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Adsorptive Removal of Six Dyes from Aqueous Solution using Normal and Modified Sawdust of *Gmelina arborea* Roxb

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Abstract
In the present study, the use of low-cost timber industry waste adsorbent normal sawdust of *Gmelina arborea* (NSGA) and alkali modified sawdust of *Gmelina arborea* (MSGA) have been reported as an alternative to the existing expensive methods of removing dyes from aqueous solution. Total six dyes *viz*., Malachite Green, Congo Red, Crystal Violet, Methylene Blue, Bromophenol Blue and Methyl Orange were removed from aqueous solution using NSGA and MSGA. Alkali and heat treatments were used to modify the sawdust to alter its adsorptive capacity. The NSGA removed 95.22 % crystal violet from the aqueous which was maximum removal by NSGA. The MSGA removed 91.6% malachite green which was highest among the six dyes used in the screening. The results indicated that most of the dyes were removed within 20-40 minutes. The efficacy of dye adsorption was improved in case of bromophenol blue, Congo red, methylene blue and malachite green after alkali and heat treatment to sawdust of *Gmelina arborea*. The result indicates that after optimization alkali modified *Gmelina arborea* sawdust can be used for the adsorptive removal of dyes from wastewater.

Keywords
Dye, adsorption, decolorization, sawdust, *Gmelina arborea* Roxb.

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INTRODUCTION
Water is most important for humans, plants, animals and every microorganism. Pure and easily available water is important for human health, environment, domestic usage, food production and in industry too. Total 10% of world's population survives under lack of drinking water requirement. Number of people and animals are forced to migrate in search of pure water. Up to 2025, majority of people will face major problem of potable water. In 2010, UN General Assembly announced the human right for water and sanitation, "Everyone has the right to sufficient,

continuous, safe, acceptable, physically accessible and affordable water for personal and domestic use" [1].

Textile industry is one of the largest sources for the water pollution. Textile industry discharges the large amount of pollutants and also produces noise pollution, air pollution, soil pollution and water pollution, too. Textile industry produce textile wastes by various type of processes like, bleaching, heat setting, desizing, dyeing, printing and finishing. The fact is that the textile contains a large amount of dyes and other hazards chemicals [2]. Dyes are used in large quantities in textile industry [3]. Their presence in water is highly visible and also undesirable [4]. Dyes also possess some kind of carcinogenic effects to the body, which also causes the skin irritation and other adverse effects on body. Colored waste water critically affects the aquatic life in the ecosystem [5]. To remove dyes, possible methods for the treatment of wastewater from textile industry are aerobic and anaerobic treatment, aerated lagoons, activated sludge, trickling filtration, oxidation ditch and ponds, microfiltration, nanofiltration, photocatalysis, electro-chemical process, ion exchange method, thermal evaporation and adsorption technique [6].

Among all this techniques, Adsorption is the most simplest, reliable and applied technique. Adsorption is the adhesion of atoms, ions and molecules from liquid to a surface. This process creates a film of the adsorbate on the surface of the adsorbent. According to IUPAC, adsorption is the increase in the concentration of a substance at the interface of a condensed and a liquid or gaseous layer owing to the operating of surface forces. Adsorption is mostly present in many systems like physical, chemical, biological and also used in many industrial applications. With the comparison of other dye removal treatments of aqueous solutions, adsorption technique is most preferable [4]. Low cost adsorbents are widely used for the adsorption method and generally agricultural/plant wastes are mostly used. A number of low cost non-conventional agricultural waste adsorbents are presence like, barley husk [7], wood apple [8], tamarind fruit shell [9], Bengal gram seed husk [10], waste potato peels [11], rice hulls [12], mosambi and cotton wastes [13], tea dust [14], Neem [15], groundnut [16] and *Tectona grandis* sawdust [17]. Sawdust is the most simple, easily available and low cost type of adsorbent. In the present study, we prepared the normal and alkali-heat treated (modified) sawdust of *Gmelina arborea* Roxb. to assess their capacity for the removal of six different dyes from the aqueous solution.

