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Evaluation of methyl red removal potential of adsorbents made from Leucaena leucocephala leaf and bark materials

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A B S T R A C T

The aim of this study was the investigation of how adsorbents made from *Leucaena leucocephala* (L. Leu) leaf and bark materials were effective in treating wastewater containing methyl red dye (MeReD). The functional groups in the phytochemical elements from the bark and leaves of L.Leu offer active surface sites and enable the adsorption mechanism. The investigations were done in batch mode. Contact time, adsorbent dose concentration, temperature, pH, and initial MeReD concentration were all examined as significant parameters impacting the sorption process. The study found that significant MeReD removal efficiency was achieved after 90 min of contact time, pH 4.0 units, 1.2 gm/L adsorbent dose concentration, 100 ppm initial MeReD quantity, and 30 ^oC temperature. The developed adsorbents were applied to samples gathered from dye companies in Hyderabad and Guntur. The extraction of MeReD (81.2% to 90.0%) from untreated wastewater contaminated with MeReD using developed adsorbents was very efficient.

1. Introduction

During the functional process activities, the textile industry uses a huge quantity of water as well as chemicals. As a result, a large amount of effluent including acids, solvents, bleaching agents, dyes, and other chemicals is typically generated, contributing to high levels of biological & chemical oxygen demands, turbidity, dyes and hazardous compounds.[1,2] Textile effluent is frequently released straight into rivers in underdeveloped nations without any further processing. This activity has a major negative impact on the environment, as it may cause the loss of aquatic species and inflict poor health effects in humans such as dermatitis, immune system repression and allergies.[3]

Amongst several dyes, methyl red dye (MeReD) is probably of the most widely used in laboratories as an acid base marker.[4] The MeReD (anionic azo dye), is hazardous, carcinogenic or mutagenic contaminant in water bodies. The presence of methyl red dye in water can cause digestive problems as well as eye and skin irritation.[5,6] As a result, before dumping such dye-containing effluent into aquatic habitats, it must be treated. To treat textile effluent, a number of new methods are presently being explored specifically, adsorption,[7] advanced oxidation procedures,[8] biological treatment,[9] membrane technology,[10] chemical coagulation technology and flocculation technology.[11]

Adsorption is the most frequently used approach due to its ease of use and cheaper cost when compared to other approaches. The generation of adsorbents from distinct biomass further contributes to the process's cost effectiveness.[12] Adsorption is a mass transference process that occurs when molecules move from a liquid to a solid state.[13]

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The IUCN Species Survival Commission's Invasive Species Specialist Group considers *Leucaena leucocephala* to be one of the top 100 most intrusive species.[14] L.leu is a type of blossoming plant has a place with Fabaceae family, plantae realm and is a typical no man's land weed.[15] L.leu has perceived restorative qualities and is generally utilized in ayurveda in the treatment of infection, ulcers, asthma, and growths.[16] The L.leu seeds have the ability to control stomach hurt, as contraception and abortifacient.

The plant materials (leaf and bark) of L.leu were employed as adsorbents in the absorption of the MeReD in polluted waters for the first time in this investigation. The object of the current work is to test the plant materials (leaf and bark) of L.leu for their sorption characteristics towards MeReD from contaminated waters. The effects of pH, dye (methyl red) concentration, adsorbent dosage, contact duration, and agitation on the absorption of the methyl red dye by plant materials (leaf and bark) of L.leu were investigated in this study.

2. Results and discussion

The capacity of sorbents generated from L. Leu plant material (bark and leaf) to extract MeReD was assessed in the proposed investigation. Surface examinations are required to understand the sorption science using current instruments such as Fourier Transform Infrared Spectroscopy (FT-IR), X-beam Photo Electron Spectroscopy (XPS), Energy Dispersive Spectrum (EDS), and Scanning Electron Microscope (SEM) methods, in addition to the traditional basic compound investigation prior to and after the sorption of t. The observations, on the other hand, can be explained as follows: The -OH/COOH groups in bio-sorbents generated from leaves and barks are dissociated by pH, which results in limited anion exchange capacity at low pH levels, according to the equilibrations:

