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# Optimization of preparation conditions for mangosteen peel-based activated carbons for the removal of Remazol Brilliant Blue R using response surface methodology

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ABSTRACT

This study investigates the optimal conditions for preparation of activated carbons from mangosteen peel (MP) for removal of Remazol Brilliant Blue R (RBBR) reactive dye from aqueous solution. The MP activated carbon was prepared using physiochemical activation method which consisted of potassium hydroxide (KOH) treatment and carbon dioxide ($CO_2$) gasification. Central composite design (CCD) was used to determine the effects of the three preparation variables; $CO_2$ activation temperature, $CO_2$ activation time and KOH impregnation ratio (IR) on RBBR percentage removal and activated carbon yield. Based on the CCD, a quadratic model and a two-factor interaction (2FI) model were respectively developed for RBBR percentage removal and carbon yield. The significant factors on each experimental design response were identified from the analysis of variance (ANOVA). The optimum conditions for MP activated carbon preparation were obtained by using activation temperature of 828 °C, activation time of 1 h and IR of 3.0, which resulted in 80.35% of RBBR removal and 20.76% of activated carbon yield.



## 1. Introduction

Almost 45% of textile dyes produced worldwide belongs to the reactive class [1]. Reactive dyes are common dyes used for dyeing cellulosic fibres due to their favorable characteristics of bright color, water-fastness, simple application techniques and low energy consumption [2]. They are usually characterized by nitrogen to nitrogen double bonds (N=N azo bonds) where the color of azo dyes is due to this azo bond and the associated chromophores [3]. The formation of a covalent bond between the dye molecule and the fibre is much stronger than the physio-chemical bond between other classes of dyes and cellulose [4]. However, reactive dyes pose the greatest problem in textiles wastewater since they are not easily biodegradable, and thus color may still remain in the effluent [5]. In fact, the expanded uses of azo dyes and their reaction products such as aromatic amines are highly carcinogenic. The discharge of these wastewaters into receiving streams causes damage not only to aquatic life but also to human beings [6].

In recent years, many processes have been applied for the treatment of reactive dyes from wastewater including biological, physical and chemical process [7]. Among them, adsorption process with activated carbon has been proved to be superior compared to other techniques in terms of its simplicity of design, high efficiency and ease of operation [8]. However, the manufacturing costs of commercial activated carbons are in fact rather high. As such, there is a need to produce activated carbon with high adsorption performance from alternative material that is cheaper and readily available. From the literature, many studies have been carried out to prepare low cost activated carbons from agricultural wastes such as sugar beet pulp [2], bamboo [5], pomegranate peel [8], date stone [9], vetiver roots [10], corn grain [11], cotton stalk fibre [12], oil palm empty fruit bunch [13], coconut husk [14] and rice straw [15].

In this work, an attempt was made in preparing activated carbon from mangosteen peel (MP) precursor. Mangosteen (*Garcinia mangostana* L.), a tropical evergreen tree has been planted in Malaysia with the total acreage between 7000 and 8000 ha [16]. It is also being planted in other countries such as India, Thailand, Vietnam and Philippines. This fruit has become an economically important species since the demand in the domestic and export markets are tremendous. However, due to the high consumption of mangosteen's edible part, massive amounts of the peels are disposed, causing a severe problem in the community as they gradually ferment and release off odours [17]. Therefore, by utilizing MP into activated carbon will decrease the cost of wastes disposal and also converted this waste into value-added product [18].

Currently no study has been done on optimization of the production of activated carbon from MP using the response surface methodology (RSM) approach. RSM has been found to be a useful

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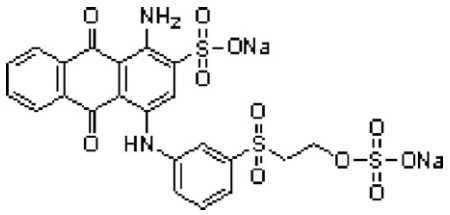

**Fig. 1.** Chemical structure of RBBR dye.

tool to study the interactions of two or more factors [19]. A standard RSM design called a central composite design (CCD) is suitable for fitting a quadratic surface and it helps to optimize the effective parameters with a minimum number of experiments, as well as to analyze the interaction between the parameters [20]. RSM has just recently been used for the optimization of activated carbon production from rattan sawdust [19], Tamarind wood [21] and Turkish lignite [22] by chemical activation whereas oil palm empty fruit bunch [13] and coconut husk [14] by physiochemical activation method. The goal of this work was to optimize the preparation conditions of activated carbon from MP for the removal of Remazol Brilliant Blue R (RBBR) dye. The effects of preparation variables; activation temperature, activation time and chemical impregnation ratio were studied simultaneously to obtain a high activated carbon yield and high RBBR percentage removal from aqueous solution using the CCD.