MATERIALS AND METHODS

Preparation of adsorbate (dye solution)

Different six dyes from different classes were used in the present study to screen the efficiency of the normal and alkali-modified sawdust of *Gmelina arborea* to decolorize these dyes. The six dyes used in the study were malachite green, congo red, crystal violet, methylene blue, bromophenol blue and methyl orange (SDFCL, India). The details of the dyes are given in the Table 1 and the structures of dyes are given in Figure 1. The dye (adsorbate) solutions were prepared with the concentration of 100 mg/L for individual dye in double distilled water.

Table 1: The particulars of six dyes used in the study

Sr. no.	Name of dye	Formula	Class of dye	Molecular weight (g/mol)	λmax (nm)
1	Malachite green	$C_{23}H_{25}N_2$	Triarylmethane dyes	364.91	617
2	Congo red	$C_{32}H_{22}N_6Na_2O_6S_2$	Azo dye	696.67	488
3	Crystal violet	$C_{25}H_{30}ClN_3$	Triarylmethane dyes	407.99	588
4	Methylene blue	$C_{16}H_{18}ClN_3S$	Thiazine dyes	319.85	665
5	Bromophenol blue	$C_{19}H_{10}Br_4O_5S$	Triarylmethane dyes	669.96	435
6	Methyl orange	$C_{14}H_{14}N_3NaO_3S$	Azo dye	327.33	468

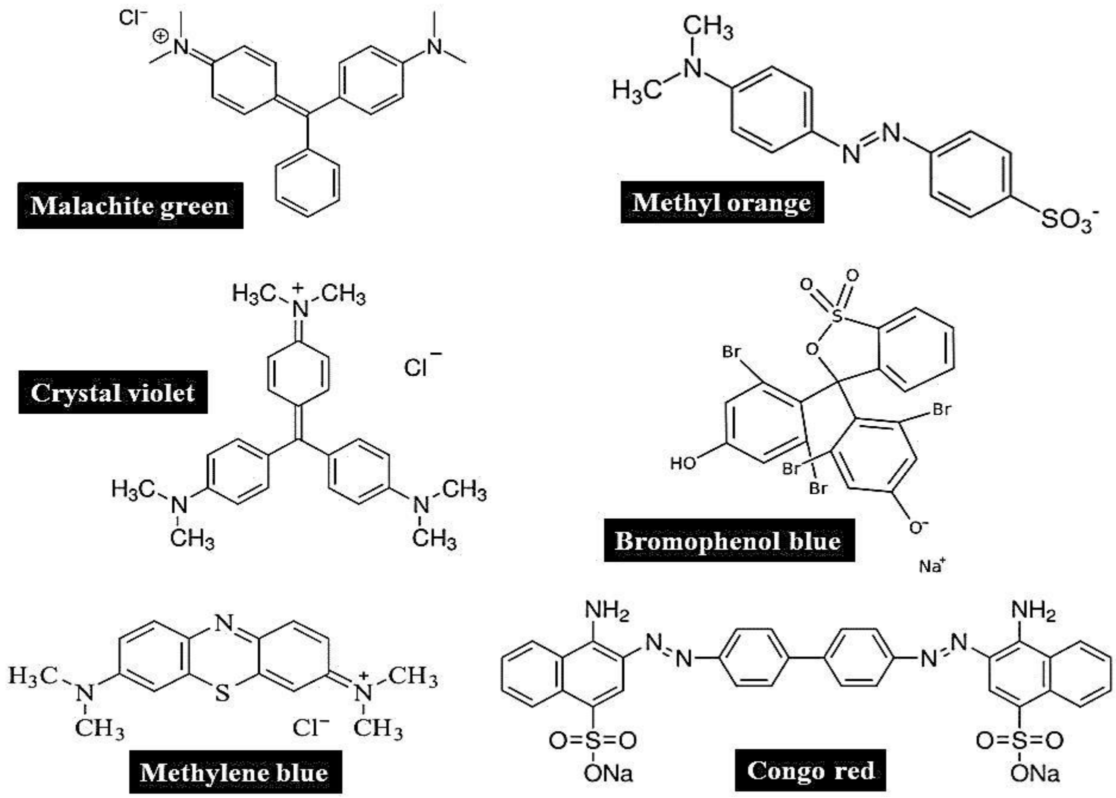

Figure 1: Structures of six dyes used in the study

Preparation of adsorbent (sawdust)

