

**REMOVAL OF INDIGO CARMINE DYE (IC) BY BATCH ADSORPTION METHOD
ONTO DRIED COLA NUT SHELLS AND ITS ACTIVE CARBON FROM AQUEOUS
MEDIUM****Ankoro Naphtali Odogu*, Kouotou Daouda, Belibi Belibi Placide Desiré,
Ndi Julius Nsami, Ketcha Joseph Mbadcam**Physical Chemistry Laboratory, Department of Inorganic Chemistry, Faculty of Science, University of
Yaoundé I, B.P 812, Yaoundé – Cameroon**DOI:****ABSTRACT**

Natural cola nut shells and its active carbons were used to remove indigo carmine dye from aqueous solution using batch adsorption test. The effect of pH, contact time (t), adsorbent dose (m) and initial concentrations (Co) were investigated. The equilibrium adsorption data were analyzed by Langmuir, Freundlich, Tempkin and Dubin-Kaganer-Raduskushkevich classical isotherm models. This equilibrium data best fits with all the four isotherm models for cola nut shells. Langmuir and Freundlich equations correlated well with data obtained using activated carbon based H₃PO₄ while Freundlich and Tempkin best agreed with activated carbon based KOH. The kinetics of indigo carmine dye was discussed by pseudo-first order, pseudo-second order, Elovich and Intra-particle diffusion models. The pseudo-second order kinetic model equation fitted best to the data from all the three adsorbents. Elovich fitted best with data obtained using activated carbon based H₃PO₄ while intra-particle diffusion model for activated carbon based KOH. D-K-R shows that adsorption process was chemisorption for all the three adsorbents. Natural cola nut shells and activated carbon based KOH show higher adsorption capacities with indigo carmine compared to activated carbon based H₃PO₄. The results indicated that these shells and its active carbon can be used as an effective and low-cost adsorbent to remove indigo carmine from aqueous solutions.

KEYWORDS: Adsorption, Indigo Carmine, Cola Nut Shells, Active carbon, Kinetic studies.**INTRODUCTION**

Textile industries in the world and the increasing demand for textile, and also the ceramic paper, printing and plastic uses large varieties of dyes as their raw materials [1]. Some dyes are used in medicine, biological stain and coloring plastics [2]. The waste from these industries in one way or the other finally ends up in water, thereby polluting the environment. Unlike other pollutants, dye pollutants especially those with the benzene ring even at low concentration is visible which reduces light penetration in to water, hence causes negative effect on photosynthesis to water plants. They are non-degradable [3]. Also anthropogenic activities have caused a great harm to the quality of our life line, which is water [4]. Indigo carmine dye is used as coloring agents in companies like CICAM in Cameroon, as additive in pharmaceuticals tablets and capsules and also for medical diagnostic purposes. Indigo carmine, however, is not readily metabolized but is rather freely filterable by the kidneys. Hence intravenous injection of IC dye for intra-operative cystoscopy is a safe technique to detect intra-operative compromises of the urinary tract [5]. The indigo carmine dye is considered a highly toxic indigoid class of dye and its touch can cause skin and eye irritations to human being. It can also cause permanent injury to the cornea and conjunctiva [5]. The consumption of the dye has also proved fatal, as it is carcinogenic in nature and can lead to reproductive, developmental, neuro and acute toxicity [5]. It has also been established that the dye leads to tumors at the site of application. It has also been reported to cause mild to severe hypertension, cardiovascular and respiration effects [5]. Waste water containing IC dye is rich in both color and organic contents and large amount of suspends solid which are broadly fluctuating in pH, high temperature and beside high chemical oxygen demand [6]. From the norms of the World Health organization, indigo carmine dye at concentration greater than 0.005 mg/L [7] is not acceptable in water. This IC dye is not only a nuisance to aquatic life but also to us the humans. Hence treatment of this waste

before deposition is essential. There are many method of treatment procedures among which are Chemical precipitation, Ultra filtration, electrochemical deposition, Coagulation-Flocculation and Adsorption. These methods differ in their efficiency to remove IC dye in water and all except adsorption are costly [8]. These techniques have certain disadvantages, which include high capital and operational cost and needs treatment and appropriate disposal of the residual dye sludge [8]. Thus, adsorption remains the out most process because of its simplicity, high efficiency and low cost. Many adsorbents have been used such as chitosan beads, palm kernel, kaolinite, clay, raw bagasse, alumina metal oxide and lots of others. The main aim of this work is was to access the removal of indigo carmine dye from aqueous solution using cola nut shell and its active carbon.

EXPERIMENTAL PROCEDURE

Preparation of Adsorbents

The preparation procedure for cola nut shells (CNS) and its active carbons (AC) used in this work has been reported by Ndi *et al.*, in 2014 [9]. 10 g each of CNS and AC based H_3PO_4 and AC based KOH were dried in an oven for 24 h at $120^\circ C$. It was then removed kept in a desiccator for adsorption. The cola nut shells used were of sizes less than $1.25 \mu m$.

Preparation of Adsorbate

1000 ppm solution of indigo carmine was prepared by dissolving 1.25 g of the indigo carmine powder in 1 L volumetric flask with distilled water. The solution was put under agitation for 2 hours. From the mother solution, standard solutions of 5, 10, 20, 25, 30, 40, 50, 60, and 70 mg/L were prepared. Solutions of hydrochloric acid (0.1 M) and sodium hydroxide (0.1M) were also prepared for pH adjustment.

Batch Adsorption Experiments

Batch adsorption tests were carried out by mechanical agitation at room temperature. For each run, 20 mL of indigo carmine dye of known initial concentration was treated with a known weight of adsorbents (cola nut shells and its active carbon). After agitation, the solution was filtered and the filtrate were subsequently analyzed for dye concentration by UV/V spectrophotometer, model ANLAGE 260. Similar measurements were also carried out at various adsorbents doses, i.e. pH and initial concentrations of indigo carmine dye.

The percentage removal (%R) of indigo carmine dye and the amount (Q_e) adsorbed per unit mass of adsorbent were calculated by using the following expressions;

$$Q_e = \frac{(C_o - C_e) \times V}{m} \dots\dots\dots (1)$$

$$\%P = \frac{(C_o - C_t)}{C_o} \times 100 \dots\dots\dots (2)$$

where C_o is the initial concentration, C_e is equilibrium concentration, V represents the volume of the solution and m is the mass of the adsorbent.

ADSORPTION ISOTHERMS

Adsorption is usually described through isotherms, which is, the amount of adsorbate on the adsorbent as a function of its pressure (if gas) or concentration (if liquid) at constant temperature.

