Adsorption Of Methylene Blue From Water Matrix Onto Naoh Modified Lantana

Joydeep Dutta[1], Bilal A Bhat[2]
[1&2]*Department of Zoology, Lovely Professional University*
Phagwara, Punjab
Email :joydeep.dutta@lpu.co.in

1. INTRODUCTION

Clean water is requirement for all. Methylene blue is one of the most commonly used substances in the textile industry. Methylene blue can damage the balance of the ecosystem and adversely affect the environment [1–3]. The methylene blue compound is very easy to obtain because its cheap price and wide use in dyeing process [4]. The dye molecule is very stable, thus difficult to be decomposed under natural conditions. Therefore, it is necessary to remove the dye molecule before being discharged to the environment [5]. The removal technique of dyestuff has been widely studied including ion exchange, membrane filtration, coagulation-flocculation, flotation,
electrochemical processes, reverse osmosis and adsorption [6–8]. But due to various advantages and disadvantages adsorption has been given the prime importance and is frequently used [9]. Table 1 depicts the merits and demerits of other technologies for dye removal from water [10]. The adsorption method is recognized as a relatively simple and an effective method [5,11].
For adsorption process various commercially available activated carbon is the most widely used adsorbent for dye removal, but they are too expensive. Therefore, numerous low-cost alternative adsorbents from have been proposed including chemically modified sugarcane, lignin, pistachio hull waste, coffee husk-based activated carbon, pine cone, rice husk, synthetic calcium phosphates, natural untreated clay, pillared clays and swelling clays, Lantana, peanut hull [12].

Lantana camara is abundantly available waste material which has shown promising results in the form of adsorbents [13-20]. Lantana biomass is a composite compound contains various functional groups ,these functional groups are notably –OH,C–O–C ,C–HN–H. Those functional groups enhances the capture of dye molecules by sharing their pair of electrons to the different cationic adsorbate molecules, resulting in the formation of complexes [18]. These properties of this adsorbent leads to facilitate greater removal of dyes from the aqueous solution. Limitations noted with number of the studies using Lantana plant and its parts for adsorption of dyes. The adsorption property of any adsorbent could be increased by cross liking it with other chemicals like KOH [12], HCl [21], NaOH [22], sulphuric acid, sodium hydroxide and PEI [23], formaldehyde and sulfuric acid [24]. Moreover the adsorption property of Lantana can be increased by modifying it with different chemicals, commonly used modifying agents are Oxalic acid, Citric acid, Formyldehyde, HCL, NaOH, SDS, Sulphuric acid KOH [18]. This study represents the study of removal of methylene blue dye using KOH/NaOH modified *Lantana camara* leaves.

In the current assessment, methylene blue is used as a shading to be expelled from its manufactured water by utilizing changed leaves of EC. Methylene blue has progressively broad applications, which consolidate shading paper, ephemeral hair colorant, shading cottons, wools, additionally, covering for paper stock. Notwithstanding the way that methylene blue isn't immovably hazardous, it can cause some hurtful effects like Heinz body game plan, heaving, cyanosis, jaundice, stun, increment pulse, quadriplegia, and tissue spoil in individuals [25].

2. MATERIALS AND METHODS

Preparation of adsorbent:

The *Lantana camara* used in this study were obtained from university area near Education Block Lovely professional university, Punjab India. The leaves were removed from the stem and were repeatedly washed using tap water followed by distilled water to remove suspended impurities and dust particles. The washed material was then dried in an oven for 18 hours at a temperature of 60^0C. Grinder (Philips) was used to crush dried leaves into small particles to powder form. The powdered form obtained was sieved to mesh size of 150 um .The remaining outsized material was again grinded for further size reduction to obtain powdered form. The sieved material obtained was stored in poly zipper bags and was stored in desiccator to avoid the moisture and dust particles for further experimental use and modification [26].

Modification of adsorbent:

Sodium Hydroxide (NaOH) Modification:

4g and 2g of NaOH pellets were dissolved in 1litre of distilled water separately, for the preparation 0.1M and 0.05 M NaOH solution respectively. For modification 1g of Lantana powder was immersed in 15ml of 0.05M and 0.1M NaOH separately stirred well and was kept for 24 hrs under room temperature. The mixtures were then washed with distilled water until the pH reaches almost 7. The material was filtered using Whatman filter paper number 4 and was kept in an oven for overnight at 60^0C. The solid residue was collected and was grinded into powder form by using pestle and mortar, the powder was then stored in zipper bags for experimental use [27].

*Preparation of adsorbates***:**

Methylene blue, color (Loba Chemie) was picked as an adsorbent on account of its known solid adsorption onto solids [27]. The color stock arrangement was set up by dissolving precisely weighed 1g of powder in 1000ml of distilled water [28].). The working solution was prepared by further dilution of stock solution using distilled water. The final concentration of working solution was 200mg/dm3 [29].

