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Review Paper

Removal of Methylene Blue Using Low Cost Adsorbent: A Review

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Abstract

In this article, adsorption process has been found to be one of the best treatment methods for Methylene blue (MB) removals. As the control of water pollution has become an increasing importance in recent years, the use of physical/chemical treatments such as membrane filtration, reverse osmosis, coagulation/flocculation and fenton reagents are not economically feasible. The use of different biosorbent as an alternative low cost adsorbent in the removal of methylene blue has been extensively studied and compiled, together with their adsorption capacities and experimental conditions such as adsorbent dose, pH of the solution, temperature and equilibrium time. But, there are issues as regards to draw back in the use of activated sorbents which were also discussed briefly. However, it is evident from the results of experiments in the literatures surveyed that various low-cost adsorbents have shown good potential for MB.

Keywords: Adsorption, methylene blue, waste water, low-cost adsorbent.

Introduction

Methylene blue is a common dye mostly used by industries involve in textile, paper, rubber, plastics, leather, cosmetics, pharmaceutical and food industries. Effluents discharged from such industries contain residues of dyes. Consequently, the presence of very low concentrations in effluent is highly visible[1,2]. Discharge of colored wastewater without proper treatment can results in numerous problems such as chemical oxygen demand (COD) by the water body, and an increase in toxicity. Currently, there are about 10,000 different commercial dyes and pigments exist and over $7x10^5$tones of synthetic dyes are produced annually world-wide[3]. It is estimated that 10–15% of the dyes are lost in the effluent during the dyeing processes. Major problems associated with colored effluent is lowering light penetration, photosynthesis and damages the aesthetic nature of the water surface[4-6]. Moreover, their degradation products may be mutagenic and carcinogenic[7-9]. Many dyes may cause allergic dermatitis, skin irritation, dysfunction of kidney, liver, brain, reproductive and central nervous system[10]. Organic dyes are harmful to human beings, the need to remove color from wastewater Effluents become environmentally important. It is rather difficult to treat dye effluents because of their synthetic origins and mainly aromatic structures, which are biologically non-degradable. Among several chemical and physical methods, adsorption process is one of the effective techniques that have been successfully employed for color removal from wastewater[11]. There are currently numerous treatment processes for effluent discharged from industrial processes containing dyes; amongst which we can mention biodegradation[12], chemical oxidation[13,14], foam flotation[15], electrolysis[16], adsorption[17], electro-coagulation[18] and photocatalysis[19]. Major aim of this review is to provide a summary of recent information concerning the use of low-cost materials as sorbents. For this, an extensive list of sorbent literature on methylene blue has been compiled.

Nomenclature:			
T °C	Temperature	S	Sips Isotherm mode
C_o	Initial Concentration (Mg/l)	Te	Temkin isotherm models.
T(min)	Equilibrium Contact Time	TY	Thomas and Yoon–Nelson models,
A.D	Adsorbent dosage (g/l)	K1	Pseudo first order kinetic model
L	Langumuir isotherm model	K2	Pseudo second order kinetic model
F	Freundlich Isotherm model	SDS	sodium dodecylsulfate
R	Redlich–Peterson isotherm model	MB	Methylene blue

Current treatment technologies for color removal involving physical and/or chemical and biological processes: The main components of dye molecules are: the chromophores, which are responsible for producing the color, and the auxochromes, which can not only supplement the chromophore but also render the molecule soluble in water and give enhanced affinity (to attach) toward the fibers[20]. The conventional methods for the treatment of colored wastewater are physical, chemical and biological treatments. However, these technologies have advantage and disadvantages. At large scale, most of these conventional methods are not applicable Because of the high cost anddisposal problems as large amount of sludge is been generated at the end of the process[21].

Physical methods: Physical treatment includes membrane – filtration process, reverse osmosis, electrolysis and adsorption techniques. The major drawback in this technology, especially membrane filtration is limited life time before membrane fouling occurs and as such the cost of periodic replacement must thus be included in any analysis of their economic viability. Among all the physical treatments, adsorption process has been reported to be the most effective method for water decontamination[22]. Adsorption is known to be a promising technique, which has great importance due to the ease of operation and comparable low cost of application in the decoloration process. Commercially activated carbon is a remarkable highly adsorbent material with a large number of applications in the remediation of contaminated groundwater and industrial wastes such as colored effluents. However, activated carbon is an expensive adsorbent due to its high costs of manufacturing and regeneration. For the purpose of removing unwanted hazardous compounds from contaminated water at a low cost, much attention has been focused on various naturally occurring adsorbents such as chitosan, zeolites, fly ash, coal, paper mill sludge, and various clay minerals[23,24]. The use of activated carbon, however, is restricted due to its high cost. An attempt to develop cheaper and effective adsorbents and many non-conventional low-cost adsorbents such as clay materials, zeolites, siliceous material, agricultural wastes and industrial waste products have also been suggested[25,26].