3.1. Langmuir model: The Langmuir adsorption isotherm is often used for adsorption of a solute from a liquid solution. It is perhaps the best known of all isotherms describing adsorption and is often expressed as [10]:

$$Q_e = Q_m \frac{K_1 C_e}{1 + K_1 C_e} \dots\dots\dots (3)$$

where: Q_e is the mass of adsorbate per g of adsorbent, C_e is the equilibrium concentration, Q_m : the maximum adsorption capacity, K_1 the Langmuir constant. This equation can further be linearized to give:

$$\frac{C_e}{Q_e} = \frac{1}{K_1 Q_m} + \frac{1}{Q_m} C_e \dots\dots\dots (4)$$

3.2. Freundlich Isotherm

The Freundlich isotherm model applies to adsorption on heterogeneous surfaces with interaction between the adsorbed molecules, and there is not restriction to the formation of a multi-layer [11, 12]. The well-known expression for the Freundlich model is given below:

$$Q_e = K_f C_e^{1/n} \dots\dots\dots (5)$$

which can be transformed to it linear form as:

$$\ln Q_e = \ln K_f + \ln C_e \dots\dots\dots (6)$$

where: Q_e is the amount adsorbed at equilibrium (mg/g), K_f is the Freundlich constant, $1/n$ is the heterogeneity factor which is related to the capacity and intensity of the adsorption, C_e is the equilibrium concentration (mgL^{-1}). The values of K and $1/n$ can be obtained from the slope and intercept of the plot of $\ln Q_e$ against $\ln C_e$.

3.3. Dubinin –Kaganer-Radushkevich isotherm model

Dubinin–Kaganer-Radushkevich isotherm is generally applied to express the adsorption mechanism with a Gaussian energy distribution onto a heterogeneous surface [13]. The model has often successfully fitted well high solute activities and the intermediate range of concentrations data.

$$\ln Q_e = \ln Q_m - \beta \varepsilon^2 \dots\dots\dots (7)$$

where: $\varepsilon = RT \ln(1 + 1/C_e)$, Q_e = amount of adsorbate in the adsorbent at equilibrium (mg/g), Q_m = theoretical isotherm saturation capacity (mg/g), β = Dubinin–Kaganer-Radushkevich isotherm constant (mol /kJ), ε = Dubinin–Kaganer-Radushkevich isotherm constant. The approach was usually applied to distinguish the physical and chemical adsorption of metal ions with its mean free energy, E per molecule of adsorbate (for removing a molecule from its location in the sorption space to the infinity) can be computed by the relationship [13].

Adsorption kinetics

The adsorption onto or into adsorbent surfaces with respect to time is known as adsorption kinetics. Adsorption is a time- dependent process, and to determine the rate of adsorption is highly important in the design and evaluation of adsorbent in removing pollutants from wastewater [14].

Pseudo-First order kinetic Model

The pseudo-first order kinetic model is the earliest known in describing adsorption rate based on the adsorption capacity [14]. The pseudo-first order equation of Lagergren is generally express as follows [15, 16].

$$\frac{dQ_t}{dt} = K_1 (Q_e - Q_t) \dots\dots\dots (8)$$

where Q_e and Q_t are the sorption capacity at equilibrium and at time t and K_1 is the rate constant of pseudo-first order sorption ($\text{L} \cdot \text{min}^{-1}$). Integrating equation (8) at $t = 0$ and at $t = t$; $Q_t = 0$ and $Q_t = Q_t$ gives:

$$\ln \frac{(Q_e - Q_t)}{Q_e} = -K_1 t \dots\dots\dots (9)$$

Pseudo-second order kinetic model

The Pseudo-second order kinetic model considers the rate-limiting step as the formation of a chemisorptive bond involving the sharing or the exchange of electron between the sorbate and the sorbent [17, 18]. The Pseudo-second order rate equation is express as:

$$\frac{dQ_t}{dt} = K_2 (Q_e - Q_t)^2 \dots\dots\dots (10)$$

where K_2 is the pseudo-second order rate coefficient ($\text{g} \cdot \text{mg}^{-1} \cdot \text{min}^{-1}$). Integrating equation (10) at $t = 0$ and at $t = t$; $Q_t = 0$ and $Q_t = Q_t$ gives:

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{t}{Q_e} \dots\dots\dots (11)$$

Intra-particle diffusion model

The intra-particle model is based on the theory proposed by Weber and Morris and is used to identify the diffusion mechanism of sorbent and the sorbate [14]. This takes place in stages: the first stage is the instantaneous adsorption or external surface adsorption of the adsorbate on the surface of the adsorbent. The second stage is the gradual adsorption stage where intra-particle diffusion into the inner layer takes place and is the rate limiting stage. According to this theory;

$$Q_t = K_{id} t^{0.5} + C \dots\dots\dots (12)$$

Where, K_{id} is the intra-particle diffusion constant, C is a constant which gives an idea about the thickness of the boundary.

Elovich model

This model is used to describe kinetic adsorption on heterogeneous surfaces [10]. It is generally represented as:

$$\frac{dQ}{dt} = \alpha e^{-\beta Q_t} \dots\dots\dots (13)$$

Linearizing equation (13) using the $\alpha\beta \gg t$, $Q_t = 0$ to $t = 0$, $Q_t = Q_t$ at $t = t$, gives:

$$Q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln t \dots\dots\dots (14)$$

where: α is the speed of adsorption ($\text{mg.g}^{-1}.\text{min}^{-1}$), β is the desorption constant (g.mg^{-1}).

Mass transfer model

The mass transfer equation is generally given by the formula [19];

$$C_o - C_t = D e^{K_o t} \dots\dots\dots (15)$$

This can be further transformed to:

$$\ln(C_o - C_t) = \ln D + K_o t \dots\dots\dots (16)$$

where: C_o is the initial concentration of adsorbate, C_t is the concentration of adsorbate at time t, D the adjustment parameter, K_o the mass transfer coefficient.

RESULTS AND DISCUSSION

Effects of pH

pH is an important parameter in the study of adsorption because it affects both the surface functional group and the adsorbate. The pH was varied between 2 and 11. Figure 2 shows that the maximum adsorption of IC dye for the three adsorbents was at pH of 2. This might be due to protonation of the surface of the adsorbents thereby increasing the electrostatic force of attraction between the positively charged adsorbents and the negatively charged anionic dye. It can also be allocated to the fact that at pH 2, there is protonation of the adsorbate, facilitating intra-particle diffusion and hence favors adsorption of IC. But for pH greater than 2, the number of positively charged surface sites decreases, which did not favors adsorption due to electrostatic repulsion between negatively charged IC dye and the adsorbents. The slight increase observe at pH greater than 5 is due to change in the structure of the dye molecule from the molecular to the ionic form.

Effect of contact time

The agitation time were varied between 5 and 60 minutes and the results displayed in figure 3. The results for the three adsorbents indicate that, adsorption rate was rapid within the first five minute for the three adsorbents, which slows down and increases again and finally attain equilibrium. This rapid speed might be due to the availability of active sites for surface adsorption. It then slows down due to the saturation of adsorption sites. The second increase might be due to pore (intra-particle) diffusion from the bulk fluid onto the inner surface of the porous adsorbents through the film due to continuous agitation. Similar results were obtained by other authors [5]. This result gives a contact time of 45 minutes for CNS and 35 minutes for AC based on H_3PO_4 and AC based on KOH.

Influence of Adsorbent dose

The study of the effect of adsorbent dosage for the removal of IC dye was done by varying the mass of the adsorbents, from 0.08 to 0.16 g and quantities adsorbed for each adsorbent respectively are represented in Figure 4. It shows that increasing the mass of the adsorbents, decreases the quantity of IC adsorbed per gram for the three

adsorbents. The decrease in adsorption capacity is basically due to masking of adsorption sites, that is, the available surface area for IC adsorption decreases due to aggregation of active sites for adsorption. Similar result was obtained by other researches [20]. The maximum quantity adsorb, Q_e of 9.997, 5.861 and 12.093 mg/g for CNS, AC based H_3PO_4 and AC based KOH respectively was attained for adsorbent mass of 0.08 g for IC adsorption.

