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Modified Mangosteen Pericarp Powder as a Biosorbent for Removal of Methyl Violet 2B from Aqueous Solutions

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Abstract

This paper proposes the removal of methyl violet 2B from aqueous medium using modified mangosteen pericarp (MMPB) as a biosorbent. The characteristic of surface biosorbent was examined by SEM and FTIR. It was found that surface morphology was porous structure and demonstrated various functional groups which were advantageous for the adsorption of cationic dye. The effect of pH solution, amount of biosorbent, and contact time on the dye removal efficiency were tested by batch experiments. The results indicated that the percentage of dye removal increased with an increase in pH solution, amount of biosorbent, and contact time. The experimental data showed that the removal efficiency of the dye was higher than 95% from 10mg/L of initial MV2B concentration with the pH solution range from 6 to 10 using amounts of biosorbent as 0.05 g in 25 mL of dye solution and contact time of 90 min. The adsorption isotherm was well fitted to the Langmuir model (R^2 = 0.982) with a maximum adsorption capacity (q_m) of 49.75mg/g, suggesting the monolayer onto a homogenous surface. This study implied that modified mangosteen pericarp can be employed in the act of an inexpensive biosorbent for methyl violet 2B removal from aqueous solutions.

Keywords: mangosteen pericarp, biosorption, methyl violet, aqueous solutions

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1. Introduction

Synthetic dyes are essential raw materials in many industries such as textiles, ink, plastics, food and beverage, cosmetics, leather, etc. A large demand for synthetic dyes was reported, as seen from the global market data in the year 2020 showing the synthetic dyes market gained a cost of nearly 18,037.1 million USD and the production volume amounted to 35,294.7 tons. The demand is expected to grow owing to the increasing usage of synthetic dyes for various industries [1]. However, the environmental problem regarding dyestuff has been mentioned [2-4]. since synthetic dyes compose of complex organic chemical compounds. Not only are they toxic, but also have difficult biodegradable properties [4,5]. If they pollute the environment, they may persist for a long time and make adverse impacts on the environment, especially aquatic ecology. When untreated wastewater containing synthetic dye is released into water body, it can produce undesirable aesthetics [5,6] as well as impair water quality to unsafe water for daily activity purposes. Furthermore, the dye may prevent sunlight penetration, causing diminished photosynthetic activity and then begetting low dissolved oxygen concentration. These may affect aquatic lives in receiving waters [5-7]. The dye contaminants can be bioaccumulated and biomagnified in food webs, linking to harmful aquatic organisms and human health impacts [2,8,9]. Previous literature reviews that synthetic dyes pose as a source of allergic, toxic, carcinogenic, and mutagenic agents [6-11], depending on their chemical compositions. They thus are blamed for the hazard which threatens human body such as skin and eye irritation, respiratory system, kidney dysfunction, reproductive system, enzymatic system, brain, and central nervous system [2,8-10].

Methyl violet 2B, or Basic Violet 1, is one of the dangerous cationic dyes. It composes of a triphenylmethane group and, pentamethyl pararosanilins [11]. It is known that triphenylmethane can form to carcinogenic agent. If it is degraded under anaerobic conditions, it produces aromatic amine, causing of mutation and cancer [8]. Moreover, it can be responsible for both acute and chronic harmful effects on aquatic organisms [10]. Thus, the elimination of this dye from wastewater is needed before discharging into receiving waters. Nowadays, biosorption is an interesting technique to uptake dye from aqueous phase. Previ-

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ous studies suggest that it is an effective, simple, and budgetary method [12,13]. The term of biosorption is classified in a subcategory of adsorption. It can be described as any system in which a solid surface of a biological material that binds with an adsorbate molecule resulting in the lower remaining adsorbate concentration in the aqueous solution [12,14]. The adsorbent employed in the biosorption process is derived from biological materials [12] and is also called a biosorbent. Various biomass materials are adopted as a precursor to prepare adsorbents such as palm kernel fiber [11], water hyacinth [13], almond shells [15,16], olive stones [17], and tarap fruits [18]. Mangosteen (*Garcinia mangostana* L.) is a popular tropical fruit that has an attractive taste, as well as containing a variety of essential nutrients, fiber, and antioxidants [19,20]. It can be consumed as a fresh fruit, in addition, it can be processed for many products in the food industry. The non-edible pericarp is found around 60 % of its total fruit weight [21], producing abundance of biowaste after consumption. These pericarps should be utilized for beneficial purposes, in spite of being discarded as solid wastes. Several articles elucidate that mangosteen pericarp presents various functional groups, e.g., phenolic compounds [19,20], hydroxyl, and carboxyl groups [19,22]. These functional groups have the ability to bind with cationic dye molecules [23]. Hence, it is a beneficial material to produce a biosorbent.

