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Utilization of Effective Microorganisms based water hyacinth compost as biosorbent for the removal of basic dyes

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ABSTRACT

The water hyacinth plants are renewable natural biomass, proliferate ubiquitously, and abundant in water bodies often posing environmental nuisances. To overcome this problem, a novel approach that utilizes water hyacinth plant has to be arrived. This research work mainly deals with application of low cost, non-conventional, newly developed novel biomaterial, Effective Microorganisms (EM) based Water Hyacinth Compost as biosorbent for the removal of basic dyes from aqueous solution. Biosorption of basic dyes like Methylene Blue (MB), Malachite Green (MG), and Basic Blue 41 (BB41) was investigated using a biosorbent of EM based Water Hyacinth Compost. The biosorbent material was analyzed for physico-chemical composition such as pH, moisture content, organic carbon, nitrogen, etc. and also the biosorbent was characterized before and after adsorption using FTIR and SEM. The batch study was performed to optimize the operational parameters such as pH, biosorbent dosage, biosorbent particle size, and initial dye concentration. The maximum experimental removal efficiency of adsorbent was obtained as 98.9, 98.4, and 89.1% for MB, MG, and BB41, respectively. Two and three parameter adsorption models were used for the mathematical description of the biosorption equilibrium and isotherm parameters were evaluated. The study confirms that the EM-based water hyacinth compost can be used as an effective biosorbent for the removal of basic dyes.

Keywords: Water Hyacinth; Compost; Biosorption; Basic Dyes; Isotherms

1. Introduction

Many industries such as textile, paper, food processing, dyeing, and cosmetics use dyes to color the products. The effluents from these industries are usually polluted by dyes. These dyes usually have aromatic complex structure which makes it very difficult towards degradation. These effluents are mixed with the natural water bodies such as rivers, streams and ecosystem which cause the serious effects on environment such as esthetic pollution, eutrophication, and perturbation in aquatic life. Dye wastewater is usually treated by physical or chemical treatment processes that include chemical coagulation/flocculation,

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precipitation, ozonation, adsorption, oxidation, irradiation, ion exchange, and photodegradation [1]. Some of these techniques have shown to be effective, although they have limitations. Among these are: excess amount of chemical usage, or accumulation of concentrated sludge with disposal problems; expensive plant requirements and operational costs; lack of effective color reduction; and sensitivity to a variable wastewater input [2].

Basic dyes, which are predominantly used in coloring acrylic fiber, are generally more toxic than other classes of dyes [3,4]. Acute exposure to Methylene Blue (MB) can cause increased heart rate, vomiting, shock, Heinz body formation, cyanosis, jaundice, quadriplegia, and tissue necrosis in humans [5]. Malachite Green (MG) is highly toxic, highly residual, teratogenic, carcinogenic, mutagenic, creates other side effects to fresh water organisms [6,7] and enters food chain and alarmed the health hazards against human being. The use of Basic Blue (BB) dye can cause eye burns to human and animals, irritation to gastrointestinal tract with symptoms of nausea, vomiting and diarrhea, and also can cause methemoglobinemia, cyanosis, convulsion, tachycardia, and dyspnea [8]. Thus, the removal of basic dyes from the environment is worthwhile noting.

To conflict these problems, the environmental friendly measures are required. Among the different conventional wastewater treatment techniques, biosorption is found to be an efficient and cost-effective technique. The possibility of recovery of adsorbate and adsorbent has made the biosorption process more attractive [9]. Since past several decades, the application of adsorption process has become the focus of intense research and utilization of waste materials as potential adsorbents is gaining stern consideration towards solid waste management [10].

Nowadays research has indicated that biosorption is one of the most promising technologies and the removal of dyes by different kinds of biosorbent materials has been receiving more attention [11–13]. Table 1 shows the adsorptive capacity of various biosorbents for the selected basic dyes. The water hyacinth (*Eichhornia crassipes*) is a floating macrophyte and its growth is enormous in the water bodies of Tamil Nadu, India. It is listed as one of the most productive plants on earth and is considered one of the world's worst aquatic plants [14]. The water hyacinth growth forms dense mats that avert river traffic, block irrigation canals, interfere with hydel power projects and destroy rice fields [15].