Alkaline pH: Adsorbent-OH ↔ H^+ + Adsorbent-O^-

Adsorbent-COOH ↔ H^+ + Adsorbent-COO^-

Acidic pH: Adsorbent-OH + H^+ ↔ Adsorbent-O H H

The existence of distinct phytochemical elements in the bark and leaves of L.Leu was reported by Zayed et al.,[17] Umaru et al.,[18] and Mohamed.[19] The major phytochemicals included 1,2-benzenedicarboxylic acid, betulin, lupeol, androstan-17-one,3-ethyl-3-hydroxy-(5à), betamethasone, β-sitosterol, diisooctyl ester, β-sitosterol, 9,12-octadecadienoic acid, 9,12,15-octadecatrienoic acid, methyl ester, 1,2-benzenedicarboxylic acid, mono(2-ethylhexyl) ester, campesterol, stigmasterol, squalene, phytol, 3,7,11,15-tetramethyl-2-hexadecen-1-ol, 3,7,11-tridecatrienenitrile, 4,8,12-trimethyl, neophytadiene, 1-octadecyne, octadecane, and hexacosane. The functional groups (i.e., carboxyl, hydroxyl, carbonyl, amino, nitro etc.,) in these phytochemical elements from the bark and leaves of L.Leu can offer active surface sites for dye interaction and enable the adsorption mechanism.[20]

2.1. Adsorption of MeReD as a function of contact time

The significant adsorption of MeReD happens during the first 90 min, according to **Fig. 1** (**Graph No:A-1, A-2, A-3 and A-4**) and the subsequent rise in adsorption capacity is significantly slower. In 90 min, the equilibrium time period was established. The accessibility of a high number of uncovered surface-active sites on leaf and bark materials of L.leu for MeReD adsorption causes a rapid rise in adsorption efficiency of leaf and bark materials of L.leu at the start of the process. With increased contact duration, the accessible active sites progressively fill up, resulting in a modest rise in adsorption capacity. It's reasonable to presume that MeReD diffusion happens in the pores of the adsorbent at this point.[21,22] It is possible to create a MeReD monolayer on the adsorbent surface, as evidenced by the slightly lower adsorption rate.[23]

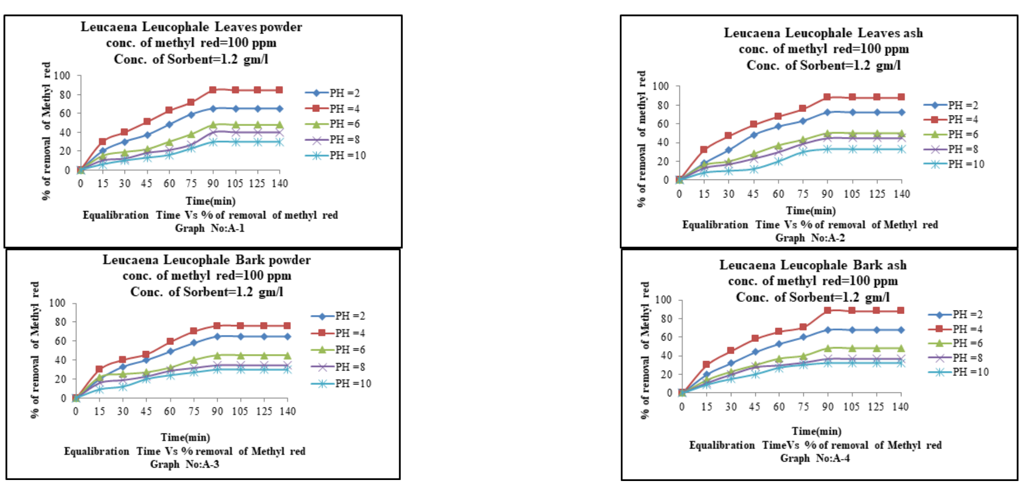

Fig. 1. Graphical representation of equilibration time Vs % removal of MeReD

According to check in data, contact times shown by different adsorbents were: 180 min by egg shell powder,[24] 3 to 4 hrs by xanathanated sal and charred saw-dust,[25] 180 min by bentonite & chitosan composite,[26] 60 min by orange peels,[27] and 90 min by oil shale.[28]

2.2. Adsorption of MeReD as a function of pH

To establish the optimal pH for MeReD sorption by leaf and bark materials of L.leu, comparative studies were conducted across a pH range 2.0 units -10.0 units. The sorption of MeReD by leaf and bark materials of L.leu increased from the starting pH of 2.0 units to pH 4.0 units, then declined across the pH range of 6.0 units – 10.0 units, as indicated in **Table 1**. As an outcome, pH 4.0 units was established as the best pH for all subsequent research.