The sawdust of plant *Gmelina arborea* Roxb.**,** was collected from the local saw mill and sieved through a mesh of No. 60 which finally gave the powder of sawdust of 0.25 mm particle size. Then, it was washed with distilled water under shaking condition at 120 rpm for the removal of other dust and particles from its surface and dried at the room temperature of 25-30°C for 24hr. Next day, it was again washed with distilled water to remove remaining unwanted particles and again dried at the room temperature for 24hr. The dried powder of normal sawdust of *Gmelina arborea* (NSGA) was collected and stored in the airtight glass bottle at room temperature.

The modified sawdust was prepared from the same normal sawdust. First of all, it was collected and sieved through a mesh of No.60, to get the 0.25 mm particle sized powder followed by two times wash of distilled water, drying at room temperature. The dried sawdust was treated with 1% NaOH (alkali) under shaking condition at 120 rpm at 37°C for 24hrs followed by filtration using Whatman filter paper. The alkali treated sawdust was washed with distilled water for 2 hrs. Then it was heated at 80°C in a hot air oven for 5 hrs. Finally, the alkali treated and heat treated sawdust was considered as modified sawdust of *Gmelina arborea* (MSGA) which was collected and stored in the airtight glass bottle at room temperature.

Batch experiment for dye removal

For the batch mode study, 100 ml dye solution having 100 mg/L concentration was taken in dry and clean 250 ml flask. The initial absorbance/optical density (O.D.) value of the dye was measured at λmax value by using UV-Visible spectrophotometer. Then 0.3 gm NSGA and MSGA powder was added in to the separate flasks containing same dye sample. Flasks were agitated in incubator shaker at 120 rpm, 37°C temperature up to 120 min. During agitation period, 2-3 ml dye samples were collected; filtered and O.D. value was measured by UV-Visible spectrophotometer after every 20 min up to 120 min. The O.D. was measured at λmax of each dye.

This experiment was conducted for all the six dyes separately. The aqueous solution containing dye without adsorbent was considered as a control. The initial and final O.D. values achieved were used to calculate percent removal of the dye by following formula [4].

$$\% \text{ removal} = \frac{\text{Initial absorbance - Observed absorbance}}{\text{Initial absorbance}} \times 100$$

RESULTS AND DISCUSSION

In the present study, NSGA and MSGA were used for the screening of their dye removal efficiency using six different dyes from various classes. The experiment was carried out using 100 mg/L dye concentration. After 120 minutes, NSGA removed 95.22 % crystal violet from the aqueous which was maximum removal by NSGA. The lowest removal of dye was observed in case of bromophenol blue which was 4.76% using NSGA after 120 min. The MSGA was used for removal of six dyes and after 120 min MSGA removed 91.6% malachite green which was highest among the six dyes used in the screening. With 6.67% removal of methyl orange, it was the least removed dye by MSGA. Overall results of dye removal efficiency by NSGA and MSGA are given in Table 2.

The results of dye removal by NSGA and MSGA indicated that most of the dyes were removed within 20-40 minutes (Figure 2). Malachite green was removed slowly up to 120 min and 100 min by NSGA and MSGA, respectively. Dye decolorization screening indicated that except malachite green, all the dyes were mostly removed within 40 min. After 40 min, dye removal process became almost steady. It suggests that most of the dye binding sites might have been occupied by the dye particles within 40 min.