Composting is the biological degradation of organic matter under aerobic conditions to a relatively stable humus-like material called compost [16]. The micro-organisms present in the organic materials are responsible for decomposition. The decomposition by Effective Microorganisms (EM) is safe and simple way of improving the composting system. EM is a mixture of groups of organisms that has a reviving action on human being, animals, and the natural environment [17] and described as a multi-culture of coexisting anaerobic and aerobic beneficial microorganisms [18]. EM can ensure high-quality compost as the microbial inoculant increases the production of aerobic bacteria and increases the composting speed. The main species involved in EM include lactic acid bacteria, photosynthetic bacteria, yeasts, actinomycetes, and fermenting fungi [19].

The aim of the present investigation is to explore the possibility and effectiveness of utilizing EM-based water hyacinth compost (EM_{WHC}) as an alternative, low-cost adsorbent for the biosorptive removal of Methylene Blue, Malachite Green, and Basic Blue 41 dyes from aqueous solution. To the best of our knowledge, this study is the first attempt on EM-based Water Hyacinth Compost onto the biosorption process.

2. Materials and methods

2.1. Adsorbent

EM-based water hyacinth compost was prepared from waste water hyacinth plant and it was collected from nearby ponds and channels. The compost bed was prepared by forming the number of layers with cow dung, water hyacinth waste, and saw dust which were moist by EM in a compost bin.

EM is available in dormant state which procured from a local vendor and requires activation before application. Activation involves the addition of water, jaggery, and dormant EM as recommended by Sekaran et al. [20]. About 45 days, the volume of compost bed had dropped substantially due to thermophillic process and the temperature was within the ambient temperature. At this point, the matured compost was collected and dried. This derived EM_{WHC} can be used for various applications especially as biosorbent. The prepared adsorbent was air dried for 24 h and oven dried at a temperature of 70°C for 2 h and sieved.

2.2. Characterization of adsorbent

The composition of EM_{WHC} was characterized and analyzed by the following procedures. The pH of biosorbent was measured by pH meter (Ecosan, EUTECH Instruments), Moisture Content by oven dry method, Organic Carbon by Walkely and Black

Table 1
Biosorption capacity of various sorbents

Dye	Adsorbent	Uptake/removal efficiency	Refs.
Methylene Blue	Coco-peat	212.8 mg/g	[38]
	Saw dust	236.16 mg/g	[39]
	Corynebacterium glutamicum	99.7%	[40]
	Waste sludge	99%	[41]
	Wheat straw	396.9 mg/g	[42]
	Rice husk	312.0 mg/g	[43]
Malachite Green	Activated carbon (mesophorous)	99%	[44]
	Activated carbon from *Cassia fistula*	28.81%	[45]
	Saccharomyces cerevisiae	17 mg/g	[46]
	Mangrove barks	129.87 mg/g	[47]
	Anaerobic granular sludge	61.73 mg/g	[48]
	Coffee bean	95%	[49]
Basic Blue 69	Peat	195 mg/g	[50]
Basic Blue 9	*Aspergillus niger*	8.3 mg/g	[51]
Basic Blue 9	Activated sludge	256.41 mg/g	[52]
Basic Blue 54	Pine wood AC	1,119 mg/g	[53]
Basic Blue 69	Bagasse pith	152 mg/g	[54]

Table 2
Physico-chemical composition of EM-based water hyacinth compost

Parameters	Level
pH	6.67
Moisture content	52 %
Electrical conductivity	1.74 dSm^{-1}
Temperature	34℃
Manganese	56 mg/L
Copper	28 mg/L
Zinc	54 mg/L
Organic Carbon	13.5% by dry mass
Nitrogen	0.64% by dry mass
Phosphorous	0.21% by dry mass
Potassium	0.045% by dry mass
C:N ratio	21.1

titration method, Nitrogen and Phosphorous by UV–visible spectrophotometer (Merck, Spectroquant Phara 300), and Potassium by digital flame photometer. The physicochemical composition of EM_{WHC} is summarized in Table 2. FTIR (Perkin–Elmer Paragon 500 FTIR) spectra of EM_{WHC} samples were obtained by preparing the samples as KBr disk and examined within the range 400–4,000 cm^{-1} to identify the specific functional groups responsible for the biosorption. To determine the morphological structure and surface characteristics of EM_{WHC} and dyes-loaded EM_{WHC}, the samples were coated under vacuum, with a thin layer of gold examined by Scanning Electron Microscopy (FESEM S-4700, Hitachi).