Batch adsorption experiments:

The batch experiments were performed in 250ml conical flasks with varying dose of adsorbent (500mg, 750mg, and 1000mg) and contact time (30, 60, 90 and 120 minutes) in 100 ml of working solution. The flasks were agitated by rotatory shaker at constant rotation (200 rpm).The constant factors were initial dye concentration (200mg/dm3) and temperature (room temp). The samples were taken at each contact time interval, the solution was filtered using Whitman filter paper No.4. The filtrate obtained is subjected to spectrophotometric analysis to measure final absorbance at 539 nm.

Calculations: ‘T’ test was employed at 5% significance level.

The final concentration of solution was measured using formula:

$$C1/C2 = A1/A2$$

Where,
C1 = Initial concentration of dye, C2 = Final concentration of dye, A1 = Initial Absorbance, A2 = Final Absorbance.

The dye removal percentage was calculated using formula:

$$q = [C1\text{-}C2/C1] \times 100$$ (Coskun *et al.*, 2017)

Where, C1 = Initial concentration of dye. C2 = Final concentration of dye. q = percentage of dye removed.

The experiments were conducted in triplets and the average value was calculated to measure the percent removal of dye.

3. RESULTS

Adsorption by 0.05M NaOH modified Lantana camara leaves:
The modification resulted in the formation of new sites on the surface of adsorbent, and leads in the improvement of adsorption capacity in comparison to unmodified. The modification was done up to 24 hours at room temperature. Both the modified and unmodified material was tested simultaneously for adsorption with varying dose of adsorbent from 500mg to 1000mg (500mg,750, and 1000mg) and varying contact time from 30, 60, 90 and 120 minutes respectively.
The adsorption of methylene blue at 500mg adsorbent dose is represented in **Fig.1** the process of adsorption was found steady in both modified and unmodified adsorbent with the increase in contact time there is not much increase in the % dye adsorption it means that the equilibrium starts establishing just quickly after the incorporation of adsorbent into the dye solution. Maximum adsorption was achieved at 30 minutes of contact time for both modified and unmodified material and the equilibrium was achieved at 90 minutes of contact time.
Representation of dye adsorption at 750mg of adsorbent dose is shown in **Fig 2**. The influence of contact time is clearly seen in the graph, the demonstrations from is clear that with the increase in contact time there is change in the rate of dye adsorption., maximum removal is analyzed at 60 minutes of contact time by both modified and unmodified material after that there is lag in adsorption, and the equilibrium was established at 90 minutes of contact time.

Illustration of dye removal at 1000mg of adsorbent dose is shown in **Fig.3** the rate of dye removal shows a little difference between modified and unmodified material. The modified material shows an increase in dye adsorption with the increase in contact time, but in case of unmodified material decrease in dye adsorption rate was seen at 60 minutes of contact, after that the abrupt increase in dye removal was seen maximum adsorption was seen at 90 minutes of contact time for both modified and unmodified material, also the equilibrium was established at 120 minutes of contact time.
The influence of varying adsorbent dose 500mg, 750mg and 1000mg of modified and unmodified material (0.05M NaOH) on the adsorption of dye is illustrated in **Fig.4**. It is clear from the figure that with the variation in the adsorbent dose there is also a change in the adsorption of dye shown by both modified and unmodified adsorbent. With the increase in the adsorbent dose the rate of dye removal decreases. Maximum adsorption in case of modified adsorbent was shown by 500mg of adsorbent dose (91.5%) ,followed by 750 mg

adsorbent dose (89%) and least by 1000mg of adsorbent dose (86%) at 60, 90 1nd 120 minutes of contact respectively. Despite a good amount of dye adsorption was also seen by the unmodified material as well maximum %age of adsorption was seen by 750mg adsorbent dose (88%) followed by 500mg (87.5%) and least by 1000mg of the adsorbent dose(86%) at 60, 30 and 90 minutes of contact time respectively.

Adsorption by 0.1M NaOH modified Lantana camara leaves:
The modification leads in the production of new positions on the adsorbent and results in increasing ability of adsorbent towards the removal of dye, both the modified and unmodified material were tested all together. Higher amount of dye removal was recorded during the adsorption process.

The rate of dye removal % by the modified and unmodified Lantana at 500mg dose of adsorbent is shown in **Fig.5**. There was not much difference in the removal % of dye between modified and unmodified material, each material shows almost equal rate of adsorption, the maximum adsorption was recorded at 90 minutes of contact time, and the equilibrium was obtained at 120 minutes.

The mean % adsorption rate at 750mg of adsorbent dose by unmodified and modified Lantana (0.1M NaOH) is demonstrated in **Fig.6** It is clear from the Fig. that unmodified material shows more efficiency towards adsorption and higher %age of dye was removed by the unmodified material, maximum dye removal percentage was seen at 60 minutes of contact time, there after it starts decreasing and the equilbrium between modified and unmodified was established at 90 minutes of contact time , however in this case the unmodified adsorbent shows better removal as compared to modified Lantana.