Influence of initial dye concentration

The concentration of IC dye was varied between 20 to 60 ppm at a pH of 2, adsorbent dose of 0.08 g and contact time of 45 min for CNS and 35 min for AC based H_3PO_4 and AC based KOH. The analytical result obtained was plotted as shown in figure 5. It is observed that as the initial dye concentration increase, the quantity adsorbed increases for the three adsorbents. This quantity increases from 4.087 to 8.169; 3.366 to 4.942 and 4.378 to 10.227 mg/g for the CNS and AC based H_3PO_4 and AC based KOH respectively. This increase is attributed to the fact that as the concentration increase, the number of collision between the dye molecules and the adsorbents increases as a result of increase in the mobility of the molecules due to high collision between the molecules themselves.

ADSORPTION ISOTHERMS STUDIES

The result from Figures 6, 7, 8 and 9 and the linear constance in Table 1 shows that the linear correlation coefficients of Langmuir and Tempkin are high in the case of CNS with a value of $R^2 = 0.9895$ and $R^2 = 0.908$. This means that the adsorption of IC dye with CNS is best described by these models; hence, it implies that the energy of attraction is high on the monolayer coverage. It can also be seen that the linear correlation constants of Langmuir and Freundlich are higher in the case of AC based H_3PO_4 with values of $R^2 = 0.9439$ and $R^2 = 0.6686$ respectively and hence describe the adsorption of IC dye with AC based H_3PO_4 on a multilayer coverage. With AC based KOH, the Tempkin and Freundlich model describes better which indicates that the maximum particles adsorbed is found on a multilayer coverage. The Values of $1/n$, respectively 0.3245, 0.1796 and 0.3812 for CNS, AC based H_3PO_4 and AC based KOH turns towards zero indicating that bond energies increases with surface density and also shows normal adsorption with high adsorption intensity. The linear correlation constants indicate heterogeneous surface binding. These results were in accordance with others [5]. From the table, it results that the theoretical maximum quantities adsorbed for the three adsorbents (Q_m) is closer to the experimental values for Langmuir and D-K-R. This equally confirms the agreement with Langmuir's monolayer adsorption. That is, the maximum particle adsorbed is found on the monolayer coverage. The Langmuir's separation constant (figure 18) decreases as concentration increases for the three adsorbents and is found to be between 0 and 1 ($0 < R_L < 1$) which confirms favorable adsorption of IC by all three adsorbents. The high Tempkin adsorption energy indicates that the attraction between the adsorbate and the adsorbents is very strong which implies a chemisorption reaction with the formation of chemical bonds. The high D-K-R energy ($E > 40$ KJ/mol) for the three adsorbents indicates that adsorption was strongly chemisorption with the possibility of multi-layer formation.

ADSORPTION KINETIC STUDY

The adsorption kinetic study describes the speed at which the solute phase is retained on the surface of the adsorbents, the time necessary for the process to attained equilibrium and also predicts the limiting step of the adsorption process. The kinetic models of pseudo-first order, pseudo-second order, intra-particle diffusion, Elovich and mass transfer models were studied using the linear transformation equation and the respective results are displayed in Figures 10, 11, 12 and 13 with their respective constant in Table II.

Looking at these results, it can be drawn that the removal of IC dye from aqueous solution by CNS, AC based H_3PO_4 and AC based KOH is adequately described by the pseudo-second order with a correlation constant of 0.956, 0.9794 and 0.9931 respectively. This model strongly described interaction between the surface of these adsorbents and the adsorbate, which indicates that the adsorption process is chemisorption with the formation of strong bonds. The high values of the correlation coefficients of the other kinetic models (pseudo-second order and Elovich) show that the adsorption process is better describe by these models with the possibility of multilayer adsorption, which is in accordance with the type of isotherm graph (Figure 6) obtained with influence of equilibrium concentration. This means that after adsorption on the first layer, there is condensation process on the second layer. The high correlation coefficients of the kinetic intra-particle diffusion model also justify the fact that the adsorption process is also being controlled by diffusion of adsorbate molecules into the internal pores of the CNS and the AC adsorbents. The small diffusion constant value K_{id} for the adsorbents implies that diffusion rate of IC is slow; this favors strong interaction between adsorbents and adsorbate. The R^2 values for kinetic intra-particle diffusion models are smaller than that of

pseudo- second order model for the three adsorbents which is an indication that the intra-particle diffusion step is not the limiting step in the adsorption mechanism. The Elovich adsorption speed constant (α) for the three adsorbents is greater than the desorption coefficient β certifies without fear of uncertainty that the adsorption process is chemisorption. The Q_e (mg/g) values for the pseudo-first order kinetic model and pseudo-second order kinetic model for all the adsorbents are approximately equal to the experimental values. It can therefore be concluded that these kinetic models greatly influence the adsorption mechanism of IC with the adsorbents and hence described the adsorption process perfectly.

CONCLUSION

The aim of this work was to study the removal of IC dye from aqueous solution onto dried cola nut shells and its active carbons. Results show that the removal of IC dye by these adsorbents was achieved at pH 2. The effect of contact time at pH 2 gave 45 min for CNS and 35 min for AC based H_3PO_4 and AC based KOH at equilibrium. Studies of the variation of mass shows that the highest quantity adsorbed was attained with an adsorbent dose of 0.08 g for the three adsorbents. The variation of initial concentration shows that, as the initial concentration increased, the quantity adsorbed increases. The isotherm models were characterized by a type IV for CNS and type II for AC based H_3PO_4 and AC based KOH. The Langmuir model describes adsorption better with high correlation coefficients for CNS and AC adsorbents. Freundlich model describes well for CNS and AC based KOH indicating the existence of multi-layer adsorption. Tempkin model gave a good description with CNS and AC based KOH while the D-K-R was better with CNS. The Langmuir separation factor shows that adsorption was favorable and the free energy for D-K-R indicates chemisorption. Kinetic studies for the adsorption of IC shows the different correlation coefficients. It was observed that the correlation coefficient of pseudo-second order kinetic model was high enough which indicates a chemical adsorption phenomenon. The adsorption speed constant α for the three cases are high implying that after adsorption, it is difficult for desorption to take place hence indicating chemisorption reaction. The adsorption capacity of CNS was higher than that of AC based H_3PO_4 and slightly lower than that of AC based KOH.