2.3. Adsorbate

Basic dyes of MB, Malachite Green, and BB41 were purchased from Merck India Ltd. and Sigma–Aldrich Chemicals with wavelength of 665, 618, and 609 nm, respectively. The dyes were used as commercial salts without further purification. The dye stock solutions were made up to a concentration of 2,000 mg/L by dissolving an accurately weighed quantity of dye in deionized water and was subsequently diluted to the required concentrations. The pH of working solutions were adjusted to the desired pH values by 0.1 M HCl and 0.1 M NaOH.

3. Results and discussion

3.1. FTIR Study

The functional groups that are responsible for adsorption present in the EM-based water hyacinth compost and MB, MG, and BB41-loaded composts were analyzed by FTIR and the spectra are illustrated in Fig. 1. The FTIR spectra exhibit a number of absorption peaks, indicating the complex nature of adsorbent examined before and after adsorption of dyes. The quantity of the characteristic functional groups can be identified by the steepness of the peaks. The absorption peaks about 3,000 cm^{-1} represents the presence of

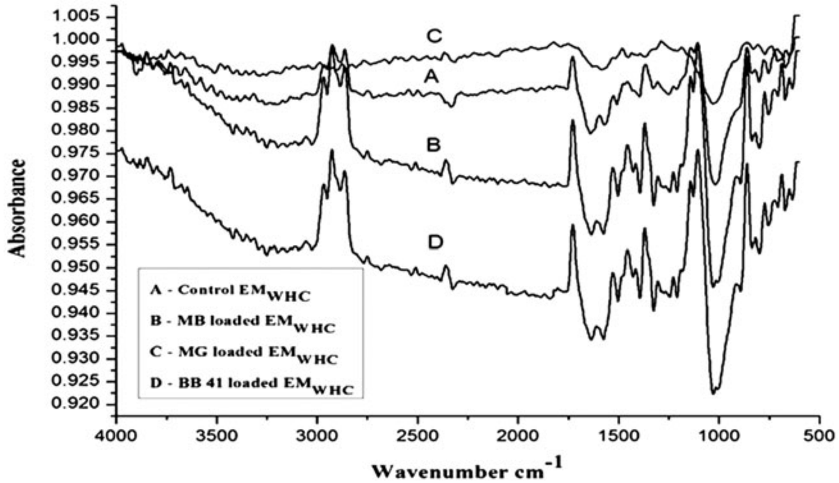

Fig. 1. FTIR spectrum for EM_{WHC} Control and MB, MG, and BB 41-loaded EM_{WHC}.

amine groups. The bands between 3,500 and 3,300 cm^{-1} are reported to occur due to amine group stretching vibrations superimposed on the side of hydroxyl group band [21]. The spectra showing the peak at 1,107 cm^{-1} indicates the carboxyl groups, 1,729 cm^{-1} indicates the C–O stretching, 862 cm^{-1} specifies the aromatic, and 500–750 cm^{-1} indicates the alkane groups [22].

3.2. SEM Study

The morphological structure of EM_{WHC} and the dyes loaded EM_{WHC} are presented in Fig. 2(a)–(d). The SEM image (Fig. 2(a)) clearly shows that the control EM_{WHC} was not smooth and has large number of pores resulting in increased surface area for bonding of dyes [23]. The images of Fig. 2(b)–(d) confirmed that the pores present in the control EM_{WHC} were occupied by the dyes of MB, MG, and BB41 which was confirmed from the smooth surface.

3.3. Effect of pH

In the aqueous solution, pH is one of the most significant parameter which affects the process of dye removal. The solution pH influences the surface charge of adsorbent, the degree of ionization processes of the dye molecules, and also the effluent chemistry. In addition, it is directly related to competition ability of hydrogen ions with adsorbate ions to active sites on the adsorbent surface [24].