In this work, mangosteen pericarp was provided as a precursor to simply prepare the biosorbent which aimed for the removal of methyl violet (MV2B) from the aqueous sample. The pericarp after chemical modification for expelling its water-soluble dyes was applied as a biosorbent. Studies on surface morphology and functional groups of the biosorbent were carried out. Batch experiments were performed to study the efficiency of MV2B removal and the effect of relevant parameters like the amount of biosorbent, contact time, and initial pH's solution. The result obtained from this study was useful to evaluate the ability of modified mangosteen pericarp biosorbent for cationic dye.

2. Materials and Methods

2.1 Adsorbate

Methyl violet 2 B (CI classification number: 42535, $C_{24}H_{27}N_3HCl$, Mr 393.96 g/mol) used in this work was purchased from LOBA Chemie Pvt.Ltd., India. A stock solution of 100 mg/L was prepared by dissolving methyl violet 0.1 g in 1000 mL of distilled water. The serial dye concentrations were prepared by further dilution of the stock solution with distilled water. The initial dye concentration and after adsorption were quantitatively analyzed with a visible spectrophotometer (Hach model DR2700, USA) at a maximum wavelength of 548 nm.

2.2 Biosorbent material

In this work, the biosorbent was derived from the waste of mangosteen pericarp (*Garcinia mangostana* L.) which was obtained from a local market in the eastern region of Thailand. The waste pericarps were cleaned with tap water and distilled water to draw out dust and impurities. Next, they were sliced into small pieces and dehydrated in sunlight until an unchanging weight was achieved. The dried pericarps were ground into powdery particles and stored in a container with a tight-fitting lid for serving as the raw material for adsorbent preparation. Dried biomass powder was reacted with analytical grade of concentrated sulfuric acid (98%) at the ratio of 1:1 (w/v), then left at room temperature for 24 h to complete a carbonized reaction. Afterward, the black carbonaceous residual was leached with distilled water to eliminate the acid and neutralized by sodium hydroxide solution. The suspension was filtered to collect the residue which was reconditioned by stirring in 1 M sulfuric acid solution for 30 min. The wet black cake was separated using Whatman no.1 filter paper and rinsed thoroughly with hot distilled water till the filtrate pH became neutral pH. It was then dehydrated in a hot air oven at 103 °C for 2 h. Finally, the modified mangosteen pericarp powder biosorbent (MMPB) was sieved to less than 75 μm particle sizes and put in an air-seal plastic box for use throughout further experiments. The preparation process is illustrated in Figure 1. The MMPB was obtained by approximately 68 % yield which started from the dried brown pericarp powder. Surface morphology was obtained by scanning electron microscope (LEO1450 VP) and surface functional groups were examined by Fourier transform infrared spectrometer (Perkins Elmer, model Spectrum One). The pH at the point of zero charge (pH_{pzc}) of MMPB was tested with the powder addition method [16]. The solution of 0.01M NaCl was adjusted in the desired pH range from 2 to 12 by using HCl (0.1N) and NaOH (0.1N) solutions. The 0.2 g of MMPB was placed into 50 mL of each solution. The suspensions were shaken for 24 h, then the final pH solutions were determined by using pH-meter. A plot of the initial pH versus the final pH was illustrated and the point where the final pH = the initial pH suggesting as the point of zero charge.