The adsorption of dye at 1000mg of adsorbent dose by unmodified and modified Lantana (0.1M NaoH) is represented in **Fig.7.** The unmodified material shows more % rate of dye removal as compared to modified, with the increase in contact time there was abrubt decrease in the removal of dye. The equilibrium was established at 120 minutes of contact time for both modified and unmodified adsorbent.

It is clear from **Fig.:5, 6 and 7** that with the increase in adsorbent dose and contact time there is decrease in mean %age of dye adsorption; the maximum adsorption was recorded at 90 minutes of contact time at 500mg of adsorbent dose for both modified and unmodified material respectively.

The effect of varying adsorbent dose on the adsorption of dy by unmodified and modified Lantana(0.1M NaOH) is shown in Fig. 8.The comparative figure clearly shows that an observable change in the adsorption is noted with the change in adsorbent dose fro 500 to 100mg (500,750 and 1000mg). The rate of adsorption decreases with the increase in adsorbent dose.For modified adsorbent the maximum adsorption rate (90%) was seen at 500mg of adsorbent dose folowed by 750mg of adsorbent dose (76%) and least adsorption rate was shown by 1000mg of adsorbent dose (57%) at 120, 90 and 60 minutes of contact time respectively. Although a better rate of dye removal was recorded by unmodified material over modified ,in case of unmodified 92% of the dye removal was shown by 500m of adsorbent dose at 120 minutes of contact time followed by 750mg and 1000mg adsorbent dose, their percentage was 83% and 70% respectively at 90 and 30 minutes of contact time.

4. DISCUSSION

The use of harmful dyes several industrial sectors eventually drain into water resources along with other wastes. The present focus of the study is to discover a lucid method for the removal of these substances. Adsorption is an economically effective method that results in efficient eradication of such dyes. The adsorption process has been suggested by the numerous researchers after their studies on dye removal [18] this study deals with the adsorption of Methylene blue by Lantana leaves powder of 150µm mesh size, this material was found effective in removing methylene blue from water The result was in steady with the studies of [15]..

Modification of the material proved an important factor in adsorption of dye, the chemical modification leads to the formation of new active sites on adsorbent as compared to unmodified material higher rate of dye removal was recorded by the modified adsorbent. The chemical modification of the material was done by NaOH.

NaOH modification increased the adsorption capacity to a greater extent. 0.5, 0.1 M NaOH solutions were used for the modification of Lantana leaves powder. As the concentration of the NaOH solution used for modification was increased, dye removal efficiency also increased, the increase could be seen due to the porous surface obtained after modification, it can be can be illustrated from the study of [12].

Adsorbent dose in adsorption process is also an important factor. The experiments were conducted at varying adsorbent dose from 500mg, 750 and 100mg respectively, it was seen that with the increase in adsorbent dose the removal of dye also increases and maximum removal of dye was recorded at 1000mg of adsorbent dose. The results indicate that the rate of adsorption increased with increasing adsorbent dose, this is observed due to presence of more sites on the adsorbent material which stick up more dye molecules it can be illustrated by the studies of [30].

Effect of contact time was clearly visible in adsorption process ,with the increase in contact time from 30 to 120 minutes the rate of adsorption also increase ,maximum adsorption was recorded at 90 and 120 minutes of contact. This increasing adsorption was due to the binding of more dye molecules with increasing their contact time with the adsorbent dos, the results were according to the adsorption studies of [31].

Various biosorbents derived from waste materials have been studied with different types of beneficial effects [32-36].

5. CONCLUSION

The adsorption of methylene blue using Lantana leaves powder was studied by varying experimental conditions. The adsorbent is an eco-friendly and economically effective method by using plant waste. The adsorption process was carried out using unmodified and modified adsorbent (Lantana leaves powder). The results showed that modified adsorbent proved to be much effective as compared to the unmodified adsorbent. It was found that dye removal capability increased with increasing contact time (30 to 120 minutes) of the dye solution. It was due to increase sticking and filling of more spaces on the adsorbent with increase in contact duration. The maximum adsorption was assessed at 90 and 120 minutes of contact time the results obtained are found to be statistically significant.

The results clearly confirmed that Lantana leaves powder can be used as an easily available low-cost adsorbent for the removal of dye.

6. REFRENCES

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Table 1:
Different Methods for the removal of dye from industrial effluents, merits and demerits (Tim Robinson *et al., 2000).*

Methods	**Merits**	**Demerits**
Photochemical	No sludge production.	Production of byeproducts.
Electrochemical Destruction	Products formed are nontoxic in nature.	More consumption of electricity.
Ozonation	No change in volume when applied in gaseous state.	short half time.
Fentons reagent	Effective removal for both hydrophobic and hydrophilic dyes.	Release of slime.
Activated carbon	Best removal of various types of dyes.	Very cheap.
Cucurbituril	Good adsorption for various dyes.	Very high expensive.
Membrane filtration	Universal removal for every dye	Formation of high concentration slime.
Ion exchange	Least loss of adsorbent.	Not effective for all types of dyes.

