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Removal of Dyes from Aqueous Solution Using Neem (*Azadirachta Indica*) Husk as Activated Carbon

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ABSTRACT

The adsorption of xylenol orange, remazole turquoise blue and procion red by neem (Azadirachta indica) husk carbon activated with $ZnCl_2$, H_3PO_4 and KOH was examined at different concentrations and at the optimized particle size, shaking time and adsorbent dosage. The adsorption obeyed both Langmuir and Freundlich isotherms but fit Langmuir isotherm than Freundlich isotherm. Adsorption studies were carried out using carbonized neem carbon, activated neem carbon and commercial adsorbent from coconut shell on xylenol orange, remazole turquoise blue and procion red. The study shows a high adsorption for remazol turquoise blue.

INTRODUCTION

Industries like plastic, paper, textile and cosmetics use dyes to colour their products. These dyes are common water pollutants and they may be frequently found in trace quantities in industrial waste water. Their presence in water, even at very low concentrations, is highly visible and undesirable. When these coloured effluents enter rivers or any surface water system they upset biological activity. Ground–water systems are also affected by these pollutants because of leaching from the soil. In addition, many dyes are difficult to degrade due to their complex aromatic structure and they tend to persist in the environment and creating serious water quality and public health problems such as allergic dermatitis, skin irritation, cancer and mutation [1] and [2]. In the past a number of conventional biological treatment processes have been used which were not effective, some of which include coagulation and chemical oxidation, membrane separation process, electrochemical, reverse osmosis and aerobic and anaerobic microbial degradation but all these methods suffer from one or more limitations and none of them were successful for the complete removal of dye. It has been discovered that dyes can be effectively removed by adsorption process in which dissolved dye compounds were attached to the surface f the adsorbents. Researchers have exploited many low cost and biodegradable effective adsorbents. They were obtained from natural sources which successfully removed dye from aqueous solutions. The present research tends to investigate neem (Azadirachta indica) husk as a low-cost adsorbent for the removal of dyes from aqueous solution.

MATERIALS AND METHODS

Materials

Sample (Neem husk) was collected from the National Research Institute for Chemical Technology (NARICT) Bassawa, Zaria. Neem husk was washed properly under rushing tap water to remove water-soluble impurities and then dried in a thermostatic oven at 105°C for 24 hours. The dried neem husk was pound using mortar and pestle. These was then sieved with three different sieves made up of brass frame and steel mesh and an aperture of $355\mu m$. The fine sieved neem husk was stored in a clean airtight container. These fine adsorbent was carbonized at 400°C and activated with H_3PO_4, KOH and $ZnCl_2$ at different optimized temperatures.

The adsorbates used were xylenol orange, procion red and remazolturqoise blue. Stock solutions of each of the dyes were prepared. The maximum wavelength of each of the dyes were determi

Methods

25ml of the different adsorbate solutions were measured into different conical flasks and 0.2g of the adsorbent were weighed into each of the flasks containing the different adsorbate solutions; this was shaken at the optimized shaken time using a mechanical shaker and then allowed to attend equilibrium. After the specific shaking time, the solutions were filtered using whatman filter paper and analyzed using a colorimeter. The experiment was repeated twice and the average taken.

3.0 Results and Discussion

3.1 Adsorption of xylenol orange on activated neem carbon, carbonized neem carbon and commercial activated carbon from coconut shell

The amount of xylenol orange adsorbed at various initial concentrations is depicted in figure 1. The initial concentration with the optimum adsorption of xylenol orange is found to be at 90ppm. Here, optimum concentration of dyes refers to the concentration at which the maximum removal of dye was noticed [3]. This may be attributed to multi layer adsorption on the surface of the adsorbed dye which is not possible at low concentration due to the availability of free sites but at high concentration these available sites most have been saturated and so there might be further adsorption on the surface of the already adsorbed adsorbate which tend to agree with one of the assumptions in the derivation of the Freundlich adsorption isotherm.

It was observed that the neem carbon impregnated with the activating reagents adsorbed better than the carbonized neem carbon, these shows that the impregnation was actually successful and effective, which agrees to what [4] reported that the activating agent acts as a support and does not allow the carbon pore network to shrink or collapse. The activating chemical achieved this by generating an activating gas that envelopes the mass of the carbon material and in this manner uniform gas activation is achieved.