The effect of pH on the decolorization of dyes such as MB, MG, and BB41 were evaluated at different pH conditions ranging from 2 to 9, and the results are described in Fig. 3. The decolorization efficiency of dyes in batch mode (dye concentration 100 mg/L, desired adsorbent dosage and agitation speed 150 rpm) increased notably from pH 2 to 8 for MB and MG, whereas from 2 to 7 for BB41. The maximum removal efficiency of dyes (95.3% for MB, 98.4% for MG, and 90.2% for BB41) was obtained at pH 8, 8, and 7, respectively. Then the removal efficiency was decreased with the increase in pH to 9. Thus, the pH value was optimized for the selected dyes as 8 for MB, MG, and 7 for BB41. At low pH, the sorption was unfavorable, probably because of excess H^+ ions competing for sorption sites. Increasing solution pH increases the number of hydroxyl groups thus, increases the number of negatively charge sites, and enlarges the attraction between dye and adsorbent surface [25].

The point of zero charge (pH_{PZC}) was determined by solid addition method [26]. pH_{PZC} gives very significant information about the type of surface active centers. The pH_{PZC} of EM_{WHC} biomass was found to be 6.7. Below pHpzc, the biosorbent acquires a positive charge which favors the attachment of dye anions to the surface of biosorbent. Above pHpzc, the biosorbent gets a negative charge and the electrostatic repulsion between dye anions and negatively charged functional groups results in lower dye removal [27].

3.4. Effect of dosage

To evaluate the optimum dosage, the decolorization of dyes was measured in an aqueous solution with adsorbent dosage varied from 2 to 10 g/L (optimum pH, dye concentration 100 mg/L and agitation speed 150 rpm). The results are illustrated in Fig. 4