Table. 1. The % removal of MeReD by leaf and bark materials of L.leu at different pH values

pH	% Removal of MeReD			
	Leaf ash	Bark ash	Leaf powder	Bark powder
0	0	0	0	0
2	72	68	65	65
4	88	88	85	76
6	50	48	48	45
8	45	37	40	35
10	33	32	30	30

The effectiveness of sorption is reliant on the solution because changes in solution pH direct to alternations in ionization degree and also the surface characteristics of the sorbent.[29] The sorbent's surface will be significantly protonated at low pH. The positive (H^+) ions create an electrostatic contact between the sorbent's surface and the MeReD molecules, resulting in maximal adsorption. However, at pH levels over 4.0 units (i.e., in the pH band of 6.0 units – 10.0 units), the proportion of protonation of the sorbent's surfaces is reduced, resulting in a decline in diffusion rate and adsorption rate owing to electrostatic repulsion.[30] Furthermore, the reduced MeReD adsorption in alkaline media might be related to competing for adsorption sites among the anionic dye units and excessive hydroxide ions (OH^-).

The current findings (**Table 1**) are consistent with former studies that report on the published findings for % removal of MeReD by xanathanated sal and charred saw-dust,[25] *Annona Squmosa* seed, [31] and *Erythrina indica* based adsorbents (leaf powder, bark powder, leaf ash and bark ash),[32] *Hyacinth* and *Tinospora Cordifolia* based leaf powder, bark powder, leaf ash and bark ash.[33]

2.3. Adsorption of MeReD as a function of sorbent concentration dose

The proportion of MeReD removed improved as the adsorbent dosage (leaf and bark materials of L.leu) was raised from 0.2 to 1.2 gm/L, then stayed steady after that. **Table 2** shows that the adsorbent dose rises in direct proportion to the number of adsorbent sites accessible.

The quantity of MeReD uptake rises as the number of adsorbent sites as well as the surface area of interaction with the MeReD increases, resulting in greater adsorption.[34] This phenomenon can be attributable to a raise in sorptive surface region as well as the availability of additional adsorption sites.[35]

Table. 2. The % removal of MeReD by leaf and bark materials of L.leu at dose concentrations

Dose concentration (gm/L)	% Removal of MeReD by			
	Leaf ash	Bark ash	Leaf powder	Bark powder
0	0	0	0	0
0.2	32	30	30	30
0.4	47	45	40	40
0.6	59	58	51	46
0.8	68	66	63	59
1.0	76	71	72	70
1.2	88	88	85	76
1.4	88	88	85	76

According to check in data, dose concentrations shown by different adsorbents were: 8 gm/ 300 ml by egg shell powder,[24] 25 mg/100 ml by xanathanated sal and charred saw-dust,[25] 0.7 gm/25 ml by bentonite & chitosan composite,[26] 3 gm/100 ml by orange peels,[27] 1.4 gm/l by *Erythrina indica* based adsorbents,[32] and 0.2 gm/100ml by banana pseudostem fibers.[36]

2.4. Adsorption of MeReD as a function of MeReD concentration

According to the findings (**Table 3**), increasing the initial MeReD concentration from 100 ppm to 250 ppm lessens the percent of MeReD cleared. The decreasing trend of the evidence suggests that as the quantity of MeReD in aqueous medium increases, the proportion of MeReD eliminated by the adsorbent (leaf and bark materials of L.leu) decreases.

This is because there were many sorption sites in the adsorbent (leaf and bark materials of L.leu) structure for low MeReD/adsorbent ratios, however as the ratio rises, the sorption spots become saturated, resulting in a drop in sorption efficacy.[37] The gradient among the sample solution and the particle's centre promotes MeReD diffusion via the film encircling the particle and the porous networks of adsorbent (leaf and bark materials of L.leu) at high starting MeReD concentrations.[38]

Table. 3. The % removal of MeReD by leaf and bark materials of L.leu at different MeReD concentrations

MeReD concentration (ppm)	% Removal of MeReD by			
	Leaf ash	Bark ash	Leaf powder	Bark powder
100	89	86	88	79
150	82	81	75	75
200	75	76	71	65
250	64	71	69	61

The same was also observed in % removal of MeReD on absorbents prepared from *Erythrina indica* based adsorbents[32] and oil shale.[39]

2.5. Effect of interfering ions

The proportion of MeReD removed in the presence of a five-fold surplus of common ions present in normal waters, such as anions like sulphate, carbonate, chloride, and phosphate, as well as cations like calcium (II), ferrous (II), magnesium (II), and zinc (II) was assessed. **Table 4** summarises the findings. At the optimal conditions of contact equilibration time, pH, MeReD concentration and adsorbent concentration, anions had only a little effect on the rate extractability of MeReD with the adsorbents (leaf and bark materials of L.leu) used in this study. Cations like ferrous (II) and calcium (II) have not messed with the maximum percent of MeReD extraction and have synergistically kept up with it, however cations like magnesium (II), and zinc (II) have messed with it to a lesser extent. The findings are consistent with those of Ramana et al.[33]