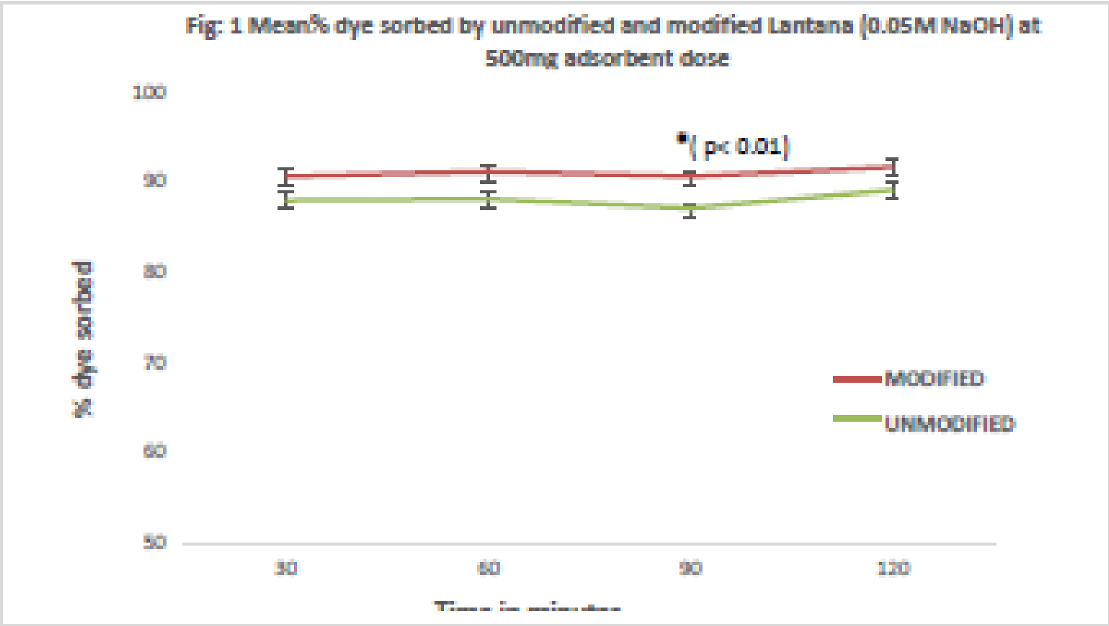
Fig: 1 Mean% dye sorbed by unmodified and modified Lantana (0.05M NaOH) at 500mg adsorbent dose
100
90
80
70
60
50
% dye sorbed
*(p< 0.01)
MODIFIED
UNMODIFIED
30
60
90
120

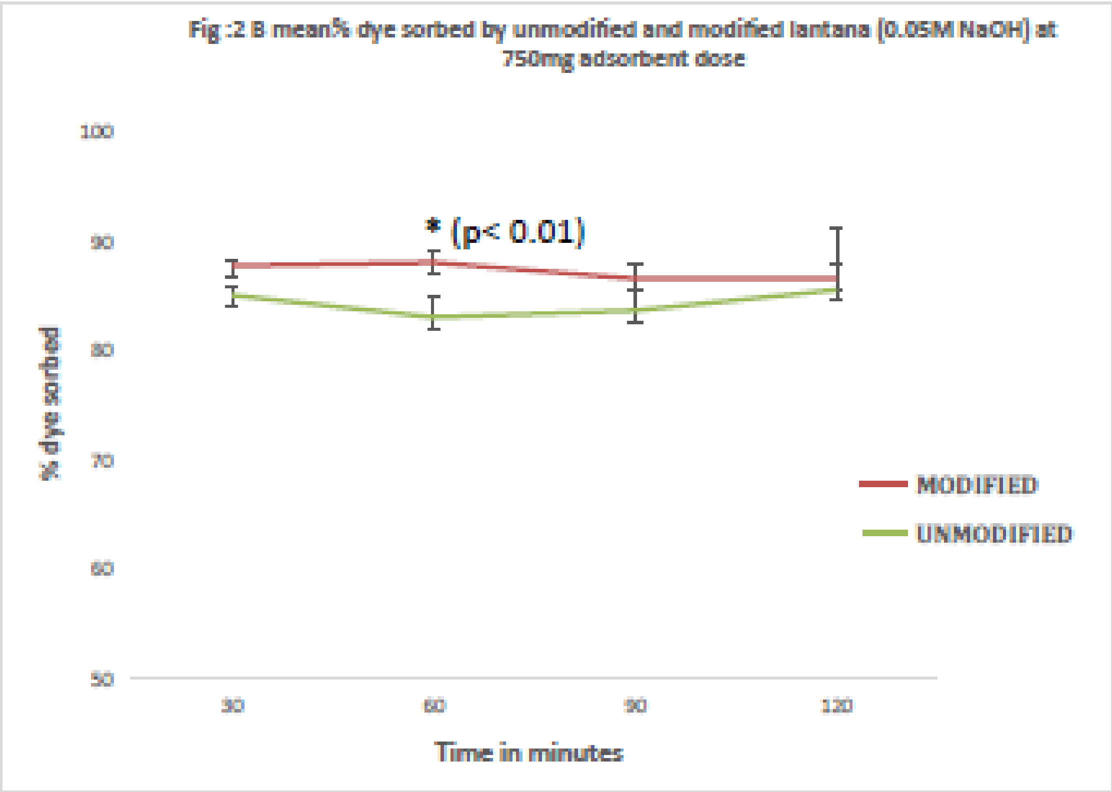
Fig :2 B mean% dye sorbed by unmodified and modified lantana (0.05M NaOH) at 750mg adsorbent dose
100
90
80
70
60
50
% dye sorbed
* (p< 0.01)
MODIFIED
UNMODIFIED
30
60
90
120
Time in minutes