Chemical treatment: The major agents of chemical treatment of dye wastewater are coagulants/ flocculants[27,28,]. It involves the addition of substances such as calcium, aluminum, or ferric ions in to the effluent, as such flocculation is induced[29]. Furthermore, Mishara[30] and Yue[15]have report the use of other agents for chemical processes such as, ferric sulphate, and some synthetic organic polymers. While Shi *et al*[27]suggests the combination of the two methods may also be added to enhance the process. Generally, chemical treatment has economic feasibility and efficiency, but major drawback is that, the cost of chemical are expensive and price fluctuation in market due to high demand and the rate at which chemicals are being produced. Moreover, even though it's efficient, the overall disadvantage of chemical treatment is the production of sludge at the final stage of the treatment which is pH dependent and brings about disposalproblems[31].

Biological methods: Biological treatment of wastewater is an alternative and most economical method as compare to physical and chemical methods. Biodegradation methods such as adsorption by (living or dead) microbial biomass, fungal decolorization, bioremediation systems and microbial degradation are commonly used in the treatment of industrial effluents. Microorganism such as yeasts, bacteria, fungi and algae are able to accumulate and degrade different pollutants, but due to some technical constraints their applications is often restricted[32,33,34]. Biological treatment may be aerobic and anaerobic[35]. But the major drawback is that, it requires substantial land area and is constrained by sensitivity toward diurnal variation as well as toxicity of chemicals [25]. Moreover, contradictory findings were reported in review of current technologies[36] which states that, with current conventional technology, biological treatment is incapable of obtaining satisfactory color elimination. Furthermore, dyes such as (azo dyes) are not easily degradable due to their complex chemical structure, synthetic organic origin and xenobiotic nature[37]. The table below, shows the advantage and disadvantages of physical and chemical treatments.

Table-1
Existing and Emerging processes for dyes removal[38]

Physical/chemical Methods	Method description	Advantages	Disadvantages
Fenton reagents	Oxidation reaction using mainly H_2O_2-Fe(II)	Effective decolorization of both soluble and insoluble dyes	Sludge generation
Ozonation	Oxidation reaction using ozone gas	Application in gaseous state: no alteration of volume	Short half-life (20 min)
Photochemical	Oxidation reaction using mainly H_2O-UV	No sludge production	Formation of by-products
NaCl	Oxidation reaction using Cl+ to attack the amino group	Initial and acceleration of azo bond cleavage	Release of aromatic amines
Electrochemical destruction	Oxidation reaction using electricity	Breakdown compounds are non-hazardous	High cost of electricity
Activated carbon	Dye removal by adsorption	Good removal of a wide variety of dyes	Regeneration difficulties
Membrane filtration	Physical separation	Removal of all dye types	Concentrated sludge Production
Ion exchange	Ion exchange resin	Regeneration: no adsorbent loss	Not effective for all dyes
Electrokinetic coagulation	Addition of ferrous sulphate and ferric chloride	Economically feasible	High sludge production

Despite the development of various technologies for dye waste water treatment, economic, effectiveness and rapid water treatment at a commercial level is still a challenging problem. Previous research efforts have focused on the adsorption technology for the dye remediation from wastewater[39]. This technique can handle fairly large flow rates, producing a high-quality effluent that does not result in the formation of harmful substances, such as ozone and free radicals[40]. Moreover, it can remove or minimize different types of organic and inorganic pollutants and thus has a wider applicability in pollution control. Adsorption is hence recognized as the most versatile process used in lesser developing countries and is currently being used extensively for the removal of organic pollutants from the aqueous media[41,42].

Natural adsorbents used for color removal

Clay: Clay are natural adsorbent classified based on their difference in layered structure.The available classes of clay materials include smectites (montmorillonite, saponite), mica (illite), kaolinite, serpentine, pylophyllite (talc), vermiculite and sepiolite[43]. The process by which adsorption takes place is as a result of net negative charge on the structure of minerals, and it's this negative charge that gives the clay mineral the capability to adsorb positively charged species. Most of Their sorption properties depends their high surface area and high porosity[44].