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REFERENCES

- [1] M. Nourozi, T. Chuah and S. Thomas, "Adsorption of reactive dyes by palm kernel shell activated carbon: Application of film surface and film pore models", *E-Journal of Chemistry*, Vol. 6, 2009, pp. 949-954.
- [2] R. Malarvizhi and N. Sulochana, "Sorption isotherm and kinetic studies of Methylene blue uptake onto activated carbon prepared from wood apple shell", *Journal of Environmental Protection Science*, Vol. 2, pp. 40-60.
- [3] C. Min-Shen, H. Pang-Yen and L. Hasing-Ya, "Adsorption behavior of dye AAVN and RB4 in acid solutions on chemically cross-linked Chitosan Beads", *Journal of China Institute and Chemical Engineering* Vol. 34, 2003, pp. 625-634.
- [4] J. Ndi Nsami and J. Ketcha.Mbadcam, "The Adsorption Efficiency of Chemically Prepared Activated Carbon from Cola Nut Shells by $ZnCl_2$ on Methylene Blue", *Journal of Chemistry*, Vol. 201, 2013, pp. 1-7.
- [5] B. Sumalatha, Y. Kumar, K. Kiran, J. Babu, N. Venkata, D. Maria and T. Venkateswarulu, "Removal of Indigo Carmine from aqueous solution by using Activated Carbon", *Research Journal Pharmaceutical, Biological and chemical science*, Vol. 5, 2014, p. 912.
- [6] P. Himanshu and R. Vashi, "Decolonization of Dyeing Mill waste water by adsorption and Coagulation", *E-Journal of chemistry*, Vol. 7, 2010, pp. 1468-1476.
- [7] B. Miriam, R. Ramero, R. Gabriela, B. Carlose, T. Teresa and V. Reyna, "Ozonation of Indigo carmine catalyzed with Fe-Pillard clay", *International Journal of photo energy*, Vol. 7, pp. 1-8.

- [8] S. Anagho Gabche, J. Ketcha Mbadcam, D. Tchuifon Tchuifon and J. Nsami Nsami “Kinetic and equilibrium studies of the adsorption of Mercury (II) ions from aqueous solution using Kaolinite and Metakaolinite clays from southern Cameroon”, *International Journal of Research and chemical Environment*, Vol. 3, No. 2013, 2012, pp. 1-11
- [9] J Ndi Nsami, J. Ketcha Mbadcam., S. Anagho Gabche, J . Ghogomu Numbonui and P. D. Belibi Belibi., “Physical and chemical characteristics of activated carbon prepared by pyrolysis of chemically treated Cola nut (*cola acuminata*) Shells wastes and its ability to adsorb organics”, *International Journal of Advance Chemical Technology*, Vol. 3, 2014, pp. 1-12.
- [10] G. Fumba, J. S. Essomba, G. M. Tagne, J. Ndi Nsami., P. D. Belibi Belibi and J. Ketcha Mbadcam, “Equilibrium and Kinetic adsorption Studies of Methyl Orange from Aqueous Solutions Using Kaolinite, Metakaolinite and Activated Geopolymer as Low Cost Adsorbents”, *Journal of Academia and Industrial Research*, Vol. 3, 2014, pp. 156-163.
- [11] P. A Kouoh Sone, G. M. Tagne, B. R. Lekene Ngouateu, P. D. Belibi Belibi, J. Ndi Nsami, D. Kouotou, J. Ghogomu Numbonui, S. Anagho Gabche and J. Ketcha Mbadcam, “Kinetics and equilibrium studies of the adsorption of nickel(II) ions from aqueous solution onto modified natural and synthetic iron oxide”, *International Journal of Basic and Applied Sciences*, Vol. 4, 2015, pp. 277-287.
- [12] A. Mohammed and O. Gemal, “Adsorption of Tatzazine on medical Activated Charcoal tablets under control condition”, *Journal of Environmental and Analytical Chemistry*, Vol. 1, 2014, pp. 1-7.
- [13] A. Dada, A. Olalekan, M. Olatunya and O. Dada, “Langmuir, Freundlich, Temkin and Dubinin–Radushkevich Isotherms Studies of Equilibrium Sorption of Zn²⁺ Unto Phosphoric Acid Modified Rice Husk”, *Journal of Applied Chemistry*, Vol. 3, 2012, pp. 38-45.
- [14] A. Mona, A. Soheir and A. Amina, “Basic Dye adsorption on Low Cost Biopolymer: Kinetic and Equilibrium Studies”, *Journal of Applied Chemistry*, Vol. 2, 2012, pp. 27-36.
- [15] J. Ketcha Mbadcam, H. Ngomo Manga, C. Tcheka, N. Abdoul Rahman, H. Djoyo and D. Kouotou, "Batch Equilibrium Adsorption of Cyanides from Aqueous Solution onto Copper- and Nickel-Impregnated Powder Activated Carbon and Clay", *Journal of Environmental Protection Science*, Vol. 3, 2009, pp. 53-57.
- [16] G. M. Tagne, J. Ndi Nsami and J. Ketcha Mbadcam., “Adsorption of Copper (II) Ions from Aqueous Solution onto Synthetic Goethite and Two Naturally Available Red Soils from Yaoundé-Cameroun”, *British Biotechnology Journal*, Vol. 2, 2013, pp. 221-235.
- [17] M. Samarghandi, M. Hadi, S. Moayedi and A. Barjasteh, “Two-Parameter Isotherms of Methyl Orange Sorption by Pinecone Derived Activated Carbon”, *Iranian Journal of Environmental Health and Scientific Engineering*, Vol. 6, 2009, pp. 285-29
- [18] D. D. Dina Joh, N. Abduol Rahman, J. Ndi Nsami and J. Ketcha Mbadcam, “Adsorption of Acetic Acid Onto Activated Carbons Obtained From Maize Cobs by Chemical Activation with Zinc Chloride (ZnCl₂)”, *Research journal of Chemical Sciences*, Vol. 2, 2012, pp. 42-49.
- [19] J. Ketcha Mbadcam., S. Dongmo and D. Dinka’a. Ndaghu, “Kinetic and Thermodynamic studies of the adsorption of Nickel (II) Ions from aqueous solutions by Smectite Clay from Sabga-Cameroon”, *International Journal of Current Research*, Vol. 4, 2012, pp. 162-167.
- [20] D. Fungaro, M. Yamaura and T. Carvalho, “Adsorption of anionic dyes from aqueous solution on Zeolite from fly ash-iron oxide magnetic nanocomposite”, *Chemical and Environmental Technology Center, Nuclear and Energy Research Institute*, Vol. 2, 2011, pp. 305-316.