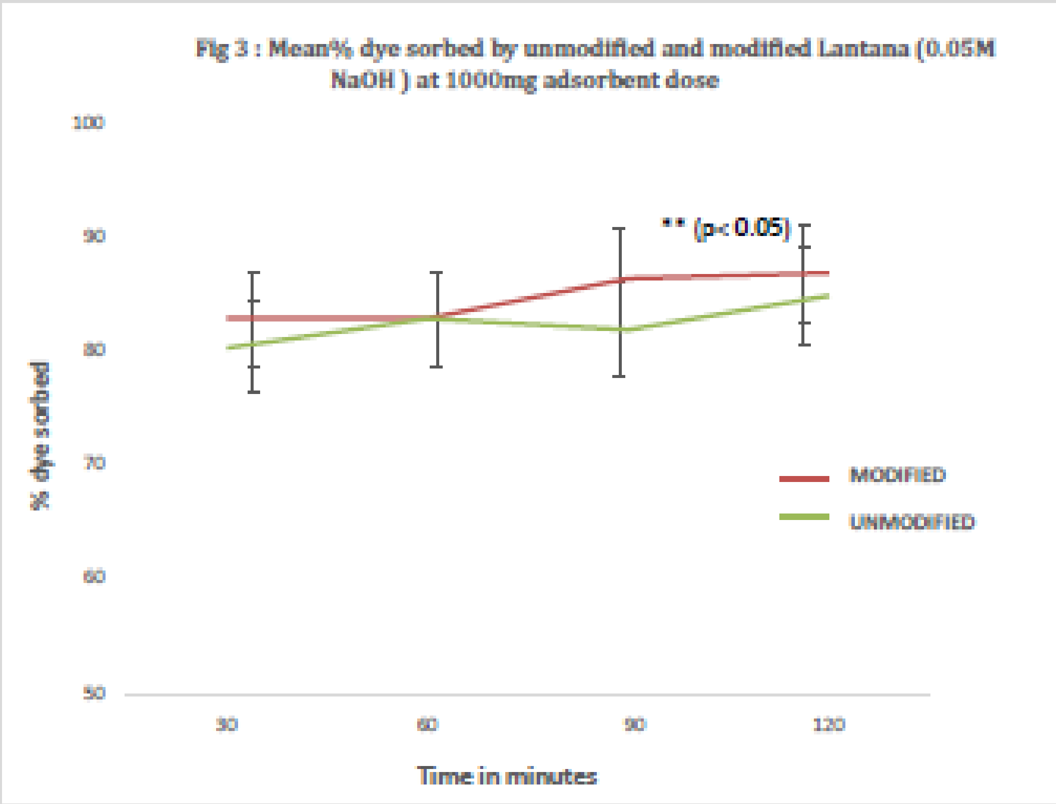
Fig 3 : Mean% dye sorbed by unmodified and modified Lantana (0.05M NaOH) at 1000mg adsorbent dose
100
90
80
70
60
50
% dye sorbed
** (p< 0.05)
MODIFIED
UNMODIFIED
30
60
90
120
Time in minutes