Dye removal from aqueous solution was achieved by adsorption mechanism by using normal and modified adsorbent. NSGA might have various functional groups which might be responsible for the binding of the dye particles on the surface of the adsorbent. After the modification of adsorbent, the functional groups on the surface of MSGA might have been changed which ultimately lead to the variation in the dye binding capacity of the adsorbent. Crystal violet and methyl orange were less adsorbed by the MSGA as compared to NSGA which might be due to variation in functional groups after alkali and heat treatment to NSGA decreased dye removal efficiency. However, the efficiency of dye adsorption was increased in case of bromophenol blue, Congo red, methylene blue and malachite green after alkali and heat treatment. The comparison of per cent dye removal by NSGA and MSGA after 120 min is shown in figure 3. The photographs of results for removal of six dyes by NSGA and MSGA are shown in figure 4.

There are some reports on the dye removal using sawdust from different plants but as per our knowledge this might be the first and primitive research on the dye removal efficiency of normal and modified sawdust of *Gmelina arborea.* The present study was preliminary study for the assessment of the dye removal efficiency of normal and modified sawdust of *Gmelina arborea.* For the practical implementation of this adsorbent, all the process parameters need to be optimized and the exact mechanism for the removal of various dyes should be studied. But the present study will help in selection of the adsorbent and methods to be used for the modification of adsorbent. It will also provide primary information of the removal efficiency of NSGA and MSGA of six different dyes. Few past reports on dye removal capacity of sawdust are there in literature which includes methylene blue removal from simulated wastewater by adsorption using Indian rosewood sawdust [18], removal of direct red 23 by wood sawdust [19], sagaun sawdust for the removal of crystal violet dye from simulated wastewater [20], timber sawdust and alkaline treated sawdust to methylene blue and methyl green [21] and poplar sawdust for removal of disperse orange 30 dye [22]. It has been reported that alkaline treatment increases dye removal efficiency of sawdust for some dye which is in agreement with past report [21]. The modification of sawdust to enhance dye removal efficiency depends on the chemical used in modification, type of dye, type of sawdust and few other parameters. Hence, the present investigation will help in the use of *Gmelina arborea* as an adsorbent for the removal of dyes from wastewater.

Table 2: Per cent removal of six dyes by normal and modified sawdust of *Gmelina arborea*

Adsorbent	Dyes	Time (min)						
		0	**20**	**40**	**60**	**80**	**100**	**120**
NSGA	CV	0.00	90.43	94.35	94.78	95.22	95.22	95.22
	MO	0.00	06.67	26.67	26.67	26.67	26.67	26.67
	BPB	0.00	04.76	04.76	04.76	04.76	04.76	04.76
	CR	0.00	61.11	66.67	66.67	66.67	72.22	72.22
	MB	0.00	40.00	44.00	44.00	44.00	44.00	44.00
	MG	0.00	14.71	19.75	28.57	42.44	63.87	75.21
MSGA	CV	0.00	78.26	78.26	82.61	86.96	91.30	91.30
	MO	0.00	06.67	06.67	06.67	06.67	06.67	06.67
	BPB	0.00	04.76	09.50	09.50	09.50	09.50	09.50
	CR	0.00	77.78	83.33	88.89	88.89	88.89	88.89
	MB	0.00	36.00	36.00	40.00	44.00	48.00	48.00
	MG	0.00	36.97	62.18	83.19	87.39	91.60	91.60

Value in table indicate % removal of dyes; NSGA: Normal Sawdust of *Gmelina arborea* **MSGA:** Modified Sawdust of *Gmelina arborea*; **CV:** Crystal Violet; **MO:** Methyl Orange; **BPB:** Bromophenol Blue; **CR:** Congo Red; **MB:** Methylene Blue; **MG:** Malachite Green

