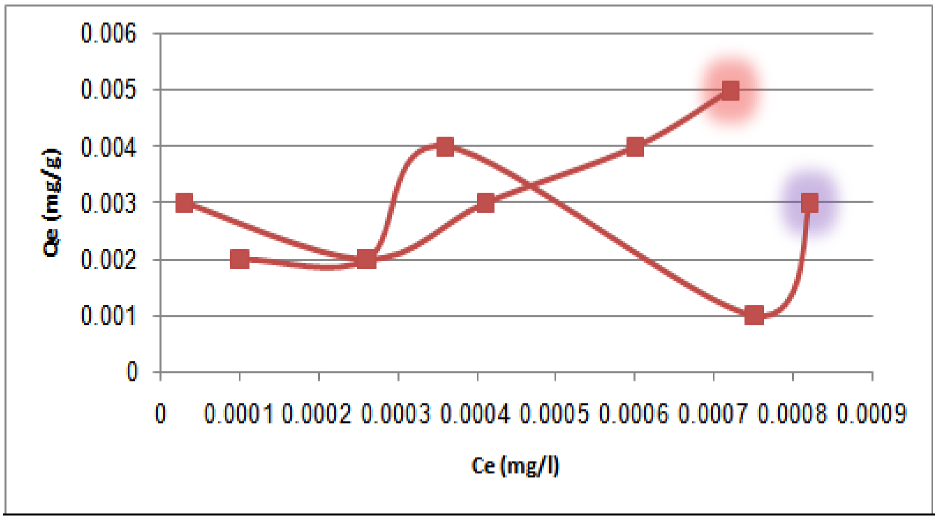

Fig (9): adsorption isotherm of methylene blue on corn seeds at different temperatures (37 and 32ºc) and at pH=1.2.

The study of the temperature effects on adsorption helps in finding the basic thermodynamic functions (ΔH ,ΔG , ΔS) of the adsorption processes. Show as that in table (1) gives X_m values at different temperatures and at pH=1.2.Table (1): effect of temperature on the maximum adsorbed quantities of mathylene blue onbentonite ,narang leaves and corn seeds.

Where X_m is the maximum uptake of adsorbate at certain value of (C_e) for all temperatures.

Adsorbent	T.c°	T.k°	1000/T. °k^{-1}	X_m(mg)	In X_m
Bentonite	37 32	310 305	3.22 3.27	0.009 0.008	-4.71 -4.82
Narang leaves	37 32	310 305	3.22 3.27	0.007 0.005	-4.96 -5.29
Corn seeds	37 32	310 305	3.22 3.27	0.0025 0.0015	-5.99 -6.5

Plotting (In X_m) versus 1000/T produced a straight line with a slope = - ΔH/R as shown in figures (10,11 and 12).