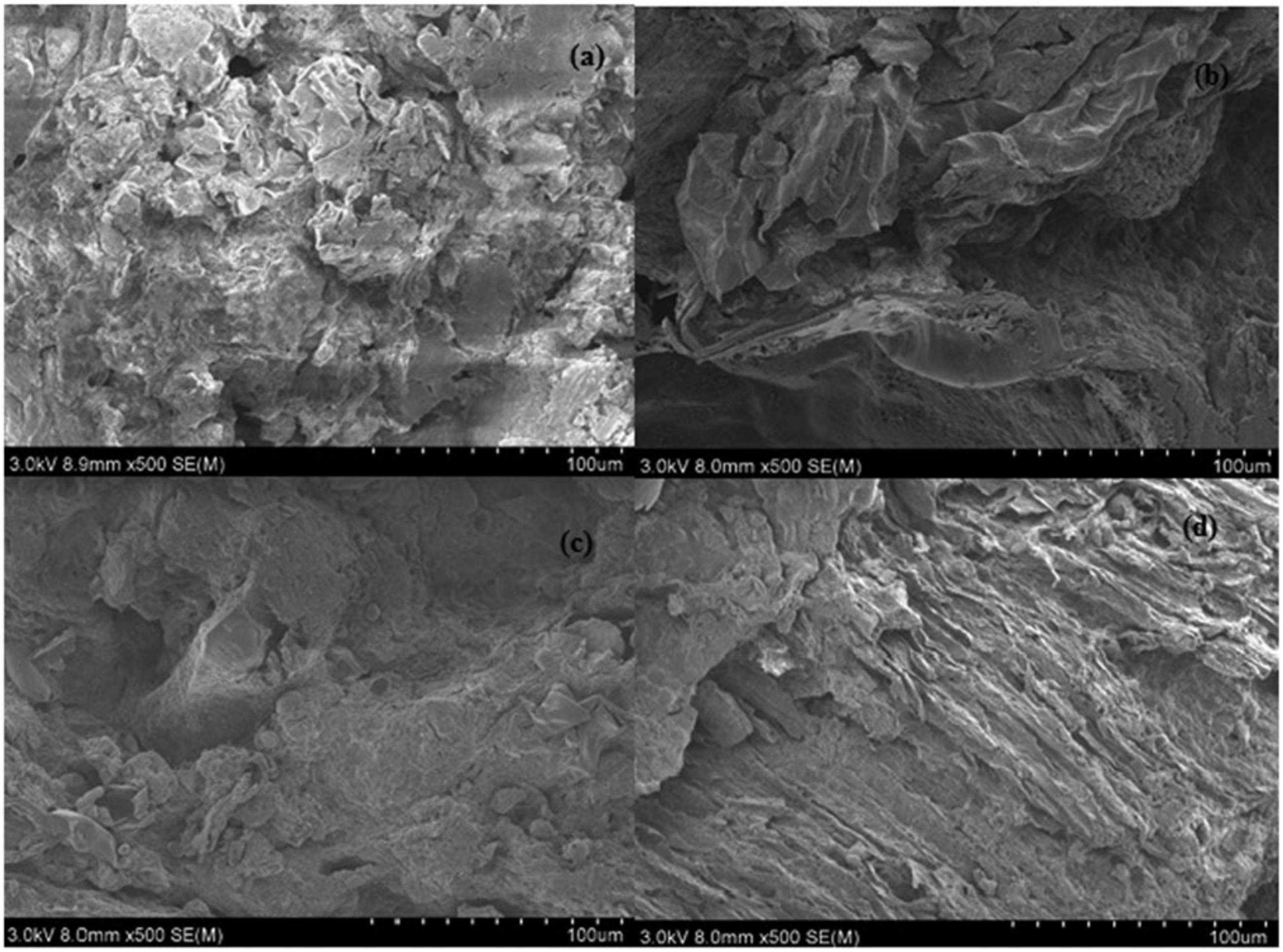

Fig. 2. SEM images of (a) EM_{WHC} Control and loaded with (b) MG, (c) MB, and (d) BB41.

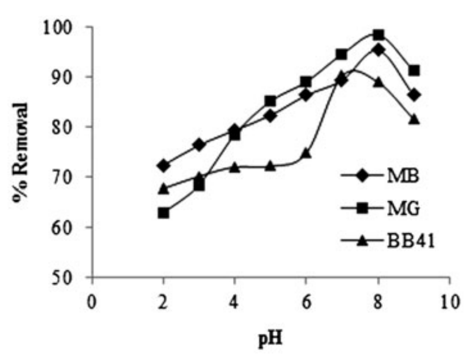

Fig. 3. Effect of pH on removal of MB, MG, and BB41.

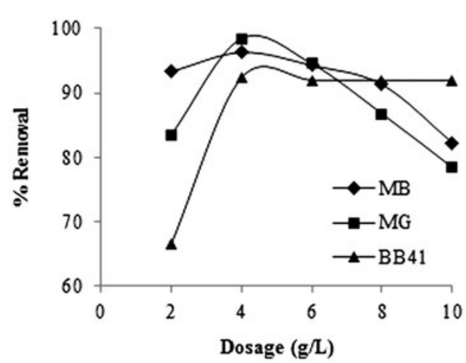

Fig. 4. Effect of adsorbent dosage on removal of MB, MG, and BB41.

and show the maximum removal efficiency of dyes (96.3% for MB, 98.44% for MG and 92.35% for BB41) with the EM_{WHC} at the dosage of 4 g/L. By increasing the amount of adsorbent, access to the residual sites for dye adsorption was restricted. In addition, the effective surface area for adsorption decreased due to the partial aggregation of adsorbent at high adsorbent dosages leading to a decrease in the adsorption capacity of dyes [28]. From the maximum removal efficiency, it has been decided to use 4 g/L as optimum adsorbent dosage.

3.5. Effect of initial dye concentration

The initial dye concentration provides the necessary driving force to overcome the resistances to the mass transfer of dye between the aqueous and solid phases [29]. The effect of initial dye concentration was experimentally studied by varying the concentration of dyes (from 50 to 1,000 mg/L) by $EM_{WHC.}$ From Fig. 5, the adsorption capacity decreased when the concentration of solution increased. The maximum removal efficiency of dyes was observed at lower dye concentration of 50 mg/L as 98.6, 98, and 88.3% for MB, MG, and BB41, respectively. At 1,000 mg/L, the removal efficiencies were recorded as 70.4, 59.1, and 55.7% for MB, MG, and BB 41, respectively. At lower dye concentrations, solute concentration to biosorbent site ratio is higher, which cause an increase in color removal and at higher concentrations, lower adsorption yield is due to the saturation of adsorption sites [30].