Siliceous materials: Natural Siliceous materials are one of the most availability and low price adsorbent. It includessilica beads, perlite and dolomite, aluniteand glasses. The use of these minerals was based on chemical reactivity of their hydrophilic surface and mechanically stable, which results from the presence of silanol groups. But among all this, silica beads is given particular attention in the use of the material as adsorbent[45,25,46]. However, Ahmed[47] reports that, a major problem with this kind of application is their low resistance toward alkaline solutions their usage is limited to media of pH less than 8.

Zeolites: Zeolites occur naturally as porous aluminosilicates consisting of different cavity structures and are linked together by shared oxygen atoms[48]. Zeolite has a wide variety of species. More than 40 natural species are available which includes clinoptilolite and chabazite. But, clinoptilolite, a mineral of the heulandite group is the most and frequently studied material, due to its have high selectivity for certain pollutants. Intensive research has been done on the use and application of zeolite as adsorbent in removing trace quantities of pollutants such as heavy metal ions and phenols with regards to their cage-like structures suitable for ion exchange[49-51].

Color removal using activated carbons from solid waste: Activated carbons are derived from natural materials such as wood, lignite or coal, which are commercially available. But almost any carbonaceous material may be used as precursor for the preparation of carbon adsorbents[52-54]. Coal is the most commonly used precursor for AC production Because of its availability and cost effective[55,56]. Coal comprises of different mixtures of carbonaceous materials and mineral matter, which results from the degradation of plants. The nature, origin and the extent of the physical–chemical changes occurring after deposition of vegetation, determines the sorption properties of each individual coal. In a research conducted by Karaka et al [57]attention has been drawn onthe use of coal as a successful sorbents for dye removal. Additionally, coal is not a pure material, and thus will have different sorption properties due to its large variety of surface properties. Recently there has been report on the use of activated carbon in the treatment of dye and heavy effluents. Material such as peanut shell[58], bael shell carbon[59], raw pine and acid-treated pine cone powder[60], Calotropis procera[61], Neem Leaf[62], Coconut Shell[63], Super paramagnetic PVA-Alginate Microspheres[64] were able to reduce the concentration of pollutants in wastewater successfully. Their sorption capacity increases with increasing in adsorbent dosage.

Agricultural waste materials used as low cost adsorbent: The use of biomass (dead or living), fungi, algae and other microbial cultures in the removal of methylene blue was the subject of many recent researches. Biological materials used to accumulate and concentrate dyes from aqueous solution are termed as bioadsorbents. Major disadvantage in these biomaterials is its non-selective (i.e. it cannot isolate each pollutant and get it removed independently of one another) all the target and non-target contaminants if present are concentrated on the surface of the adsorbent. Unlike the conventional ion exchange the process are selective to the ions it needs to adsorb by selecting the ion in such a way that it is having affinity only that ion. Bioadsorption is a novel approach, and considered to be relatively superior to other techniques because of its low cost, simplicity of design high efficiency, availability and ability to separate wide[65]. Recent literature on the methods of removal of dye from wastewater focuses on MB adsorption.

Adsorption capacities of different biosorbent for the removal of MB from wastewater; The excellent ability and economic promise of adsorbents prepared from biomass exhibited high sorption properties from selected literatures in the last one decade are summarized in the tables below.

With the recent development on the use of low cost adsorbent, this review has made tremendous effort to cover a wide range of current researches on nonconventional adsorbents in order to enlighten researchers on the adsorption capacities of different biological material used in recent times as shown from the tables above. In all the studies compiled, it was observed that Equilibrium isotherms and kinetic studies were all determined as observed. Different adsorption isotherm models ranging from lagmuir, freundlich, BET, Temkin and Redlich- peterson were used during to analysed the fittness. Furthermore, based on the knowledged acquired so far, the process of studies on biosorption shuould further be widen in the light of regeneration of bioadsorbents and recovery. Directional modeling, and disposal of the waste material in order to achieve high efficiency. Moreover, it is also observed that most of the studies were reported in batch process, and as such this will provide a room for continuous flow systems design with viable industrial applications, which can be more economical and efficient at commercial level. Having done this, we hope Such a strategy will fulfill the goal of a zero waste.