LIST OF FIGURES

**Figure 1:
Carmin**

Structure of Indigo

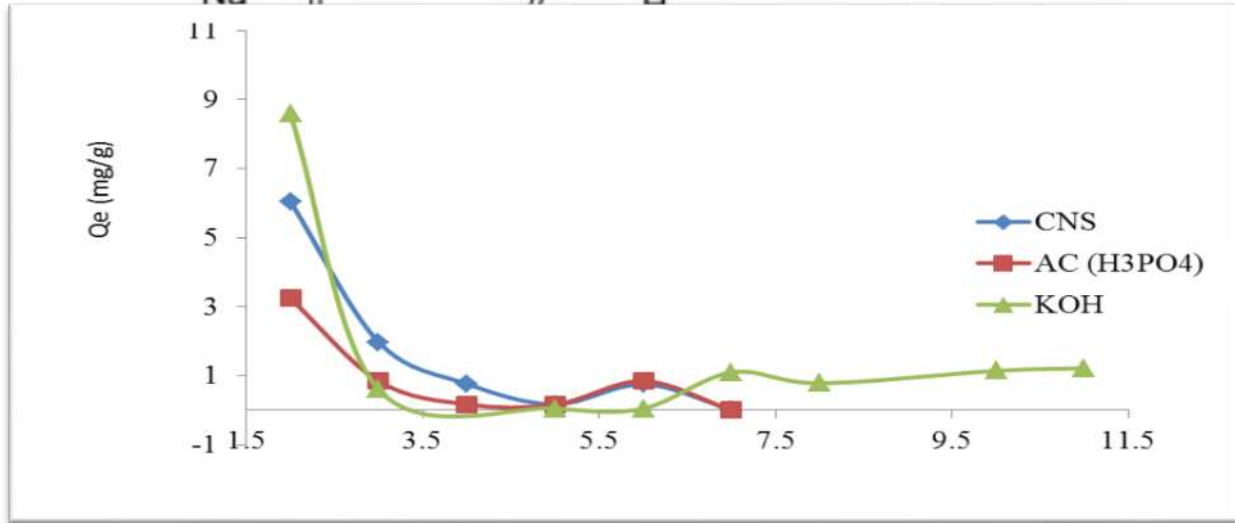


Figure 2: Influence of pH for IC adsorption

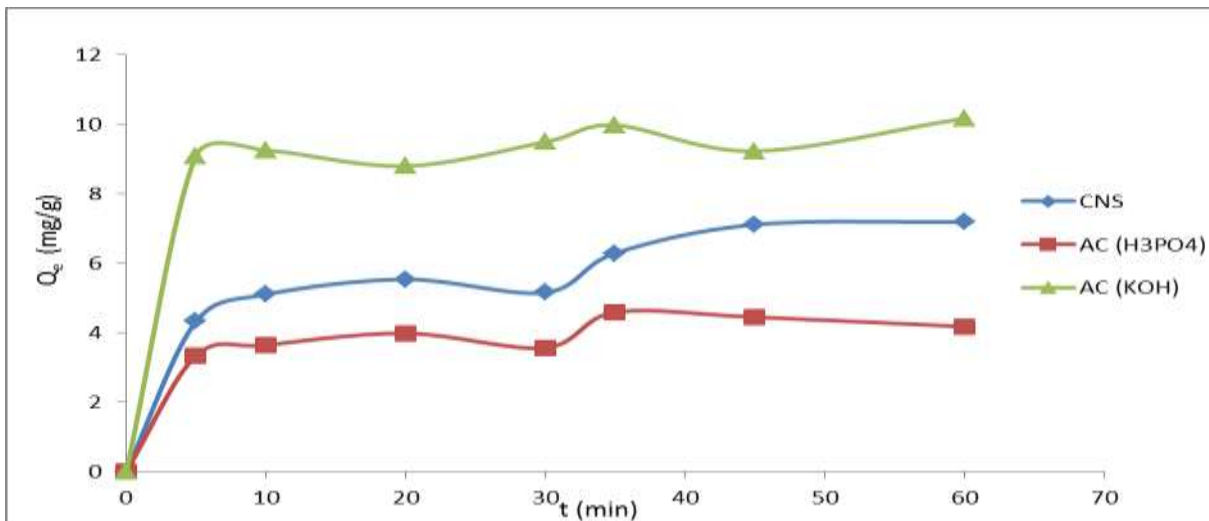


Figure 3: Influence of contact time on IC adsorption

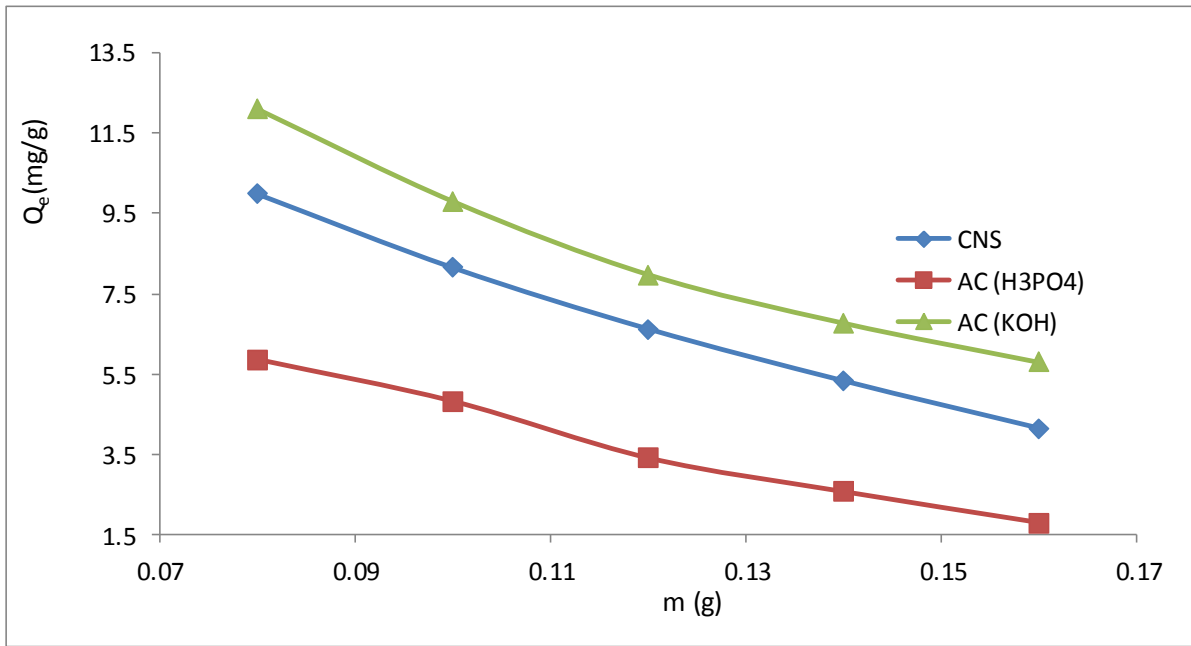


Figure 4: Variation of adsorbent dose on adsorption of IC dye.