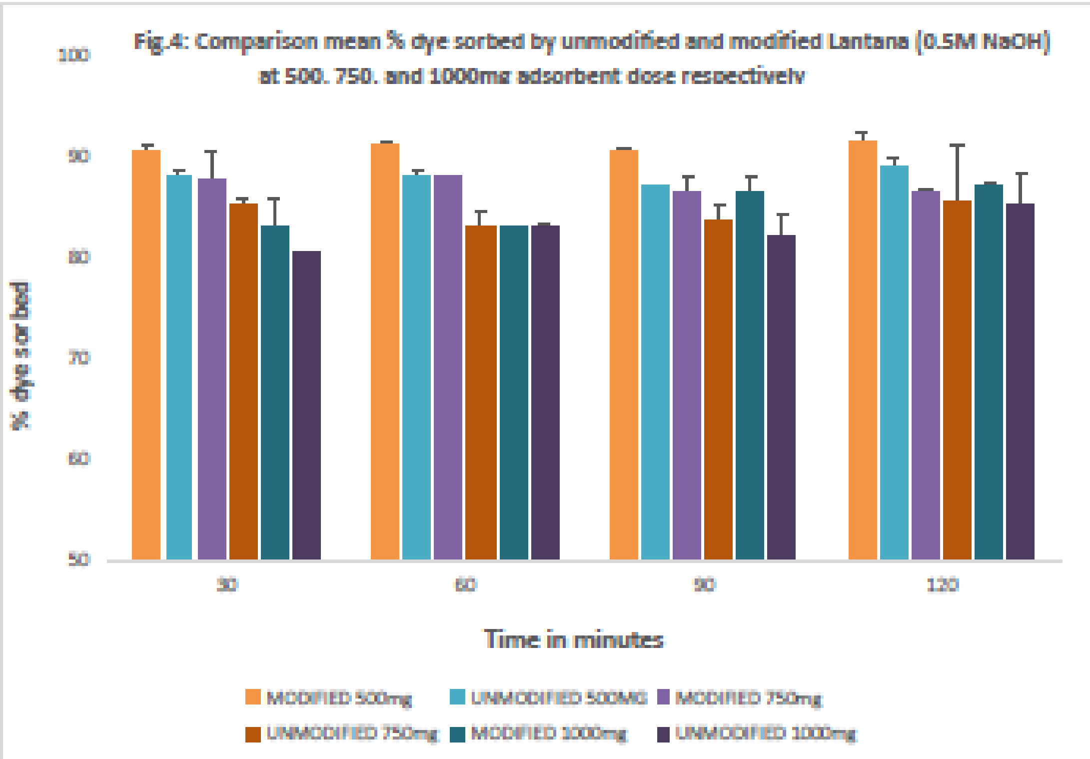
Fig.4: Comparison mean % dye sorbed by unmodified and modified Lantana (0.5M NaOH) at 500. 750. and 1000mg adsorbent dose respectively
100
90
80
70
60
50
% dye sorbed
30
60
90
120
Time in minutes
MODIFIED 500mg
UNMODIFIED 500MG
MODIFIED 750mg
UNMODIFIED 750mg
MODIFIED 1000mg
UNMODIFIED 1000mg

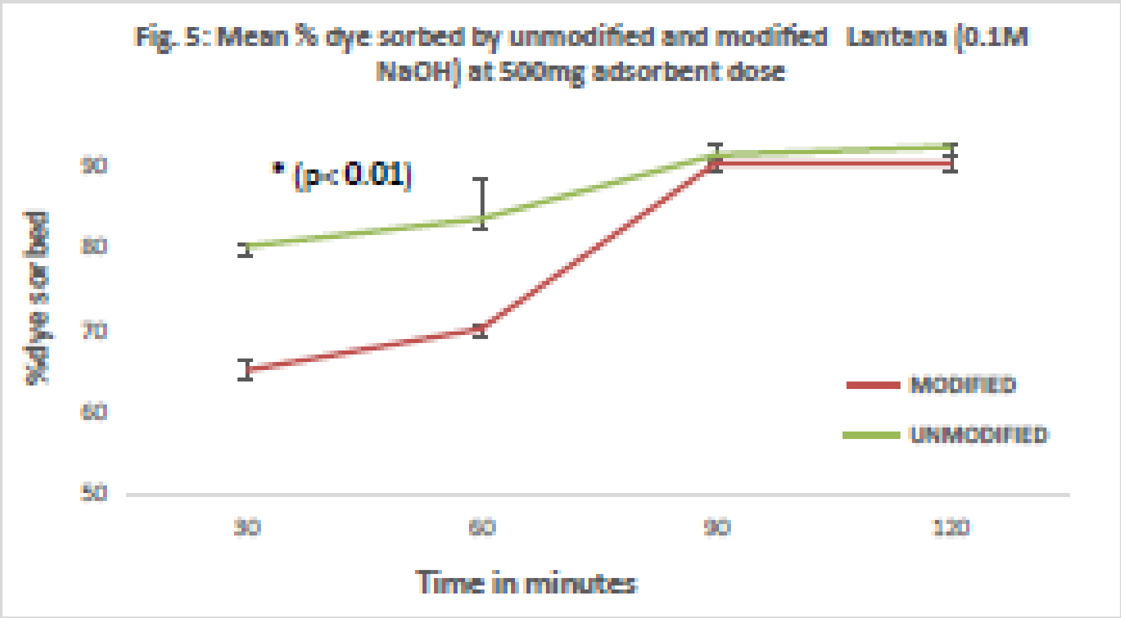
Fig. 5: Mean % dye sorbed by unmodified and modified Lantana (0.1M NaOH) at 500mg adsorbent dose
* (p< 0.01)
90
80
70
60
50
%dye sorbed
30
60
90
120
Time in minutes
MODIFIED
UNMODIFIED