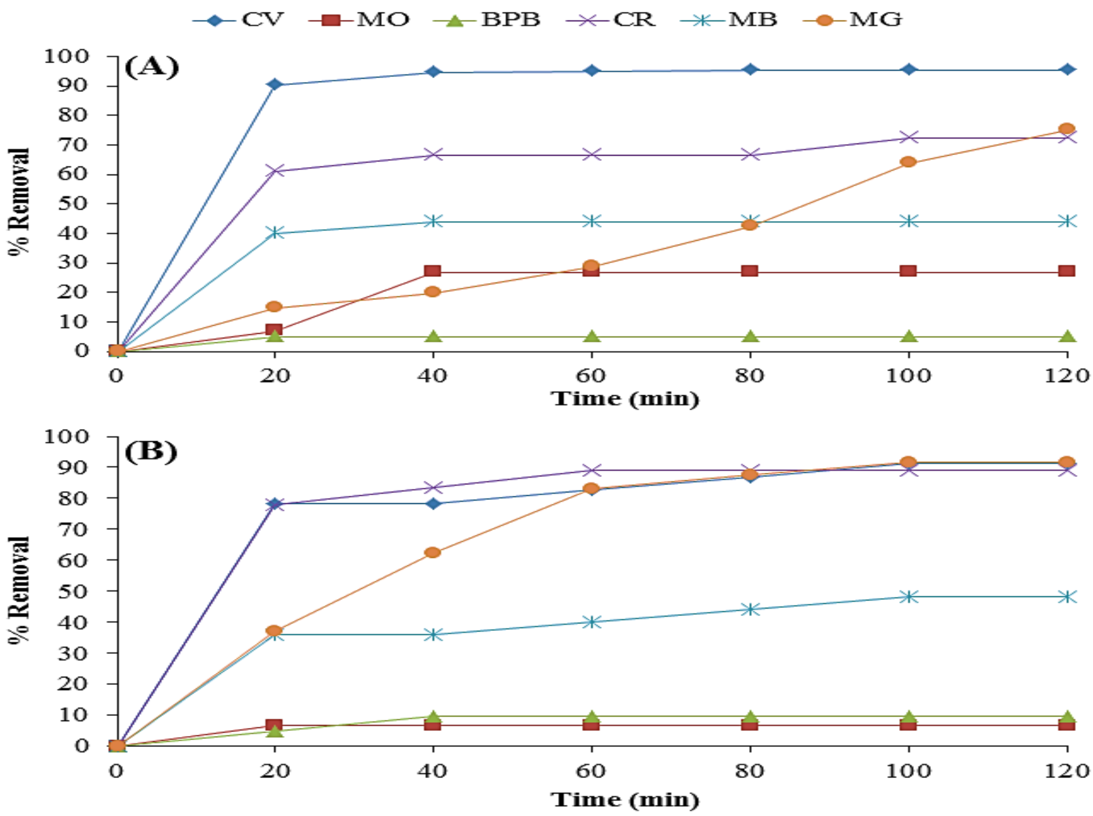

Figure 2: Per cent removal of six dyes by (A) NSGA and (B) MSGA. (CV: Crystal Violet; MO: Methyl Orange; BPB: Bromophenol Blue; CR: Congo Red; MB: Methylene Blue; MG: Malachite Green)