2.3 Batch adsorption studies

Methyl violet adsorption experiments were performed to study the effect of various factors on the dye adsorption namely biosorbent dosage (0.025 – 2 g), contact time (0-240 min), and initial pH (2-10). The methyl violet 2B solution of 10 mg/L was used. The initial dye solution of 25 ml was filled in 125 mL Erlenmeyer flask with the desired amount of biosorbent. The batch experimental sets were agitated on the orbital shaker with the speed of 120 rpm at room tem-

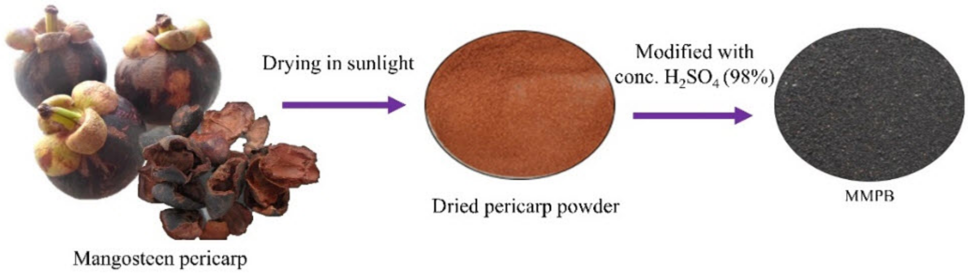

Figure 1: Preparation of modified mangosteen pericarp biosorbent (MMPB)

perature. The percentage of dye removal (R%) was calculated according to the following equations:

$$R = \frac{(C_i - C_e)}{C_i} \times 100\% \quad (1)$$

where C_i and C_e (mg/L) are the initial and equilibrium concentrations of methyl violet in the solution.

3. Results and Discussion

3.1 Characterization of MMPB

The morphology of MMPB (SEM image in Figure 2) confirmed that the MMPB was an irregularly shaped particle. Its surface was obviously rough structure with various pores, which is advantageous application as active site for dye molecule adsorption [23]. FTIR analysis was used to characterize functional groups of MMPB. The FTIR spectrum is shown in Figure 3. The broad band around 3383 cm^{-1} was assigned to -OH stretching vibration indicating the existence of hydrogen-bonded OH groups [16-17,24]. The absorption peak located at 2971 and 2929 were attributed to the -CH stretching vibration [16,24]. The peak at around 1701 cm^{-1} was caused by C=O stretching of the carbonyl functional group from carboxylic acid [23]. The sharp peak occurring near 1594 cm^{-1} corresponded to C=C stretching vibration of aromatic groups [17]. The results informed the presence of various functional groups in MMPB which participated in the adsorption of cationic dye pollutants such as hydroxyl and carbonyl groups [16,23,25]. The pH at the point of zero charge (pH_{pzc}) is used to explain variable charge surfaces of biosorbent. The pH_{pzc} of MMPB was tested to be around 2.1. In the case that the pH of aqueous solution is lower than the pH_{pzc} value, the adsorbent has a positive charge on the surface. While the pH of solution is higher than the pH_{pzc} value, the surface of the adsorbent has a negative charge [26].