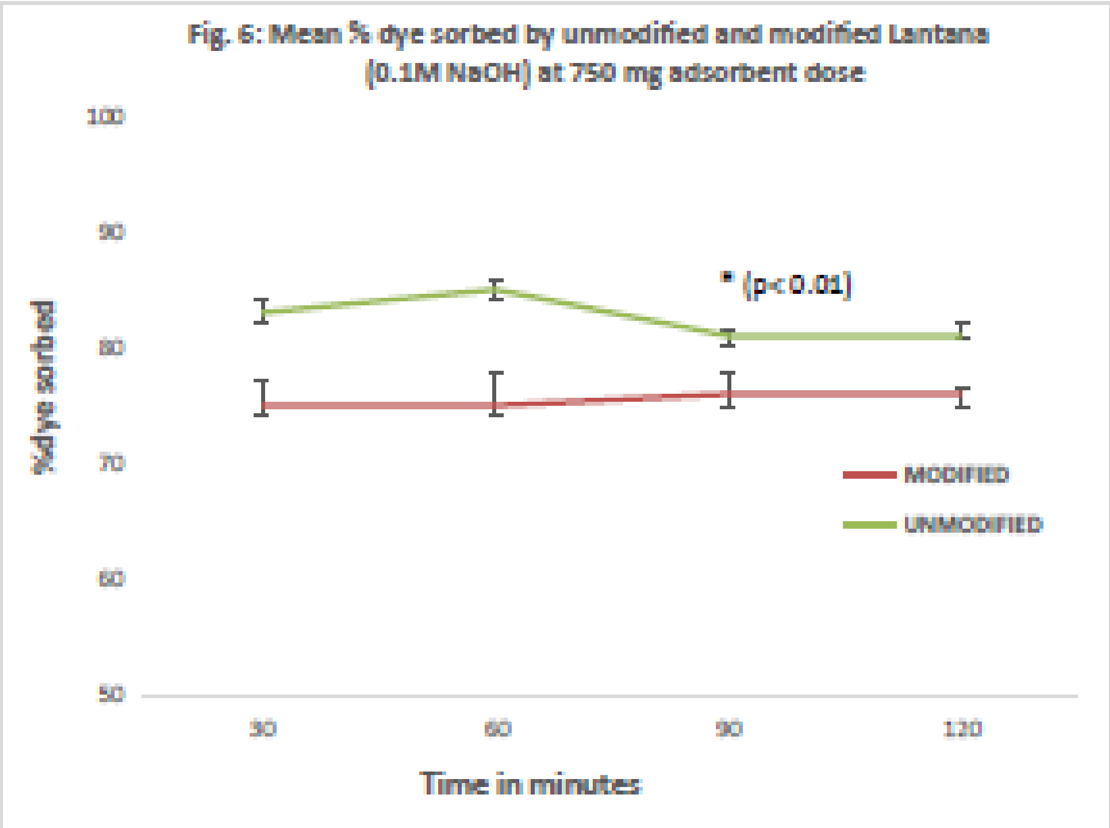
Fig. 6: Mean % dye sorbed by unmodified and modified Lantana (0.1M NaOH) at 750 mg adsorbent dose
* (p< 0.01)
100
90
80
70
60
50
%dye sorbed
30
60
90
120
Time in minutes
MODIFIED
UNMODIFIED