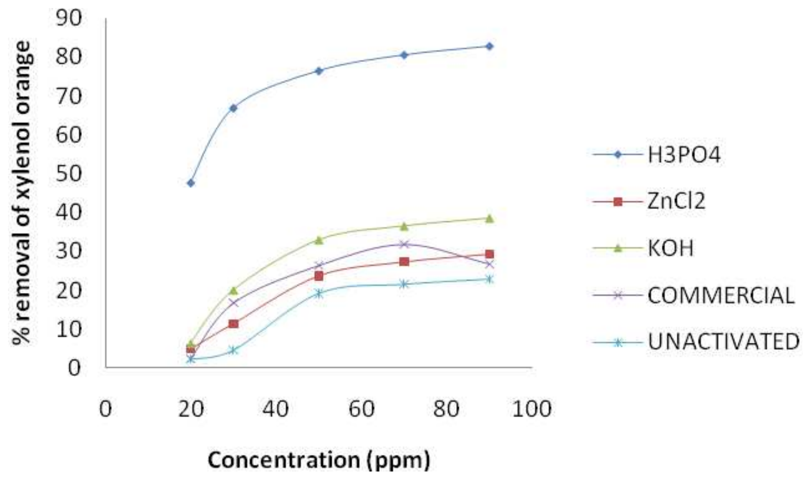

Figure 1 Amount of xylenol orange adsorbed on neem carbon and commercial activated carbon

*3.2 Adsorption of procion red on activated neem carbon, ca*rbonized *neem carbon and commercial activated carbon*

The initial concentration of procion red with the optimum adsorption of xylenol orange was found to be at 5ppm. The adsorption sites took up the available solute more quickly at low concentrations [5]. The percent removal of procion red decreases from 5 to 60ppm for all the adsorbents used as shown in Figure 2. The adsorbent activated with $ZnCl_2$ gave the highest percentage procion red adsorption (81%) compared to the other adsorbents, followed by the adsorbent activated with H_3PO_4, then the commercial activated carbon while the neem carbon activated with KOH was the least.

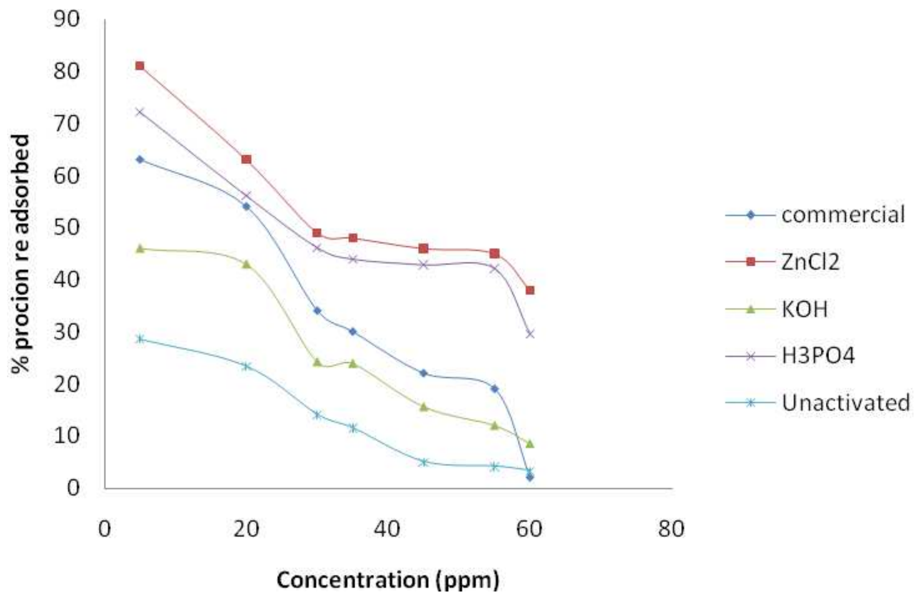

Figure 2 Adsorption of Procion red on neem carbon and commercial adsorbent

3.3Adsorption of remazol turquoise blue on activated neem carbon, carbonized neem carbon and commercial activated carbon

The lowest initial concentration among the ranged studied gave the optimum adsorption of remazol turquoise (12-20ppm). The neem carbon activated with $ZnCl_2$ gave the highest percentage removal of remazol turquoise (99%) compared to the other adsorbents, followed by the adsorbent activated with H_3PO_4 (79%), then the commercial activated carbon (74%) while the neem carbon activated with KOH (68%) gave the least adsorption and if these is compared with the carbonized neem carbon value (32%), it means the activating agent actually inproves the pore system (see figure 3).

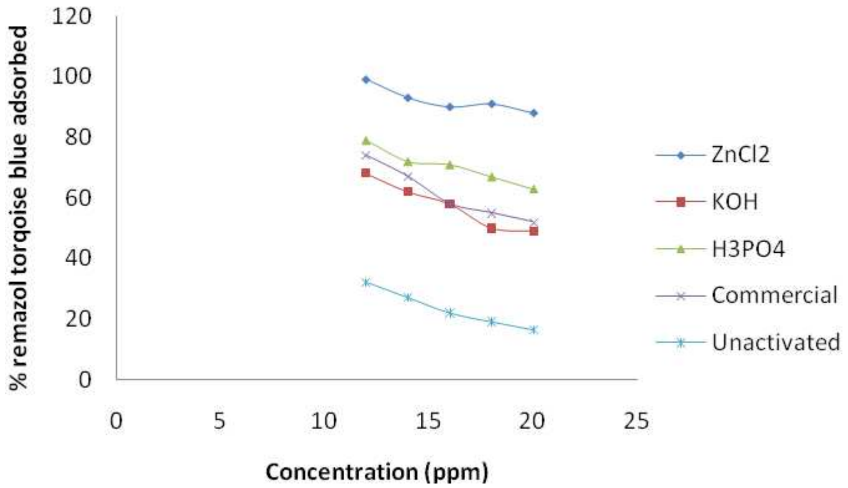

Figure 3 Adsorption of remazole turquoise blue on neem carbon and commercial adsorbent.