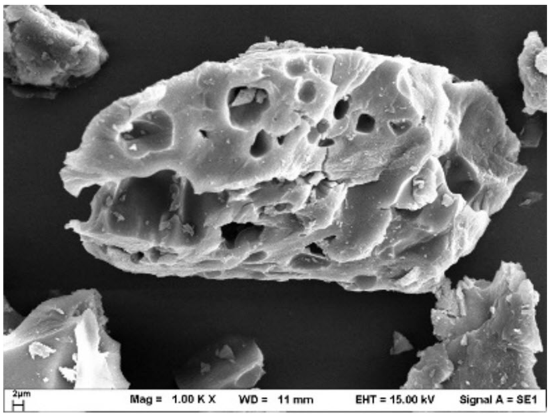

Figure 2: Scanning electron microscopy of MMPB x 1000 magnitude

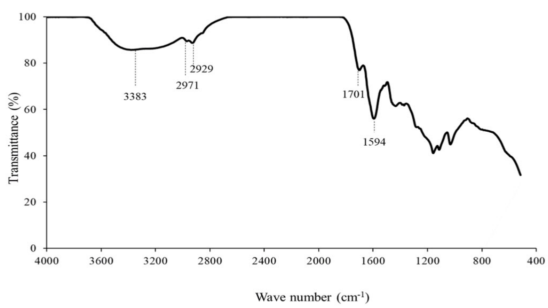

Figure 3: FTIR spectrum of MMPB before adsorption of MV 2B dye

3.2 Effect of biosorbent dosage

The effect of MMPB amount in the range of 0.025 to 0.5 g on methyl violet adsorption was depicted in Figure 4. The removal percentage of dye raised from 60.46 % to 96.85 % with those increasing MMPB amount. It can be described by increasing in the surface area which leads to more active sites for the dye molecule adsorption [4,13]. In the event that the amount of MMPB was varied from 1 to 2 g, the removal percentage dropped slightly from 95.70% to 94.75%. This may be accredited to the agglomeration of biosorbent particles which obstruct the interaction between dye molecules and active sites on the biosorbent surface [13,16]. As a result, the amount of MMPB at 0.05 g exhibited high adsorption performance attaining 95.42% of dye removal. Consideration from an economic perspective this amount was appropriated for further batch experiments.

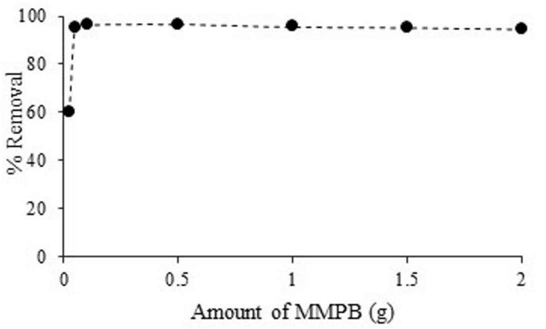

Figure 4: Effect of adsorbent amount on the adsorption of MV 2B dye using MMPB

3.3 Effect of contact time

The effect of contact time in the methyl violet 2B adsorption was conducted by varying contact time from 15 to 240 min. The result was illustrated in Figure 5. It indicated that the percentage of dye removal increased with increasing time. Rapid adsorption process was noticed within the first 60 minutes showing high removal efficiency of around 97 % since the availability of active sites for dye adsorption [13,15,18]. After that the dye removal percentage was nearly constant with the approximate value of 98 %. As shown in the curve suggesting the equilibrium time was achieved at around 90 minutes.

3.4 Effect of initial pH

The pH is regarded as a significant factor to the adsorption behavior. It has influence over the surface charge of adsorbent and the property of dye [9,18]. The effect of pH on the adsorption of MV 2B dye onto the MMPB surface was studied within the initial pH range from 1.8 to 10. As can be seen from Figure 6, the pH increased from 1.8 to 4.1, percentage of dye elimination increased sharply from 88.22 % to 94.54%. This phenomenon can be explained that, in the case of acidic conditions, the excess H^+ ions compete with dye cations for adsorption on the active sites causing unfavorable dye adsorption [4]. Moreover, when the initial pH was less than pH_{pzc} (at pH 2.1), bringing the surface of the MMPB had more positive charges. This is suppressed condition for the binding between MV2B dye molecule and the positively charged adsorbent surface, resulting in lowering adsorption efficiency. When the initial pH was higher than pH_{pzc}, the surface had more negatively charged sites, favoring the adsorption of biosorbent to the MV2B cations [15]. However, the efficiency of dye removal was almost constant from pH 6 to 10 with the percentage of dye removal at approximate 96 %. It can assume that at these pH values, they effect insignificantly to the adsorption process without a loss of color of positively charged MV 2B molecules. A neutral pH is an optimal condition to apply the further batch adsorption studies because this pH value is higher than pH_{pzc} of the MMPB affected to the favorable adsorption process by electrostatic interaction. In the pH range between 11.0 and 13.0, a chemical reaction between the MV2B and OH- ions occurs to produce a colorless base carbinol form resulted the shift to the UV region and then it is not possible to investigate the adsorption process using the maximum wavelength of 548 nm.