Table-2
2003-2006

Biosorbent	Q_{max} (mg/g)	pH	Experimental Parameters/ Result					Source
			T(^{O}C)	C_o (mg/l)	T (min)	AD (g/l)	Best fitted model	
Modified polysaccharide	48	8	25	50	150	0.5	-	66
Mango seed kernel powder	142.86	8	30	100		0.02	L and K1	67
wheat shells	21.50	6.5	50	100	60	1.0	L	68
Neem leaf powder	8.76	5-8	30	40	240	2	L and F	69
jute fibre carbon:	22.5	5-10	28	50-200	250	1	L	70
Rice husk	40.58	8	32	100	40	0.12	L and K2	71
Giant duckweed	144.93	9	25	300	144	0.2	K1	72
Date pit	80.3	8	25	5	NA	NA	K2 and L	34
Wool Fibre (sheep wool)	94.3%	5		5g	30	1.0	F and L	73
Cotton Fibre	97%	5		3.5	10	1.50		

Table-3
2007

Biosorbent	Q_{max} (mg/g)	pH	Experimental Parameters/ Result					Source
			T(^{O}C)	C_o (mg/l)	T (min)	AD (g/l)	Best fitted model	
Rattan sawdust	294.14		30	100-200	300	0.1	L and k2	74
Guava Seeds	198.12	8-8.3	500		40		L	75
Dehydrated wheat bran carbon	99.84	2.5	45	200	300	2	L and K2	76
bamboo-based activated carbon:	454.2	7	30	100-500		0.2	L and K2	77
Dehydrated peanut hull	161.30	3.5	50	400	150	1.0	L an K2	78
Paspalum notatum	31	8	30	100	300	1.33	L and K2	79
Rice husk (Coir pith carbon)	5.87	6.8		10-20	60	2	L	80
Wheat Bran	3.08	2.97	20-50	5-20	180	-	K2	81

Table-3
2008

Biosorbent	Q_{max} (mg/g)	pH	Experimental Parameters/ Result					Source
			T(^{O}C)	C_o (mg/l)	T (min)	AD (g/l)	Best fitted model	
Yellow passion fruit waste	44.70	7-10	25	28.7	2880	10	F,R,Land S. K1 and K3	82
leaf powder:	295	7.5	30	517	120	2	L and K2	83
Banana Stalk waste	243.90	4-12	30	50-500	330	1.0	L, F and T and K2	84
Hevea brasiliensis seed coat	227.27		30	50-500	300	0.1	F and K2	85
activated desert plant	23	3-8	24	150	65	4	NA	86
periwinkle shells	500	7	25	400-500	360	0.2	L and K2	87
Sesame stalk	502.68	NA	NA	NA	410	2	NA	88

Table-4
2009

Biosorbent	Q max (mg/g)	pH	Experimental Parameters/ Result					Source
			T(°C)	C_o (mg/l)	T (min)	AD (g/l)	Best fitted model	
Grass waste	457.64.	3-10		380	70	0.05–1.20	L and k2	89
Pummelo peel	390.6	8	30	300	30	2	L and K1	90
Gulmohar plant leaf powder	186.22	7.5	30	10-100		0.5-2.5	L and K2	91
Guava seed	0.10	6	25	50		0.03	-	92
Meranti sawdust	120.48	9-12	60	50-200	180	0.1-1.2	L and K2	93
Tea waste	85.16	4.3+0.2	27+2	20-50	300	0.2	L and K2	94
Garlic peel,	142.86	4-12	50	25–200	210		F and K2	95
Carica papaya seeds	1250	6.25	30	10	120	1.5		96
Pineapple stem waste	119.05	9	30	250	330	03	L and K2	97
Jackfruit peel	285.713	2-11	30	35-400		0.05-1.20	L and K2	98
Papaya seeds (PS)	555.557	3-10	30	50-360		0.05-1.0	L and K2	99
Water hyacinth	426.9	8	30	250		2.0	L	100
Hydrolyzed Oak sawdust	67.78	8	25	300	90	2.5	L and K2	101

Table-4
2010

Biosorbent	Q max (mg/g)	pH	Experimental Parameters/ Result					Source
			T (°C)	C_o (mg/l)	T (min)	AD (g/l)	Best fitted model	
Rhizopus arrhizus	370.3	10	25	50	240	1	F and K2	11
Rhizopus with SDS	1666.6	10		288.4				
Brazil nut shells	7.81	3-10	30	1100	120		L and K2	102
Bamboo	286.1	3.7	25	400	1440	0.1	F, L and K2	103
Activated carbons from walnut shells	315	7.0	25	200	1440	0.75	R, L and F	104
Pretreated rice husk (RH) and rice husk ash (RHA)	1347.7 and 1455.6	7	150	NA	30	NA	L and F	105
Modified sugarcane bagasse	115.3	8						106
Walnut shells via vacuum chemical activation	315		177			0.75	R, L and F	107
Algal biomass	860	4 -10					L	108
Treated sawdust	263.16	7	25	300	480	0.2	L and K2	109
Activated carbon	8.77	6.8	25	25	120	5.0	L and F	110