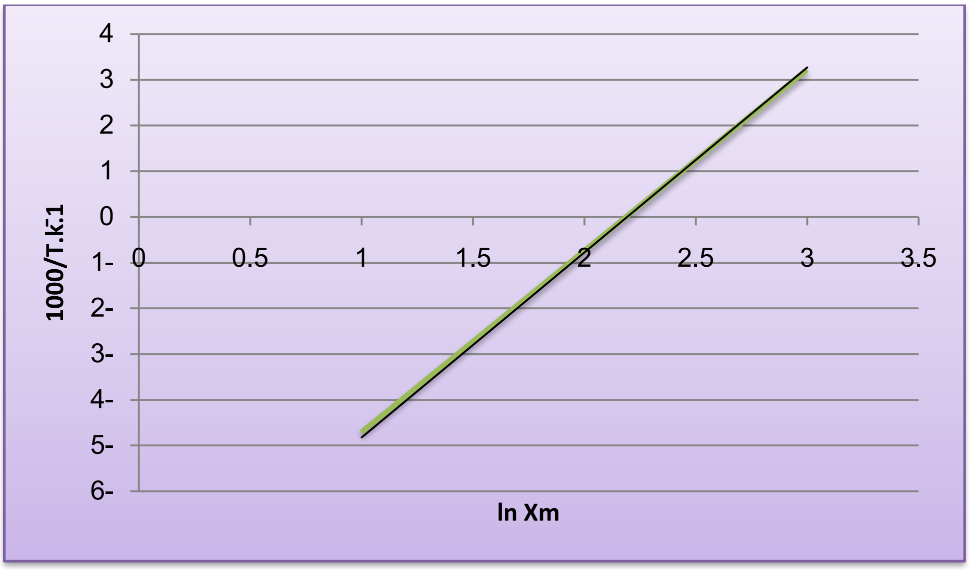

Fig10. In Xm plotted against reciprocal absolute temperature for the adsorption of methylene blue on bentonite at pH=1.2.

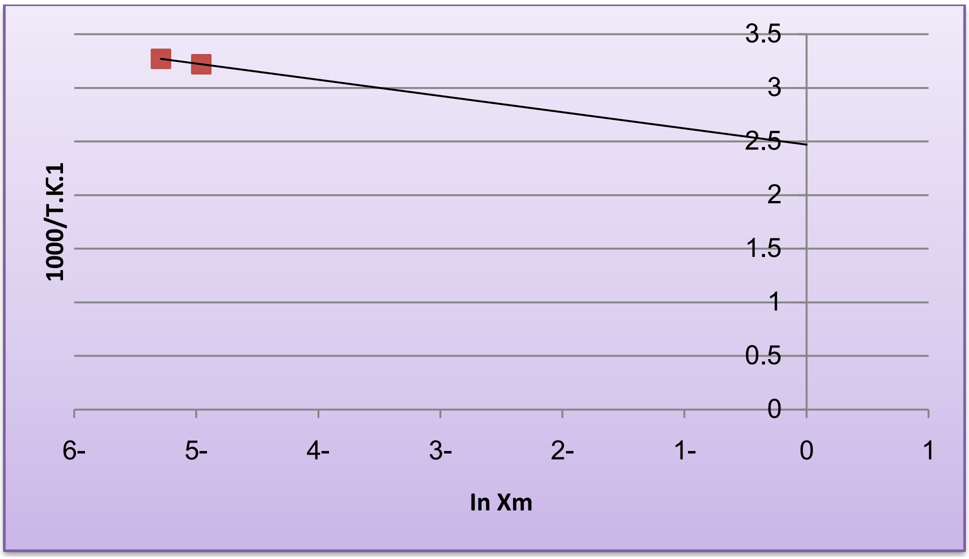

Fig11. In Xm plotted against reciprocal absolute temperature for the adsorption of methylene blue onnarang leaves at pH=1.2.

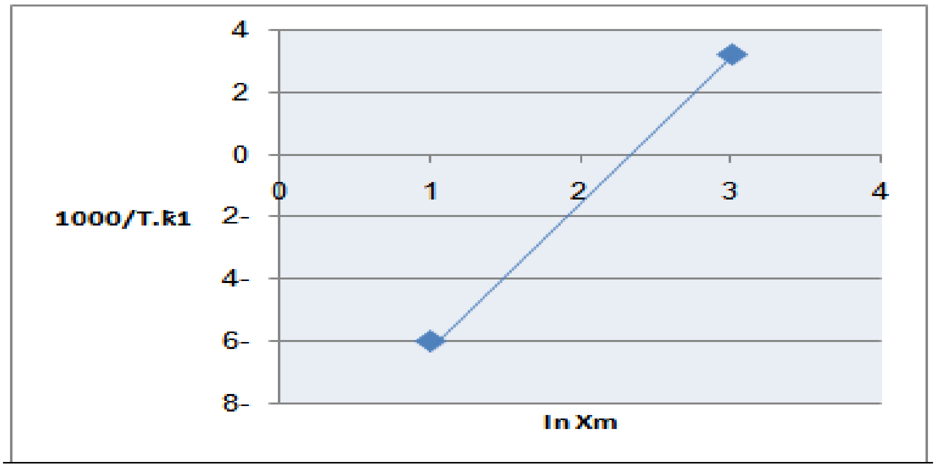

Fig12. In Xm plotted against reciprocal absolute temperature for the adsorption of methylene blue on corn seeds at pH=1.2.