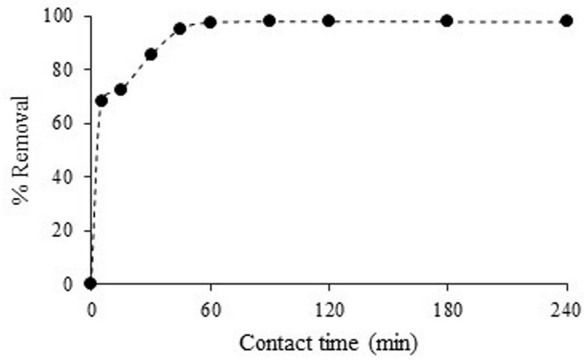

Figure 5: Effect of contact time on the adsorption of MV 2B dye using MMPB

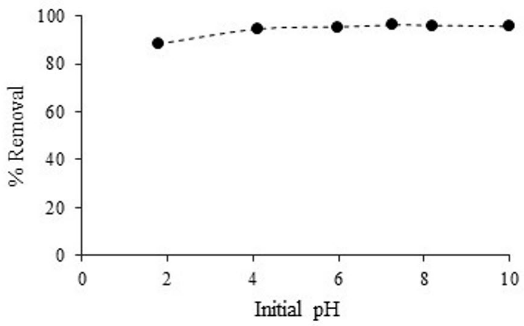

Figure 6: Effect of pH on the adsorption of MV 2B dye using MMPB

3.5 Adsorption isotherm

Adsorption isotherm are the main issue for describing adsorbate-adsorbent interactions at equilibrium. The most well-known Langmuir and Freundlich adsorption isotherm models were applied to examine the validity of the experimental data for MV2B adsorption onto biosorbent. The Langmuir theory is based on the process type of monolayer adsorption. Moreover, it assumes a structurally homogeneous surface, in which all adsorption sites are identical and energetically equivalent [16]. The Langmuir model is presented by the following linear transform:

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \qquad (2)$$

where C_e(mg/L) is the equilibrium MV concentration in the solution; q_e (mg/g) is the equilibrium amount of MV adsorbed per unit mass of the solid biosorbent; q_m (mg/g) is the maximum adsorption capacity at saturation of the biosorbent surface by a monolayer and K_L (L/mg)is the Langmuir constant related to the free energy of biosorption.

The Freundlich model is defined as the process type of multilayer adsorption, and it describes a structurally heterogeneous surface [18]. The Freundlich model is expressed in linear form as below:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (3)$$

where K_F is the Freundlich constant related to the adsorption capacity (L/mg), and $\frac{1}{n}$ is the parameter related to heterogeneity factor. The heterogeneity factor points out that adsorbate adequately adsorbs on the adsorbent ($\frac{1}{n} < 1$) or the adsorption is unfavorable ($\frac{1}{n} > 1$) [17].

Langmuir and Freundlich models in linear plots of the MV2B adsorption system on biosorbent are displayed in Figure 7. The Langmuir and Freundlich parameters were computed by the slope and the y-intercept of each corresponding linear plot and their parameters are summarized in Table 1. The results demonstrated that the Langmuir model with the highest R^2 values (0.982) is a well-behaved fit linearly for describing the MV adsorption onto biosorbent than the Freundlich model. Another useful characteristic of the Langmuir isotherm model can be determined in terms of the dimensionless factor R_L as called the Hall equilibrium separation factor, which is obtained by the following equation:

$$R_L = \frac{1}{1 + K_L C_0} \quad (4)$$

where C_i (mg/L) is the initial concentration of MV 2B in the liquid phase and K_L (L/mg) is the Langmuir constant related to the energy adsorption. The R_L factor indicates the shape of isotherm and predicts the type of adsorption process to be unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable which occurs spontaneously ($0 < R_L < 1$), or irreversible ($R_L = 0$) [27]. In this work, the calculated R_L value was found to be in a range of 0.03 to 0.75, which is well corresponding to $0 < R_L < 1$. This condition indicates the adsorption of MV 2B on biosorbent occurs spontaneously.

Table 1. Langmuir and Freundlich parameters calculated for MV 2B adsorption onto MMPB

Model	MMPB
Langmuir	
q_m (mg/g)	49.75
K_L (L/mg)	0.34
R_L	0.03 - 0.75
R^2	0.982
Freundlich	
K_F (L/mg)	9.08
$1/n$	0.71
R^2	0.957

3.6 Adsorption mechanism of MV2B with the MMPB

Based on the result of FTIR analysis, there were hydroxyl (-OH) and carbonyl (-C=O) functional groups on MMPB outer surface, which have potentiality for binding to MV 2B dye. The nitrogen atoms in dimethylaniline groups of dye interact with hydroxyl groups of MMPB via hydrogen bonding [15], play a central role in adsorption mechanism. Moreover, the ion-dipole interaction between the positive charges of MV 2B dye and the partially negative charges on oxygen atoms in carbonyl could be involved during on adsorption mechanism. The schematic of dye adsorption mechanism is proposed in Figure 8.