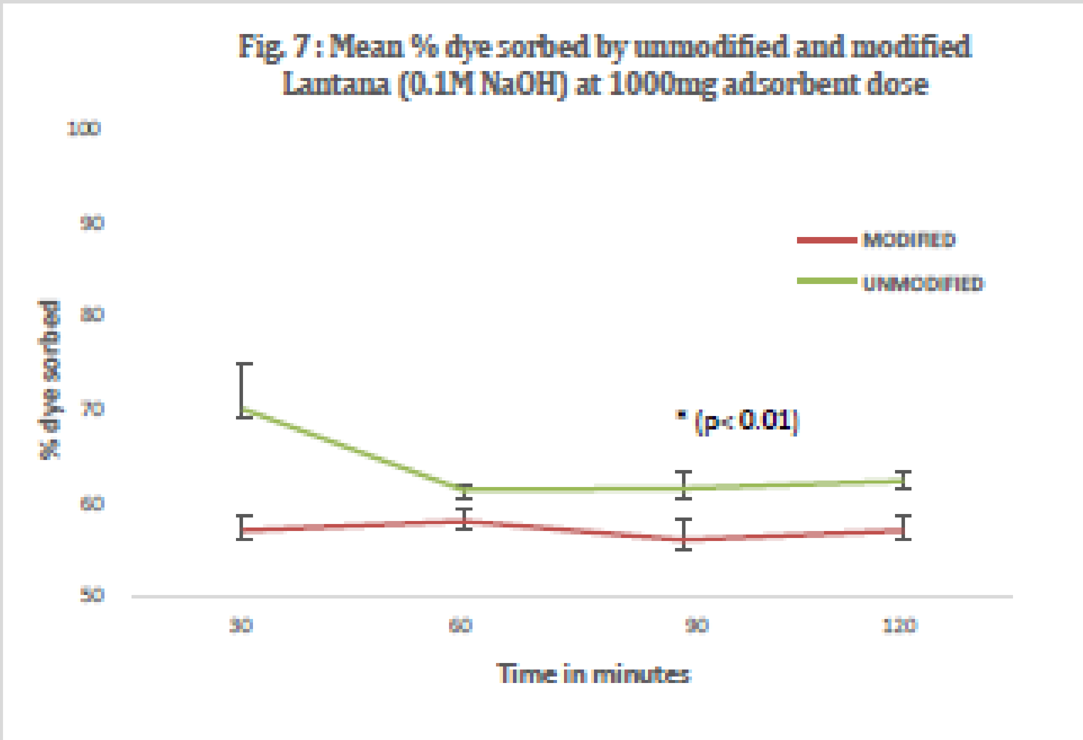
Fig. 7 : Mean % dye sorbed by unmodified and modified Lantana (0.1M NaOH) at 1000mg adsorbent dose
100
90
80
70
60
50
% dye sorbed
MODIFIED
UNMODIFIED
* (p< 0.01)
30
60
90
120
Time in minutes

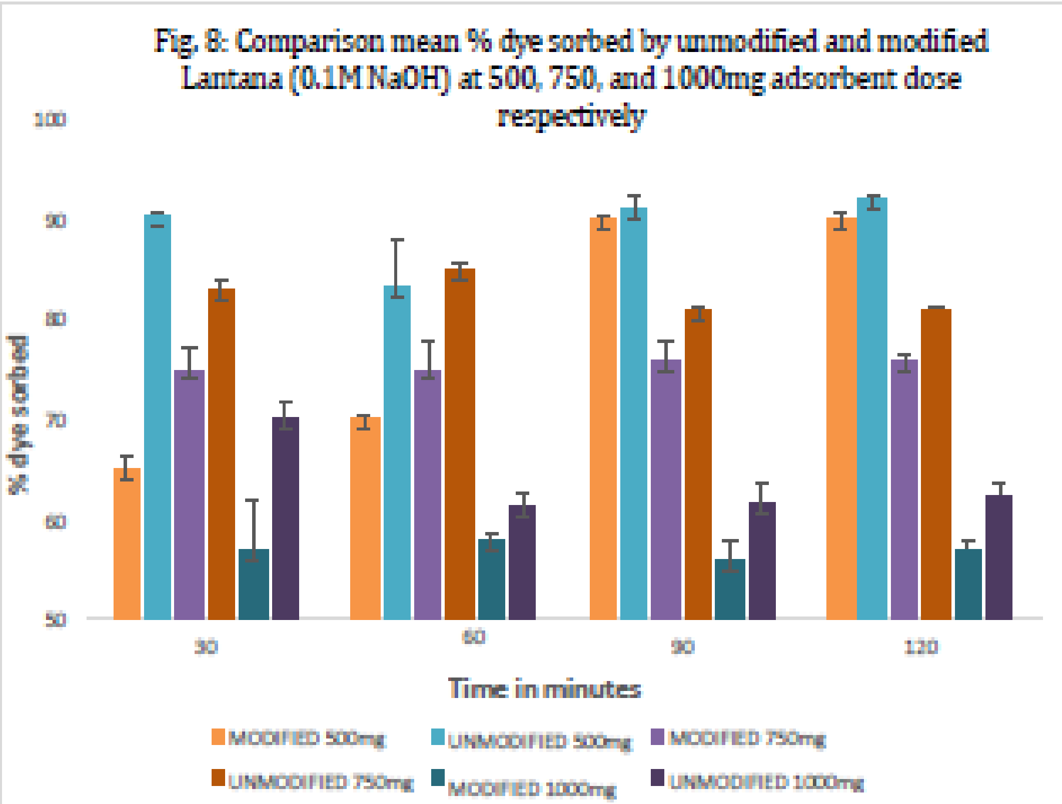
Fig. 8: Comparison mean % dye sorbed by unmodified and modified Lantana (0.1M NaOH) at 500, 750, and 1000mg adsorbent dose respectively
100
90
80
70
60
50
% dye sorbed
30
60
90
120
Time in minutes
MODIFIED 500mg
UNMODIFIED 500mg
MODIFIED 750mg
UNMODIFIED 750mg
MODIFIED 1000mg
UNMODIFIED 1000mg