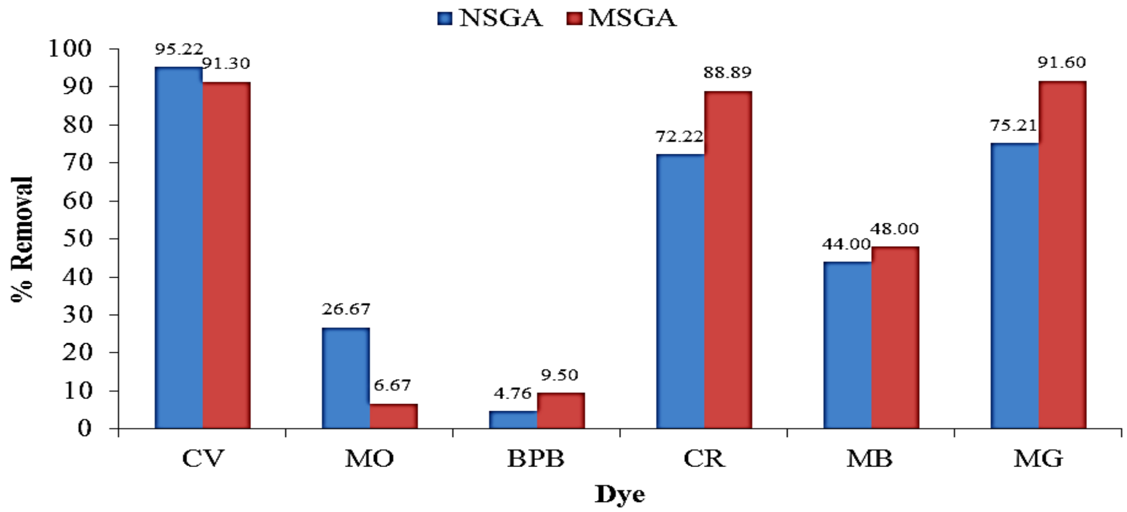

Figure 3: Comparison of % removal of six dyes by NSGA and MSGA after 120 min. (CV: Crystal Violet; MO: Methyl Orange; BPB: Bromophenol Blue; CR: Congo Red; MB: Methylene Blue; MG: Malachite Green)

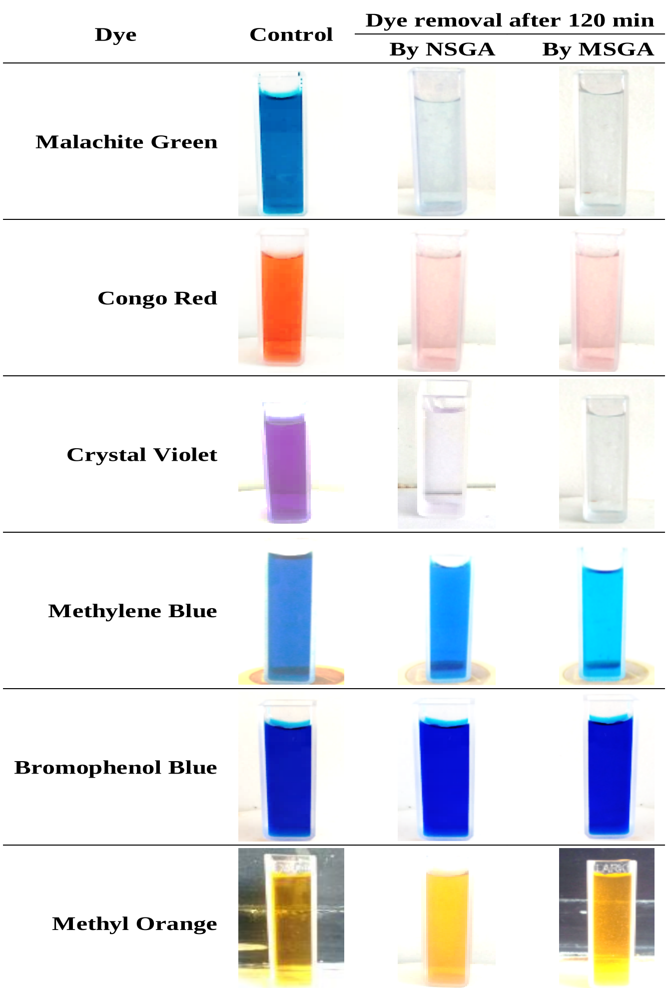

Figure 4: Removal of six dyes by NSGA and MSGA after 120 min.

CONCLUSION

The present study was pilot study for the elucidation of the dye removal efficacy of normal and modified sawdust of *Gmelina arborea.* The NSGA removed 95.22 % crystal violet from the aqueous which was maximum removal by NSGA. The MSGA removed 91.6% malachite green which was highest among the six dyes used in the screening. The results indicated that most of the dyes were removed within 20-40 minutes. The efficiency of dye adsorption was increased in case of bromophenol blue, Congo red, methylene blue and malachite green after alkali and heat treatment to *Gmelina arborea* sawdust. It indicates that after optimization alkali modified *Gmelina arborea* sawdust can be used for the removal of dyes from wastewater.

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