The removal efficiency of MV 2B dye obtained in this present work was similar to those of previous studies of hair dye and toxic metal ions [28,29]. The MMPB will be a promising biosorbent produced from the biowaste for the removal of chemical species such as cationic dyes, hair dye, and metal ions in an aqueous solution.

4. Conclusions

Modified mangosteen pericarp (MMPB) was used as a biosorbent for methyl violet 2B elimination from an aqueous solution. The study found that its surface exhibited plenty of porous structure which is appreciable for adsorbing dye molecules. In addition, various functional groups were detected on the surface of MMPB, which enhanced interaction with cationic dye molecules. The value of pH_{pzc} of MMPB was at 2.1, if the initial pH was above this value, it could be readily used without any pH adjustment. Langmuir isotherm better described the adsorption of methyl violet 2B on the MMPB, showing the maximum monolayer adsorption capacity of 49.75 mg/g. The results obtained from the batch experiments showed good effectiveness on MV 2B removal. A removal efficiency of MV2B was higher than 95% after 90 min of contact time at the amount of MMPB of 0.05 g. Based on the resulting data, it was concluded that the MMPB could satisfy the application as an inexpensive biosorbent for the removal of methyl violet dye.

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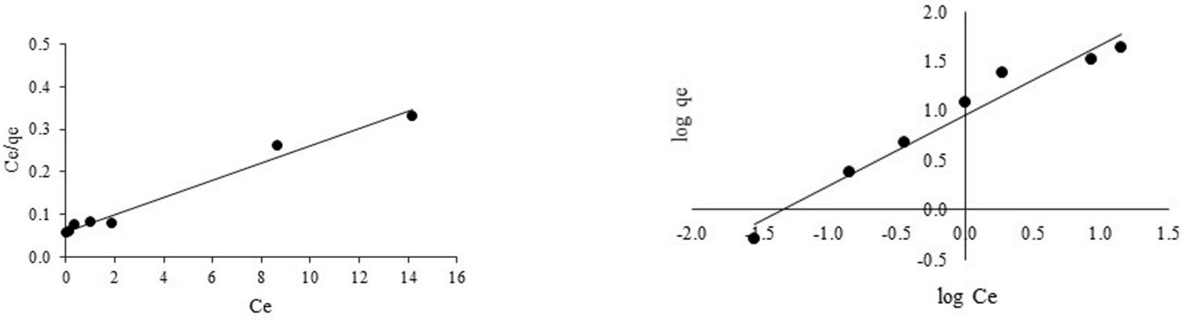

(a) (b)

Figure 7: Adsorption isotherm of MV 2B dye onto MMPB (a) Langmuir (b) Freundlich

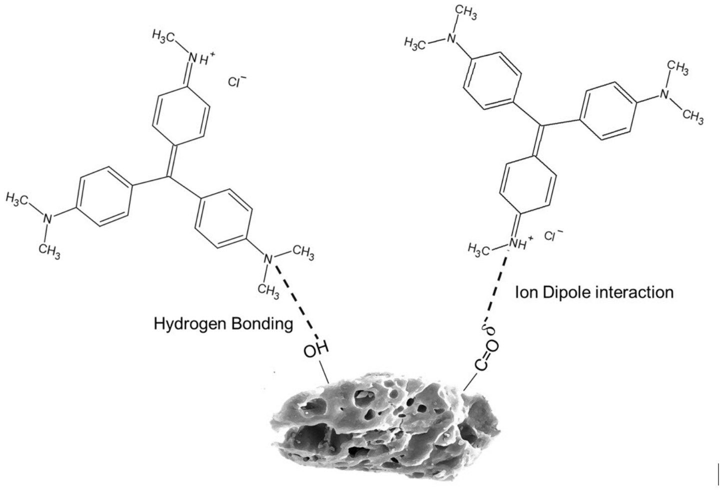

Figure 8: Schematic interaction between MV 2B dye and MMPB functional groups

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