3.4 Adsorption Isotherm

The Freundlich constants n and K_f and Langmuir constants Q_o and b for xylenol orange, procion red and remazole turqoise adsorption on neem carbon activated with KOH, H_3PO_4, and $ZnCl_2$, carbonized neem carbon and commercial activated carbon were calculated. Where the Freundlich constant n is the index for adsorption intensity, K_f is for adsorption capacity where as Q_o and b are the Langmuir constants which is a measure of the monolayer adsorption capacity and surface energy of adsorption respectively . Another important parameter is the correlation coefficient R^2 which measures the fitness of the experimental investigations to either Freundlich or Langmuir model. According to [6], that the higher the value of Q_o and b the better the effectiveness of adsorbent capacity. The parameters K_f and n values of the Freundlich isotherm are characteristic of the adsorption system and give important information about adsorption phenomenon and that K_f parameter increases with the total adsorption capacity of the adsorbent to bind the adsorbate whereas the n value is a useful index of adsorption efficiency and may vary along the adsorption process [7]. A range of values for the Langmuir and Freundlich constants base on 120 data point were evaluated according to [8]; b ranges from 0.0092 to 333.33 while Q_o is from 0.00109 to 2.58 while n values ranges from 0.4428 to 25 and K_f from 0.06 to 727.05. Table 1, is the summary of the parameters obtained from both Freundlich and Langmuir models. The isotherm for xylenol orange fit the Freundlich isotherm with an R^2 of 0.914 than the Langmuir Isotherm (R^2 is 0.54), procion red fit better into the Langmuir Isotherm as shown from there R^2 in table 1 while remazole turquoise blue fit both into the two isotherms but it better fit in the Langmuir isotherm.

Table 1: Langmuir, Freundlich constants and separation factor (R_L) for adsorbents

Xylenol orange						
Model	**Parameters**	**KOH**	**H_3PO_4**	**$ZnCl_2$**	**Commercial**	**Unactivated**
Freundlich	R^2	0.902	0.77	0.949	0.982	0.966
	n	0.34	0.25	0.312	0.402	0.37
	K_f	51.64	12.22	54.08	26.12	49.09
Langmuir	R_L	5.12×10^{-3} - 0.463				
	R^2	0.523	0.445	0.696	0.382	0.65
	Q	0.104	47.62	0.21	0.311	0.097
	b	2.16	0.058	0.085	0.07	0.191
Procion red						
Freundlich	R^2	0.431	0.949	0.984	0.54	0.061
	n	3.5	1.92	2.04	3.56	8.85
	K_f	12.56	16.29	20.99	18.41	9.64
Langmuir	R_L	1.79×10^{-3} - 0.169				
	R^2	0.913	0.909	0.929	0.913	0.914
	Q	0.04	0.09	0.07	0.026	0.108
	b	5.16	1.834	1.71	9.3	0.986
Remazolturqoise blue						
Freundlich	R^2	0.926	0.948	0.635	0.887	0.985
	n	3.344	3.846	1.124	7.634	4.587
	K_f	23.714	37.584	72.95	39.084	30.69
Langmuir	R_L	7.79×10^{-4} - 0.0128				
	R^2	0.904	0.99	0.964	0.989	0.997
	Q	0.028	0.0133	0.0116	0.0182	0.0695
	b	11.2	48.88	61.03	64.88	6.402

CONCLUSIONS

The present investigation showed that neem (*Azadirachta indica*) husk activated with $ZnCl_2$, H_3PO_4 and KOH can be effectively used as a raw material for the removal of xylenol orange, procion red and remazolturqoise blue. Neem activated carbon competed favorably with the commercial adsorbent from coconut shell for the adsorption of xylenol orange, procion red and remazolturqoise blue. The three dyes used were subjected to both Langmuir and Freundlich isotherm model; Xylenol orange fit better into the Freundlich isotherm model than the Langmuir isotherm model, procion red fit better to the Langmuir isotherm model than the Freundlich model while remazolturqoise blue adsorption model fit to the two isotherms but fit better in the Langmuir model. Both the Freundlich and Langmuir constants falls within the range reported in literatures as shown in table 1.

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