Table-4
2011

Biosorbent	Q max (mg/g)	pH	Experimental Parameters/ Result					Source
			T(°C)	C_o (mg/l)	T (min)	AD (g/l)	Best fitted model	
Date Stones and Palm-Tree Waste	43.47, 39.47	6.3	20-70	100	240	10	K2	111
NaOH modified rejected tea	242.11	7	30	50-100		0.5	L and K2	112
teak tree bark powder (*Tectonagrandis*)	333.33	7	50	300	30	1	L ,F and K2	113
Cold plasma-treated and formaldehyde-treated onion skins	250, 166.67.	10		50	150	0.15	L and K1	114
oil palm (Elaeis) empty fruit bunch	344.8	2-12	30	200	15	0.1	L	115
Date stones	316.11	2-12	30	300	8	0.1	F, L and T	116
cotton stalk	111.3586	7	35 ± 2	825	120	4	L and K2	117
sulphuric acid treated cotton stalk	381.68	7	35 ± 2			4	F and K2	
and phosphoric acid treated cotton stalk	242.13	7	35 ± 2			4	L and K2	
peanut husk	72.13			80	20	NA	L and K2	118
lotus leaf	221.7	7	20	50-150		1	L, F and Koble–Corrigan	119
palm kernel fibre	95.4	10-11	55	20-160	60	0.4	L, F and T	120

Table-4
2012

Biosorbent	Q max (mg/g)	pH	Experimental Parameters/ Result					Source
			T(^{O}C)	C_o (mg/l)	T (min)	AD (g/l)	Best fitted model	
Pink Guava	250		30	50–500	300	-	L and K2	121
Alkali-modified malted sorghum mash	357.1	7.3	33		18	0.1	L	122
sugar extracted spent rice biomass	8.13	5.2	25	50		0.5	L and K2	123
Water Hyacinth Root Powder	8.04	8		20-10	80	1	L,F and K2	4
Date stones	398.19	7	30	450	270	0.5	S and K2	124
Oil palm shell	133.13		30	150	10		TY	125
Swede rape straw	246.4						L	126
bio-char from pyrolysis of wheat straw	12.03	8-9	20	100	50		S	127

Table-4
2013

Biosorbent	Q max (mg/g)	pH	Experimental Parameters/ Result					Source
			T(^{O}C)	C_o (mg/l)	T (min)	AD (g/l)	Best fitted model	
Pea shells *(Pisum sativum)*	246.91	2 and 11.5	25	100-350	180	1	L	128
coconut husk	500	7.8	30	100-200	30	o.o3	L and K2	129
papaya leaf	231.65	2-10	30	200	300		L	130
Coconut fiber	50					0.6		131
untreated Alfa grass	200	12	20	10–150	180	0.25–12.5	L and K2	132
Neem leaf powder (Azadirachta indica) activated NLP and NLP	401.6 , 352.6	7	87	200	60	3	F and K2	133
Corn husk byZnCl2 activation(CHACZ)	662.25	4	25	50	120	0.4	F	134
HCl Treated SawDust (Lagerstroemia microcarpa):	229.8		30	50-200	360	1.0	L and K2	135
sugarcane *bagasse*:	95.19%	NA		72		0.18	NA	136
watermelon (*Citrullus lanatus*)	489.80	NA	30	50	30	0.5	L and K2	137
Artocarpus odoratissimus (Tarap) skin	184.6	4.4		0.577	2010		L ,S and K2	138
Sugarcanebagasse:	95.19%	8.76	25	72	193	0.19	Na	139
Fallen leaves of platanus	145.62	7		50-500	300	0.5	Te, L andF and Ho	140
Pine sawdust	16.75		35	20-50	180		L and F	141

Conclusion

Based on the literature reviewed so far, it is evident that, recently, there has been an increase in production and utilization of dyes, resulting in an increase in environmental pollution. Various techniques have been utilized in the removal of dyes. However, practically a successful methodology for removal of all types of dyes at low cost has not been established. The results of the literatures above and methods employed during the researches lead to a conclusion that for removal of MB using bio-materials, a collection or combination of different processes involving adsorption yields a rewarding results, but it is also observed that there exist some drawback in biosorption as the cost becomes higher when it is activated. Moreover, despite their upright efficiency and applicability, an economic consideration has restricted the use of some varieties, because substantial amount of adsorbent is loss during regeneration processes. Treatment of industrial wastewater has gained so much importance in recent years, regulations become stricter and researchers have shown clearly for many years, its health, safety and environmental problems if not properly treated before final discharge. Finally, from the data available in literatures, these suggest that MB removal can be achieved to some extent by low cost adsorbent, as some have advantages where by many of them are renewable and available natural resources which are currently under use.

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