Table (2) shows the basic thermodynamically values of adsorption of methylene blue onbentonite ,narang leaves and corn seeds.

Adsorbent	ΔH	ΔG	ΔS	pH	Temprature
bentonite	+12.14 +12.22	-5.9 -3.8	+0.05 +0.05	1.2 1.2	37 32
Narang leaves	+12.78 +13.41	-2 -1.5	+0.04 +0.04	1.2 1.2	37 32
Corn seeds	+15.91 +16.72	-15.7 -13.6	+0.1 +0.09	1.2 1.2	37 32

The negative values of the ΔG for the adsorption of methylene blue on bentonite ,narang leaves and corn seeds indicated that the adsorption process of folic acid is spontaneous.The positive values of ΔH at different temperatures indicated an endothermic reation.The positive values of ΔS indicated the increase in the degree of freedom of the adsorbed species (methylene blue).Adsorption isotherms The adsorption isotherms of methylene blue on bentonite,narang leaves and corn seeds at pH=1.2 and different temperatures are shown in figures (7,8 and 9) , as indicate offrundlich isotherm depend on Giles classification.Figures (13,14 and 15) show a linear relationship between (log Q_e) and (log C_e)

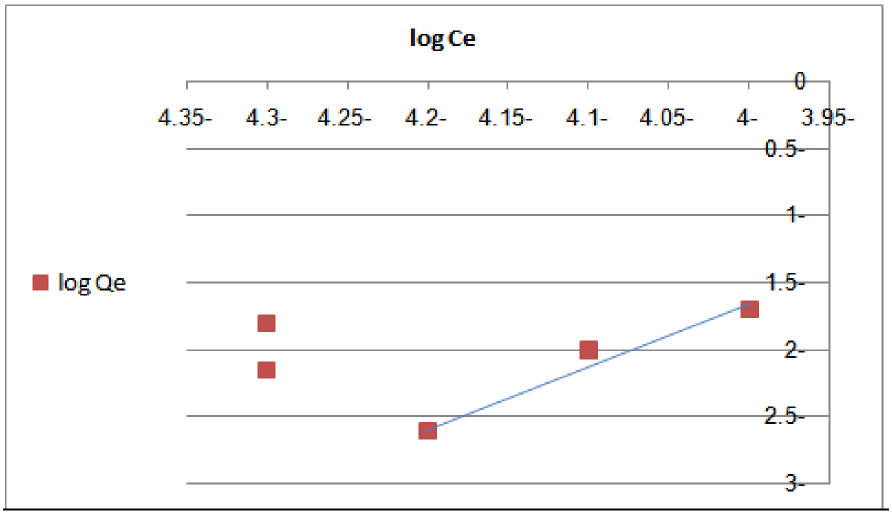

Figure 13. Linear form of freundlich isotherm of mathylene blue adsorbed on bentonit at 37 C^{o} and pH=1.2.

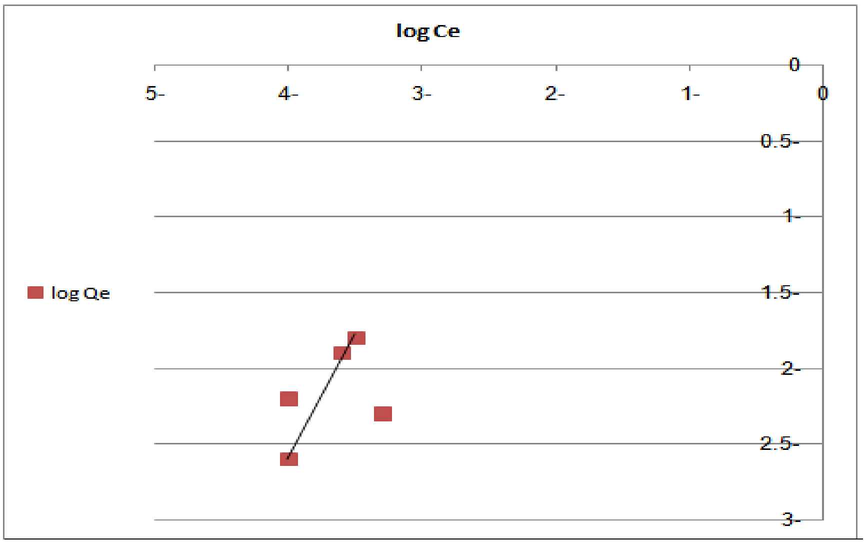

Figure 14. Linear form of freundlich isotherm of mathylene blue adsorbed on narang leaves at 37 C^{o} and pH=1.2.