3.6. Effect of particle size

Adsorbent particle size has significant role in exhibit the active sites in the adsorption process. The adsorbent particle sizes from 0.6 to 2.36 mm were used to optimize the size with the optimal parameters of pH, dosage, and initial dye concentration and the results are presented in Fig. 6. The maximum removal efficiencies were recorded at the particle size of 1.18 mm for MB, MG, and at 0.75 mm for BB 41. The increase in sorption depends on the large external surface area for smaller particles. Apparently, reductions in particle size resulted in increased external surface area of the adsorbent particles yielding more binding sites for dye adsorption and therefore, more efficient adsorption [31]. In order to compete the industrial applications and the recycling of adsorbent, the optimum size of particle was selected as 1.18 mm.

3.7. Sorption isotherms

The isotherm models are widely used to describe the equilibrium between biosorption capacity (q_e) and sorbate concentration (C_e) at a constant temperature [32]. The two parameter models of Langmuir and Freundlich isotherms and three parameter models of Redlich–Peterson, Toth, and Sips isotherms were discussed as follows:

$$\text{Langmuir model: } Q = \frac{Q_{\max} b_L C_f}{1 + b_L C_f} \tag{1}$$

$$\text{Freundlich model: } Q = K_F C_f^{1/n_F} \tag{2}$$

$$\text{Redlich-Peterson model: } Q = \frac{K_{RP} C_f}{1 + a_{RP} C_f^{\beta_{RP}}} \tag{3}$$

$$\text{Toth model: } Q = \frac{Q_{\max} b_T C_f}{[1 + (b_T C_f)^{1/n_T}]^{n_T}} \tag{4}$$

$$\text{Sips model: } Q = \frac{K_S C_f^{\beta_S}}{1 + a_S C_f^{\beta_S}} \tag{5}$$

where Q is the equilibrium dye uptake (mg/g); C_f is the final dye concentration (mg/L); $Q_{\max}$ is the maximum dye uptake (mg/g), b_L is the Langmuir equilibrium coefficient (L/mg), K_F is the Freundlich coefficient $(L/g)^{1/n_F}$, n_F is the Freundlich exponent, K_{RP} is the Redlich–Peterson isotherm coefficient (L/g), a_{RP} is the Redlich–Peterson isotherm coefficient

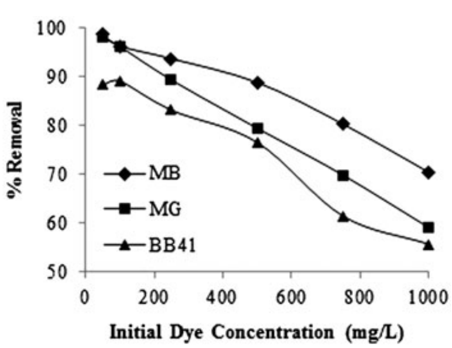

Fig. 5. Effect of initial dye concentration on removal of MB, MG, and BB41.

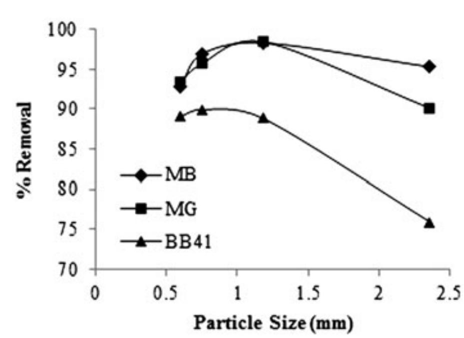

Fig. 6. Effect of adsorbent particle size on removal of MB, MG, and BB41.

$(L/mg)^{\beta_{RP}}$, β_{RP} is the Redlich–Peterson model exponent, b_T is the Toth model constant (L/mg), n_T is the Toth model exponent, K_S is the Sips model isotherm coefficient $(L/g)^{\beta_S}$, a_S is the Sips model coefficient $(L/mg)^{\beta_S}$ and β_S is the Sips model exponent.

The average percentage error between the experimental and the predicted values is calculated using,

$$\%\ \text{Error} = \frac{\sum_{i=1}^{N}\left(Q_{\text{exp},i} - Q_{\text{cal},i}/Q_{\text{exp},i}\right)}{N} \times 100$$

where Q_{exp} and Q_{cal} represent the experimental and calculated uptake values, respectively, and N is the number of measurements.