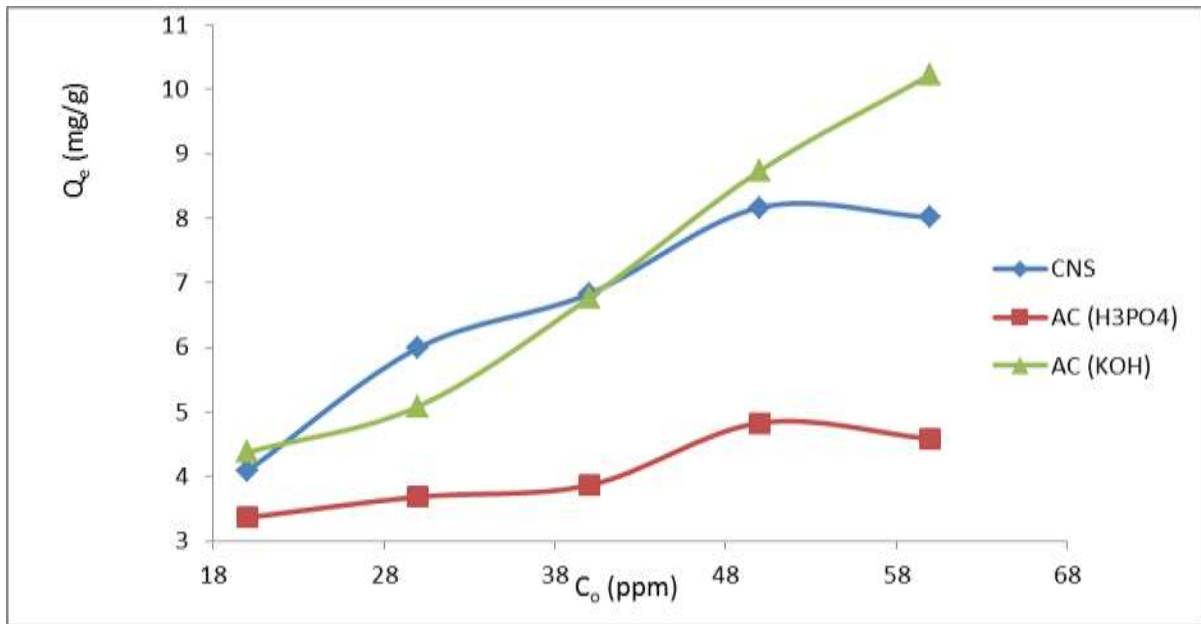


Figure 5: Influence of initial dye concentration with the quantity adsorbed

ADSORPTION ISOTHERM STUDIES

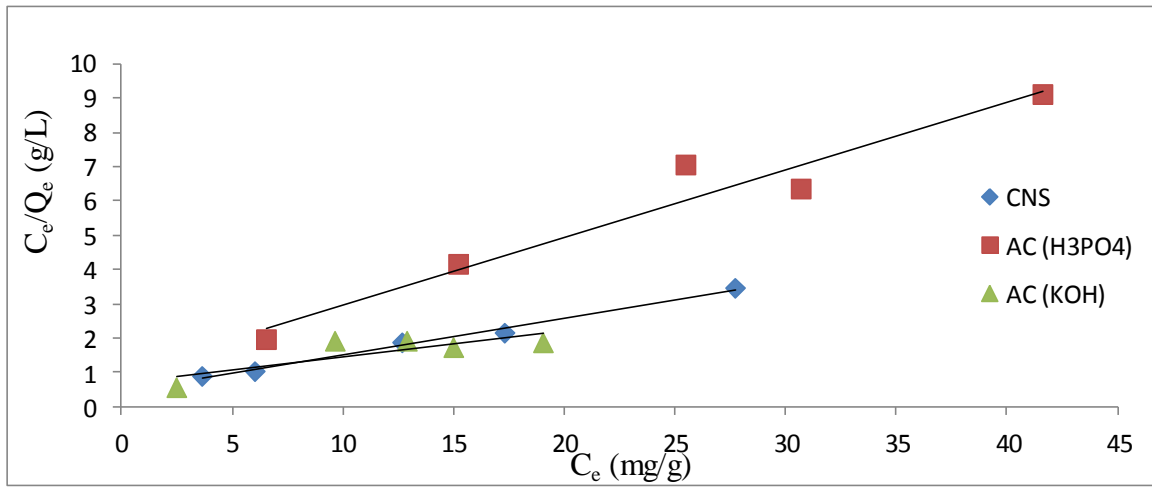


Figure 6: Linear transformation of Langmuir isotherm

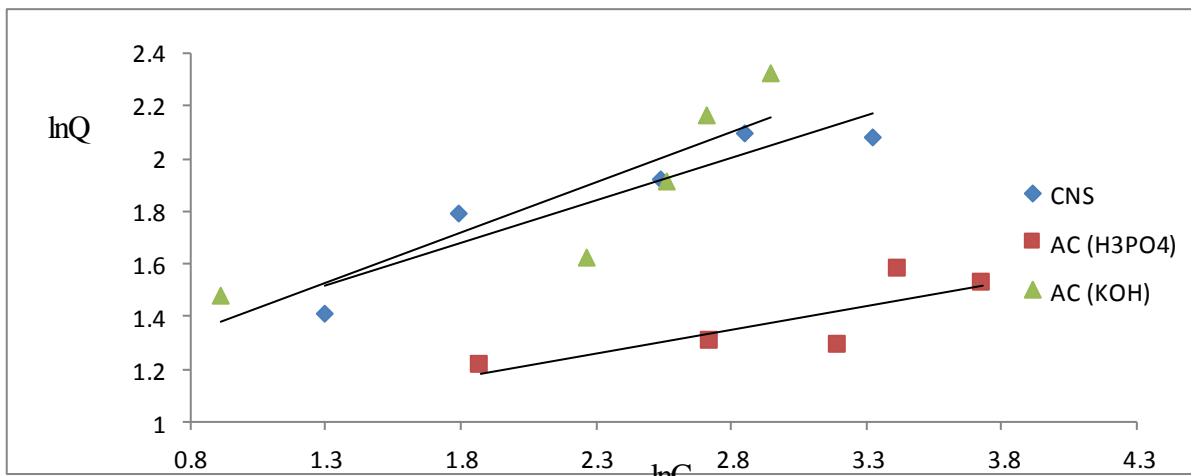


Figure 7: Linear transformation of Freundlich isotherm

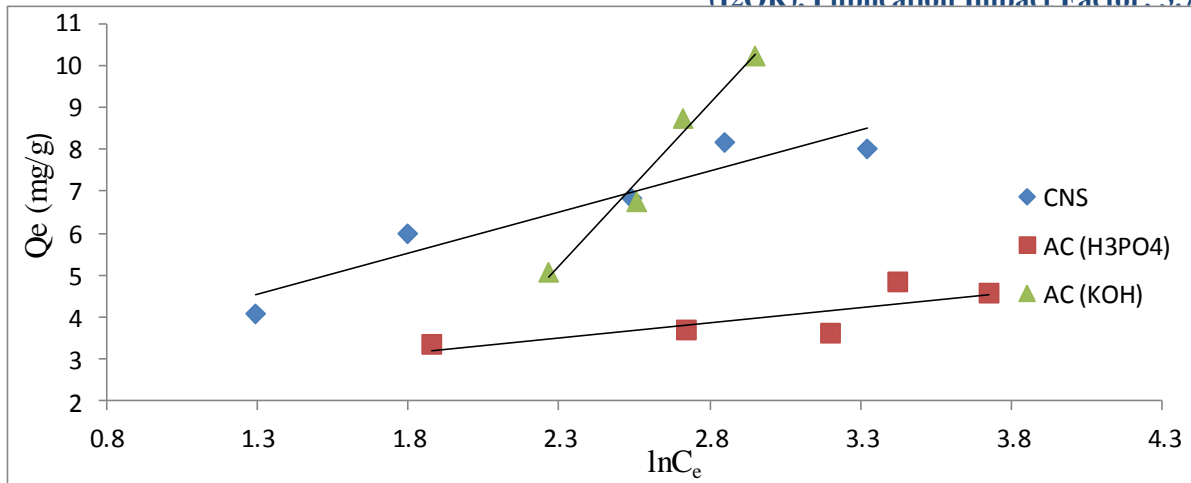


Figure 8: Linear Transformation of Tempkin Isotherm

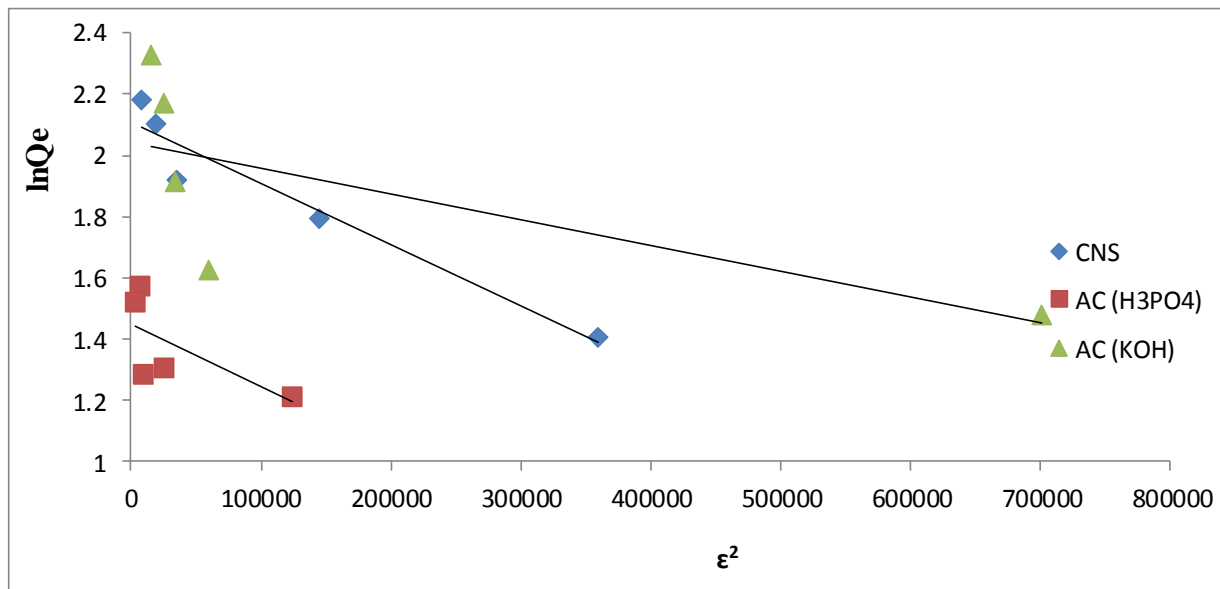


Figure 9: D-K-R linear plot

ADSORPTION KINETIC STUDIES

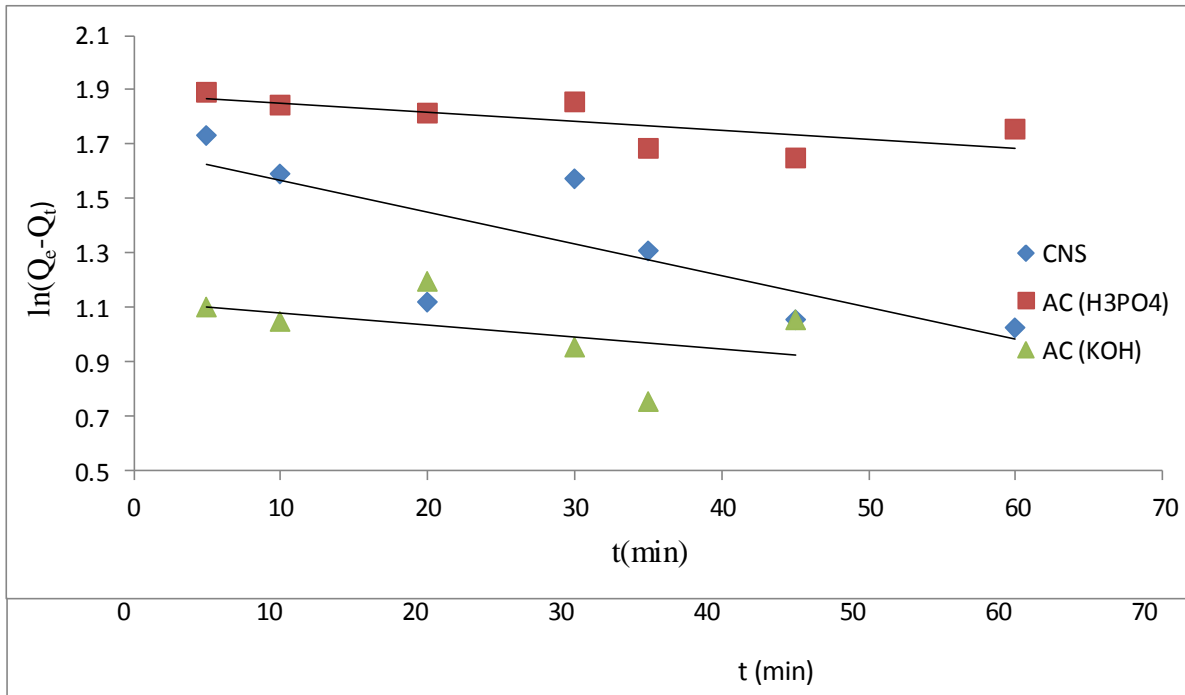


Figure 10: Linearized form of pseudo-first order kinetic model

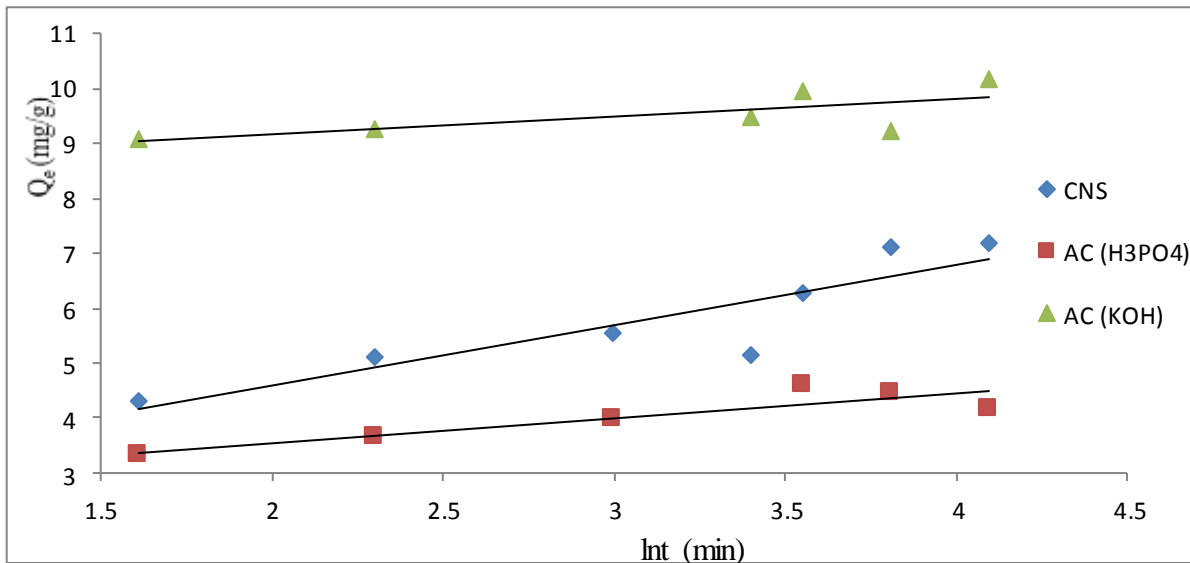