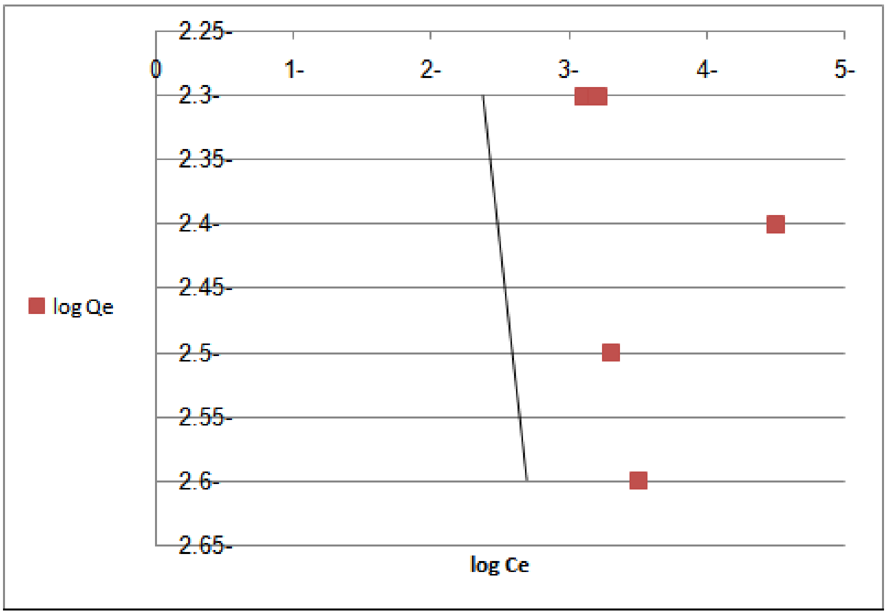

Figure 15. Linear form of freundlich isotherm of mathylene blue adsorbed on corn seeds at 37 C^{o} and pH=1.2.