Table. 4. Effect of co-ions on the MeReD extractability with leaf and bark materials of L.leu

Adsorbent	MeReD extractability (%) in the presence of a fivefold surplus of interfering ions (500 ppm) under optimal experimental settings*							
	SO_4^{2-}	PO_4^{3-}	CO_3^{2-}	Cl^-	Ca^{2+}	Mg^{2+}	Fe^{2+}	Zn^{2+}
Leaves powder	96.0	98.5	100.0	95.9	99.4	99.5	98.1	88.9
Leaves ashes	98.2	97.6	100.0	99.3	99.5	94.9	100.0	91.9
Bark powder	94.7	93.8	96.5	87.9	95.6	96.8	98.5	97.6
Bark ashes	92.9	96.6	99.1	94.2	96.4	89.7	91.8	92.5

* Optimal conditions: pH – 4: Agitation time - 90 min: Sorbent Conc. - 1.2 gm/L

2.6. Adsorption of MeReD as a function of temperature

In both scientific as well as practical applications, investigation into the impact of reaction temperature is essential and recommended. Temperature variation is a realistic limitation that can impact the adsorption process in either a positive or negative way. The studies on adsorption of MeReD as a function of temperature with leaf and bark materials of L.leu were carried out at temperatures of 30, 40, 50, 60, and 70 degrees Celsius. **Table 5** shows the experimental results, which show that the MeReD removal process is thorough and unaffected by temperature changes, demonstrating significant chemisorption of MeReD molecules on the adsorbent (leaf and bark materials of L.leu) surface.

Table. 5. The percent removal of MeReD by leaf and bark materials of L.leu at different temperatures

Temperatures (^oC)	% Removal of MeReD by			
	Leaf ash	Bark ash	Leaf powder	Bark powder
30	89.12	86.56	88.79	79.22
40	89.21	86.45	88.75	79.12
50	89.45	86.56	88.73	79.23
60	89.10	86.52	88.65	79.25
70	89.11	86.57	88.71	79.13

Different researchers reported a procedure temperature of 25 °C for highest MeReD removal by egg shell powder[24] as adsorbent, xanathanated sal and charred saw-dust[25] as adsorbent, bentonite & chitosan composite[26] as adsorbent, orange peels[27] as adsorbent and banana pseudostem fibers[36] as adsorbent. When utilizing Annona Squmosa seed, Krishna et al. detected that ambient temperature provided the highest MeReD removal.[31] Chukka et al. and reported a procedure temperature of 27 °C using *Erythrina indica* based materials as adsorbents.[32] Using oil shale, Noha et al. mentioned a process temperature of 30 °C for highest MeReD removal.[39]

2.7. Applications of the developed biosorbents

Numerous researchers generated organic chemical compounds, then examined their chemical architectures using spectroscopic investigations, and studied their performance for biological processes including insecticidal activity, metal ion clearance activity, hydrocarbon disposal performance, and antibacterial behavior.[40-46]

In this study, the acceptability of biosorbent systems established using the novel adsorbent obtained from L. Leu plant materials for the extraction of MeReD from wastewaters was tested using real effluent collections from a few industries. At the optimal conditions of MeReD extraction obtained, the collected samples were treated to MeReD removal using the adsorbents (leaf and bark materials of L.leu) established as part of this investigation. The outcomes got were in **Table 6**.

Table. 6. Percent MeReD extractability from collected effluents using leaf and bark materials of L.leu

Bio-sorbents	Percent extraction of MeReD in sample fed with						
	10.0 ppm MeReD	15.0 ppm MeReD	20.0 ppm MeReD	25.0 ppm MeReD	30.0 ppm MeReD	35.0 ppm MeReD	Average
Leaves powder	81.5	88.3	82.3	81.2	85.5	86.3	84.1
Bark powder	88.1	82.2	85.6	88.3	81.5	83.5	84.86
Leaves ashes	86.3	81.5	85.5	88.4	89.0	85.2	85.98
Bark ash	86.5	90.0	89.0	85.2	83.5	86.5	86.78

3. Conclusion

The leaves and bark materials of the L.leu plant was utilised to produce efficient adsorbents. These established adsorbents were applied for the removal of azo dye, MeReD. The batch technique was used to investigate the adsorption of MeReD on these developed adsorbents as a function of contact duration, MeReD concentration, pH, co-ions, and temperature. The developed adsorbents were exploited successfully to remove MeReD, under optimal extraction conditions established, from actual test effluent samples gathered from dye companies in Hyderabad and Guntur. The findings suggested that established adsorbents could be used in the efficient removal of MeReD from waste aqueous solutions at a low cost.