Figure 11: Linearized form of pseudo-second order kinetic model

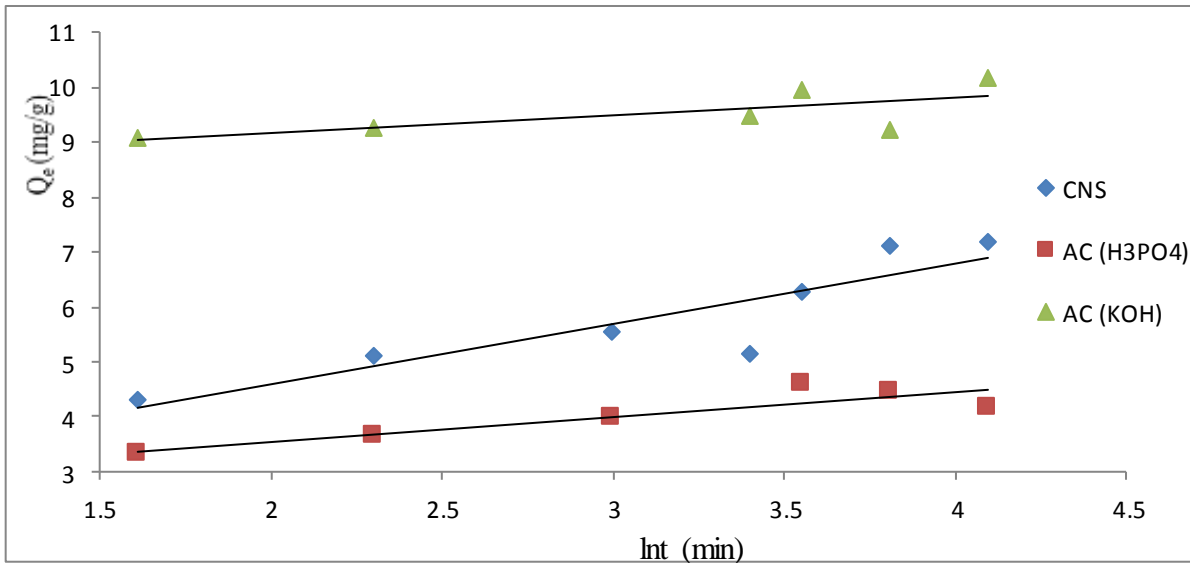


Figure12: Linear form of Elovich model

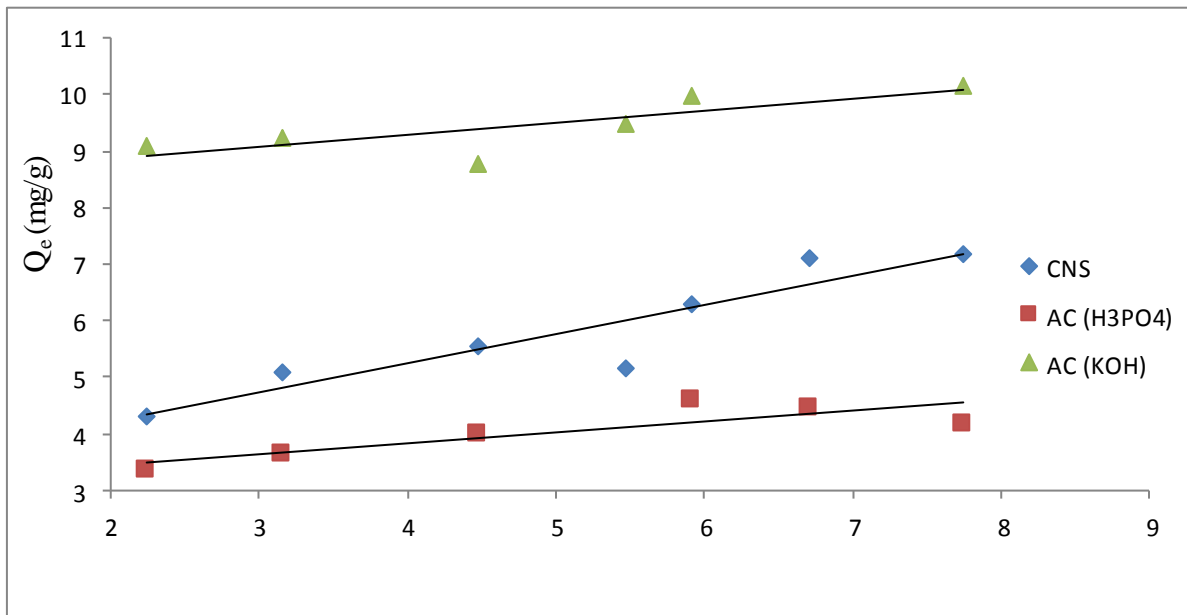


Figure13: Intra-particle diffusion model

LISTE OF TABLES

Table I: Correlation coefficients and linear constants of Langmuir, Freundlich, Tempkin and D-K-R for the adsorption of IC.

Model	Parameter	CNS	AC (H ₃ PO ₄)	AC (KOH)
Langmuir	R²	0.9895	0.9439	0.6519
	Q _m (mg/g)(theoretically)	9.346	5.089	13.405
	K _L (L/mg)	0.253	0.194	0.105
	Q _m (mg/g)(Experimentally)	8.017	4.587	10.227
Freundlich	R²	0.8766	0.6686	0.7427
	K _F (mg/g)(L/mg) ^{-1/n}	2.986	2.324	2.809
	1/n	0.3245	0.1796	0.3812
Tempkin	R²	0.908	0.6427	0.9773
	K _T (L/mg)	2.773	13.344	5.112
	b _T (KJ/mol)	1265.552	3442.507	318.409
D-K-R	R²	0.9375	0.442	0.4989
	Q _m (mg/g)	8.213	4.261	7.709
	E (KJ/mol)	500	500	790.569

Table II: Correlation coefficients and linear constants of kinetic models.

Parameter		CNS	AC (H ₃ PO ₄)	AC (KOH)
Pseudo-first order	R²	0.6129	0.4964	0.2032
	K ₁ (min ⁻¹)	0.0116	0.4964	0.0044
	Q _e (mg/g)	5.385	6.606	3093
Pseudo-second order	R²	0.956	0.9794	0.9931
	K ₂ (g.min ⁻¹ .mg ⁻¹)	0.0173	0.051	9.821 x 10 ⁻³
	Q _e (mg/g)	7.633	4.442	10.091
Elovich	R²	0.8007	0.8015	0.4935
	α (mg/g/min)	33.228	13.226	1.340 x 10 ⁸
	β (g/mg)	0.909	2.232	3.119
Intra-particle Diffusion	R²	0.8534	0.7122	0.641
	K _{id} ((g.min ⁻¹ .mg ⁻¹)	0.5136	0.1898	0.2121
	C	3.1915	3.0712	8.431