IV. Discussion

Many treatment methods have been used to remove dyes from wastewater, which can be divided into chemical, biological andphysical methods. Physical methods which include adsorption.[1]The Adsorption is one of the methods commonly used to remove any substance from various aqueous solutions with relatively low concentrations. The efficiency of adsorption relies on the capability of the adsorbent to adsorb molecules from the solutions onto its surfaces.[17] in addition , a number of intermolecular forces have been suggested to explain this aggregation, these forces include (van der Waalsforces,ion dipole forces and dipole dipole forces), which occur between dye molecules in the solution.[1].The most important in the investigation of adsorption mechanism is the equilibrium adsorption isotherm. These properties such as surface property and the maximum adsorption capacity can be determined from the adsorption isotherm and correlative constants.[18]The process of dye removal by adsorption is being from of use the lower cost adsorbents so that the process becomes economically feasible. Therefore, theresearchers are focused on to use of low-cost, reusable, locally available, biodegradable adsorbents made from natural sources like wheat bran, rice husk, waste apricot, bagasse fly ash, powdered peanut hull etc. Natural and modified clays like zeolite, perlite and bentonite are being considered as low-cost adsorbents.[19]Three adsorbents used in this work for adsorption methylene blue dye Bentonite , narang leaves and corn seeds powder were chose.From this work the adsorption isotherms isFreundlich Isothermis S type depend on Giles classification it used to describe the adsorption characteristics for the heterogeneous surface and formation multilayers. [20,21], this is indicate for bentonite , narang leaves and corn seeds surfaces is heterogeneous.[21]rom structure of methylene blue iscationic in nature[22] as shows in figure (1) it is carry a net positive charge inthe presence of protonated amine or sulfur containing groups[1]. It can be adsorbed on different site of each adsorbents due to the polar ends of the dyemolecule have a tendency to attachthe heterogeneous surfaces of adsorbentand can separates out from aqueoussolution[22].Therefore,in this work the adsorption processes between the methylene blue and negative site of hydroxyl group in the bentonite site surface [23]. Is attributed to the isomorphous replacement of Al^{3+} for Si^{4+} in the tetrahedral layer and Mg^{2+} for Al^{3+} in the octahedral layer. This negative charge is balanced by the presence of replaceable cations (Ca^{2+}, Na^{+}, etc.) in thestructure [24].In addition the functional groups ofheterogeneous surfaces analysis for narang leaves surface and corn seeds surface were detected by used the FT-IRspectrum it found at 3400 cm^{-1} isO- Hband of these surface compounds, a 2922 cm^{-1} vibration of CH_2, 2852 cm^{-1} vibration of CH_2, 1797 cm^{-1} C- O of carboxylic acids band ,1637 cm^{-1} vibration of COO, C- O and C- N bands , peptide bond of proteins, a 1426 cm^{-1} was of phenolic O-H and C-O stretching bond,a 1033 cm^{-1} band was vibration Of aliphatic C- O bands. The <1000 cm^{-1}indicated finger print zone which were phosphate and sulphuregroups. [25]Andthere are two bands at 1022 cm^{-1} and 465 cm^{-1} could be attributed to the stretching and bending vibrations of the SiO_2 in the structure ofnarang leaves surface[4]he functional groups of corn seeds surface are two bands at 1022cm^{-1} and 465 cm^{-1} could be attributed to the stretching and bending vibrations of the SiO_2 in the structure. Also there is at 2900 cm^{-1} could be attributed to C-H band stretching for aliphatic chain andat 1720 cm^{-1} there is a band related to the C=O of the carboxylic group[4].

In this studied used pH (1.2) at low pH showing that adsorbents have positively charged surface. [17]Which enhance adsorbing basic dyes [24].e. the adsorbent have a zero point charge at pH lower, the surface of the adsorbent becomes positive chargedwhich enhances the adsorption of negatively charged from functional groups of the adsorbate as shown in figure (1)through the electrostatic forces of attraction on the surface of adsorbent. [26]n this study found the temperature effect on the adsorption process increasingwith increasing temperature. It might be that stronger bonds areformed at higher temperatures and in this fact that the adsorption is endothermic[27]Thermodynamic parameters such as free energy change (ΔG°),enthalpy change (ΔH°) and entropy change (ΔS°) used toknow the nature of adsorption process. [19 ,29] n this work the negative values of_ΔG◦ for all temperatures indicate the spontaneous nature of adsorption with a high preference of Methylene Blue onto (bentonite,narang leaves and corn seeds) [27]. The value of_ΔH◦was positive, indicated that the adsorption reaction was endothermic.The positive value of_ΔS◦shows that increasing randomness atThe solid/liquidinterface during the adsorption of Methylene BlueOn adsorbents.[22,27 ,29]

V. Concluded

It conclude the bentonite surfaceshowed ability to adsorb the methylene blue dye from its aqueous solution it higher than narang leaves surface and its higher than the corn seeds surface at pH=1.2 and different temperatures , therefore, these adsorbents can be used as antidotes for decreasing of methylene blue dye concentration .The adsorption isotherms of methylene blue onbentonite ,narang leaves and corn seedsproposal a Freundlich isotherm model. These results indicated the surface heterogeneity of the adsorbents leading to differentadsorption affinities towards dye molecules and adsorption strengths from site to site. The FT-IR spectrum resultsshowed the adsorbents have characteristicBandsof proteins,lipids and carboxylic acid groups which are able to react or adsorbed with functional groups ofdye molecules from aqueous solution.Thermodynamic parameters such as enthalpy change (ΔH°), free energy change (ΔG°) and entropy change (ΔS°) showed that the adsorption process of methylene blue dye on each adsorbents was spontaneous andexothermic.

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