The experimental equilibrium uptakes and their predicted values by five different models, Langmuir, Freundlich, Redlich–Peterson, Toth, and Sips for the three basic dyes are plotted in Fig. 7. The five isotherm model constants and correlation coefficients with percentage error were evaluated for the dyes MB, MG, and BB41 are presented in Table 3.

The derivation of the Langmuir isotherm assumes ideal monolayer adsorption on a homogenous surface. From the observed data, the correlation coefficient R^2 for the Langmuir model was greater than those for Freundlich model, suggesting the monolayer coverage of the EM_{WHC} surface by dye molecules [33]. According to Langmuir isotherm, the maximum uptake was obtained as 295.65, 153.0, and 158.04 mg/g for MB, MG, and BB 41, respectively. The Freundlich isotherm is used for non-ideal adsorption on heterogeneous surfaces. The heterogeneity arises from the presence of different functional groups on the surface, and the various adsorbent–adsorbate interactions [34]. From the Fig. 7 it was evident that, the maximum predicted uptake for all the dyes were observed for Freundlich isotherm compared with other two- and three-parameter isotherms. The values of Freundlich constant $(1/n)$ for the examined basic dyes were predicted as 0.29, 0.32, and 0.44, which is less than 1 indicates the favorable dye adsorption. This indicates the fact that the active sites on sorbents have different affinities to dye molecules [35]. However, the prediction of isotherm data by the Freundlich model was not agreeable because of high % error (Table 3).

In order to improve the suitability of isotherm data, the three parameter models of Redlich–Peterson, Toth, and Sips were used. The Redlich–Peterson model is a combination of Langmuir and Henry's model which incorporates three parameters (K_{RP}, a_{RP}, and β_{RP}) and thus can be applied to either homogenous or heterogeneous systems [36]. When β_{Rp} = 0, the model reduces to the Langmuir model, while β_{Rp} = 1, the model transforms to Henry's law form. From the data obtained, β_{RP} was estimated as 0.88, 0.97, and 0.88 for MB, MG, and BB 41, respectively, which are close to unity indicating that the isotherms are approaching the Langmuir, but not the Henry isotherm. The Redlich–Peterson model resulted in better R^2 (0.99) and low % error (<5.51%) values. The Toth model derived from potential theory has proven useful in describing sorption in heterogeneous systems. It assumes an asymmetrical Quasi-Gaussian energy distribution with a widened left-hand side, i.e. most sites have sorption energy less than the mean value [37].