If this article has been published, it will be referred by many other researchers which may enhance the application of natural techniques and may be a platform to develop a new technique in a natural way and hence supporting elimination of dye pollution in an eco-friendly manner. This (our article) might be a small initiative from our side with the support of learned reviewers and esteemed journal.

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4. Experimental

4.1. Plant materials

L.leu plant was collected in Ramanagar, at Bapatla town, Guntur District, Andhra Pradesh and was authenticated by the taxonomist at Acharya Nagarjuna University, Guntur. L.leu leaves and barks were freshly plucked from the plants, cleaned with water, then again cleaned using distilled water, and finally sun dried for nearly 48 hr. This dried plant materials (leaf and bark) of L.leu was powdered using a mechanical grinder to a fine cross section with size nearly > 75 microns. The L.leu powdered dry plant materials (leaf and bark) were activated in an air oven set at 105 ^{o}C for 5 hr. Dried plant materials (leaf and bark) were burned in a muffle electric furnace for one hour to obtain L.leu leaf and bark ash. The ash and powder samples of L.leu plant materials were then utilized in this investigation to test their sorption characteristics towards methyl red dye from contaminated waters.

4.2. Methyl red dye solution

One gram of MeReD was mixed with one litre of deionized water to make a 1000 ppm stock MeReD solution. Further necessary concentrations of MeReD solutions were made by diluting the stock MeReD solution appropriately.

4.3. Batch adsorption experiments

While investigating L.leu plant material for its ability to bind MeReD, it was observed that adsorbents originated from the leaves and barks of L.leu showed affinity for the MeReD. Dilution of the stock MeReD solution yielded aqueous solutions with varied MeReD concentrations (100, 150, 200, and 250 ppm). In a 1000 mL round bottom flask, a batch mode adsorption investigation of 500 mL MeReD solution with adsorbents (leaf and bark materials of L.leu) was carried out and thoroughly churned at 200 rpm till the equilibrium was attained. A 100 ppm MeReD solution was used in batch form adsorption tests. The parameters, i.e., contact time (15 min-140 min), dosage effect (0.2 g-1.6 gm per liter), solution pH (2−10), and temperature effect (30–60 ^{o}C) were varied to find out the ideal conditions for MeReD adsorption. Well after batch operation, the adsorbate solution was centrifuged to segregate it from the adsorbent, and the ultimate absorbance and the quantity of the filtrate were measured employing a UV–visible spectrophotometer tuned at 661 nm (maximum absorption of MeReD). Equations below were used to compute the quantity of MeReD adsorbed, qe (mg/g), and the percent of MeReD eliminated at equilibrium:

$$\text{Percent of MRD removed (\%)} = \frac{\text{MRD IC} - \text{MRD EC}}{\text{MRD IC}} \times 100 \quad (1)$$

$$\text{Percent of MRD removed (\%)} = \frac{\text{MRD IC} - \text{MRD EC}}{\text{AS}} \times \text{V} \quad (2)$$

In the equation, MRD IC = initial MeReD concentration (mg/l); AS = Sorbents (leaf and bark materials of L.leu) mass; MRD EC = MeReD equilibrium concentration (mg/l); V = test MeReD solution.

4.4. Study on influence of interfering ions (Co-Ions)

The obstructing ions used in this investigation are the most fundamental particles found in ordinary water. They include anions like sulfate, carbonate, chloride and phosphate and cations like calcium (II), ferrous (II), magnesium (II), zinc (II) and copper (II). The co-ions concentration was kept five times higher than the MeReD concentration (100 ppm) to create the synthetic MeReD and co-ions mixes. In a round bottom flask, 500 mL of synthetic MeReD and co-ions mixes were added, followed by precisely weighted optimal quantities (1.2 gm/L) of adsorbents (leaf and bark materials of L.leu). Optimum pH (4.0 units) was balanced in reaction mix. The reaction mix (co-ions, MeReD and adsorbent) was subjected to rapid agitation (200 rpm) for optimal contact time period (90 min) which is followed by 1 hr of settling. Following that, a volumetric pipette was utilized to retrieve the sample (5 mL), and the starting as well as final concentrations of MeReD were measured at a peak wavelength of 661 nm. The percent of MeReD removal was computed utilising information collected.

4.5. Applications of the developed biosorbents

The adsorbents (leaf and bark materials of L.leu) established in this investigation were exploited to remove MeReD, under optimal extraction conditions established, from actual test effluent samples gathered from dye companies in Hyderabad and Guntur.

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