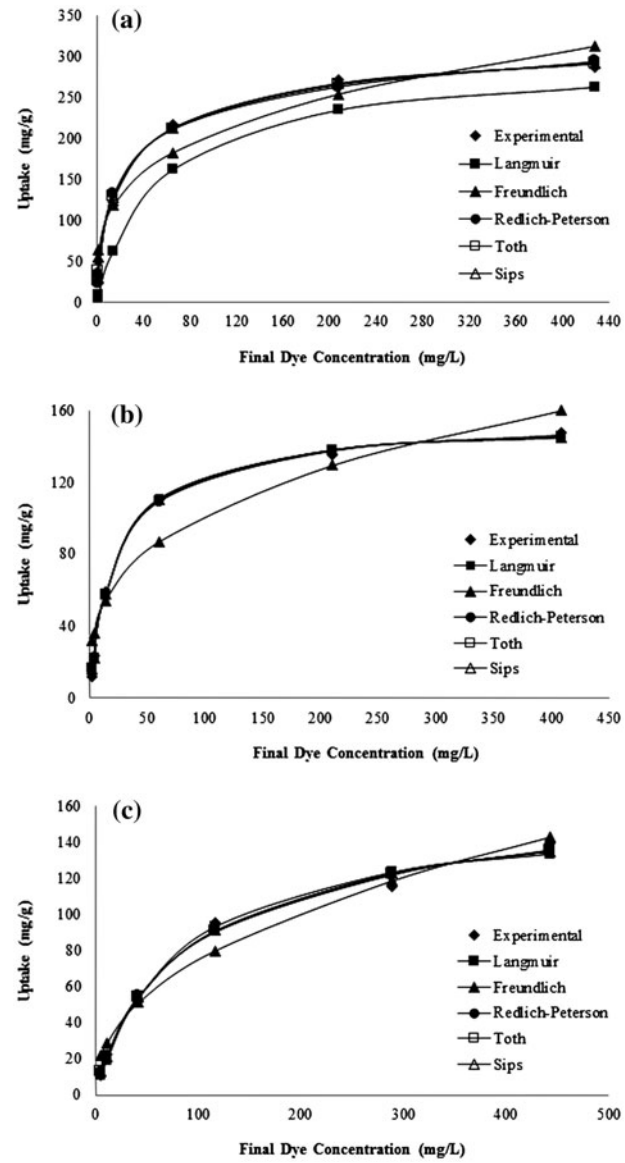

Fig. 7. Biosorption Isotherms for (a) MB, (b) MG, and (c) BB41 onto EM_{WHC}.

Table 3
Isotherm model parameters for MB, MG, and BB41 onto EM_{WHC}

Models	Parameters	MB	MG	BB41
Langmuir	Q_{max} (mg/g)	295.65	153.00	158.04
	K_a (L/mg)	0.018	0.044	0.0123
	R^2	0.99	0.99	0.99
	% error	42.65	3.94	2.71
Freundlich	K_F (mg/g) $(L/mg)^{1/n_F}$	54.95	23.47	9.97
	$1/n$	0.29	0.32	0.44
	R^2	0.97	0.92	0.97
	% error	22.89	32.45	18.24
Redlich–Peterson	K_{RP} (L/g)	27.08	7.15	2.44
	a_{RP} $(L/mg)^{\beta_{RP}}$	0.19	0.054	0.033
	β_{RP}	0.88	0.97	0.88
	R^2	0.99	0.99	0.99
	% error	5.27	5.51	1.42
Toth	Q_{max} (mg/g)	366.65	155.30	185.90
	b_T (L/mg)	0.13	0.046	0.014
	n_T	1.94	1.06	1.37
	R^2	0.99	0.99	0.99
	% error	1.43	5.03	1.52
Sips	K_S $(L/g)^{\beta_S}$	31.28	6.94	2.89
	a_S $(L/mg)^{\beta_S}$	0.09	0.045	0.017
	β_S	0.69	0.99	0.88
	R^2	0.99	0.99	0.99
	% error	0.31	4.56	1.84

The Toth model exhibited higher R^2 (0.99) values and less % error for the dyes MB, MG, and BB 41. The Sips model is postulated with the assumptions that surface active sites are of different strengths and that one molecule of adsorbate interacts with one active site [38]. At low and high sorbate concentrations, the Sips model effectively reduces to the Freundlich and Langmuir equations, respectively. The Sips model parameter β_S was estimated as 0.69, 0.99, and 0.88 for MB, MG, and BB 41, respectively. Based on the correlation coefficient and % error values, MB adsorption was well explained by Sips model, MG removal was described well by Langmuir model, and Redlich–Peterson model fitted well with the experimental data of BB 41 adsorption.

4. Conclusion

The effectiveness of EM-based water hyacinth compost onto the removal of basic dyes MB, malachite green, and BB41 was investigated.

(1) It is evident that water hyacinth, an abundantly available organic waste material, can be converted as compost using EM.
(2) The maximum removal efficiency of EM_{WHC} onto MB, MG, and BB 41 was observed as 98.9, 98.4, and 89.1%, respectively.
(3) The experimental results revealed that, the maximum uptakes (286.15 mg/g for MB, 147.81 mg/g for MG, and 139.29 mg/g for BB 41) were exhibited at the optimum operating conditions.
(4) Two- and three-parameter isotherms were modeled for dyes adsorption. Based on correlation coefficients and percentage error values, Sips isotherm model described the experimental data well.
(5) The EM_{WHC} can be used as a good biosorbent for MB, Malachite Green, and BB41 and it is suggested for the removal of basic dyes from wastewater.
(6) The present study concludes that EM-based water hyacinth compost could be employed as a low-cost, novel, and green biosorbent in removing the dyes from textile effluents.

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