## 2. Materials and methods

### 2.1. Adsorbate

Remazol Brilliant Blue R (RBBR) supplied by Sigma–Aldrich (M) Sdn Bhd, Malaysia was used as an adsorbate. Deionized water was used to prepare all solutions. RBBR has a chemical formula of $C_{22}H_{16}N_2Na_2O_{11}S_3$ with molecular weight of 626.54 g mol$^{-1}$. The chemical structure of RBBR is shown in Fig. 1.

### 2.2. Preparation of activated carbon

Mangosteen peel (MP) used for preparation of activated carbon was obtained from the local market in Nibong Tebal, Penang, Malaysia. MP was firstly washed with water and subsequently dried at 105 °C for 24 h to remove the moisture contents. The dried MP was ground and sieved to the size of 1–2 mm before loaded in a stainless steel vertical tubular reactor placed in a tube furnace. Carbonization step was carried out at 700 °C for 2 h under purified nitrogen (99.99%) flow at flowrate of 150 ml min$^{-1}$. The char produced was mixed with KOH pellets with different impregnation ratio (IR), as calculated using following equation:

$$\mathrm{IR} = \frac{w_{\mathrm{KOH}}}{w_{\mathrm{char}}} \quad (1)$$

where $w_{\mathrm{KOH}}$ is the dry weight of KOH pellets (g) and $w_{\mathrm{char}}$ is the dry weight of char (g). Deionized water was then added to dissolve all the KOH pellets.

The mixture was then dehydrated in an oven at 105 °C for 24 h to remove moisture. The activation step was done using similar reactor as in carbonization step. Once the final activation temperature was reached, the gas flow was switched from nitrogen to $CO_2$ at flowrate of 150 ml min$^{-1}$ for different period of time. The activated product was then cooled to room temperature under nitrogen flow. Then, the sample was washed with hot deionized water and HCl (0.1 M) until the pH of the washed solution reached 6.5–7.

### 2.3. Adsorption studies

For batch adsorption studies, 300 mg of adsorbent were mixed with 200 ml aqueous dye solutions of 100 mg/l initial concentration in 20 sets of 250 ml Erlenmeyer flasks. The mixture was agitated at 120 rpm at 30 °C until equilibrium was reached. The pH of the solution was natural without any pH adjustment. The concentration of RBBR dye solution was determined using a double UV–vis spectrophotometer (UV-1800 Shimadzu, Japan) at maximum wavelength of 590 nm. The percentage removal at equilibrium was calculated by the following equation:

$$\text{Removal}\ (\%) = \frac{(C_o - C_e)}{C_o} \times 100 \quad (2)$$

where $C_o$ and $C_e$ are the liquid-phase dye concentrations at initial state and at equilibrium (mg/l), respectively.

### 2.4. Activated carbon yield

The activated carbon yield was calculated based on the following equation:

$$\text{Yield}\ (\%) = \frac{w_c}{w_o} \times 100 \quad (3)$$

where $w_c$ and $w_o$ are the dry weight of final activated carbon (g) and the dry weight of precursor (g), respectively.

### 2.5. Design of experiments

RSM is a collection of statistical and mathematical techniques that uses quantitative data from appropriate experiments to determine regression model equations and operating conditions which are useful for developing, improving and optimizing processes [13]. In this work, a standard RSM design, known as central composite design (CCD) was applied to study the variables for preparing the activated carbons from MP. This method can reduce the number of experimental trials needed to evaluate multiple parameters and their interactions [14]. Generally, the CCD consists of $2^n$ factorial runs, $2(n)$ axial runs and six center runs, where n is the number of factors.

In the present study, the activated carbons were prepared using physiochemical activation method where the variables studied were $CO_2$ activation temperature ($x_1$), $CO_2$ activation time ($x_2$) and KOH:char IR ($x_3$). These three variables together with their respective ranges were chosen based on the literature and preliminary studies as given in Table 1. For each categorical variable, a $2^3$ full factorial CCD for the three variables, consisting of 8 factorial points, 6 axial points and 6 replicates at the center points were employed, indicating that altogether 20 experiments for this procedure for

**Table 1**
Independent variables and their coded levels for the central composite design.

| Variables (factors) | Code | Units | Coded variable levels | | | | |
|---|---|---|---|---|---|---|---|
| | | | $-\alpha$ | −1 | 0 | +1 | $+\alpha$ |
| Activation temperature | $x_1$ | °C | 648.87 | 700.00 | 775.00 | 850.00 | 901.13 |
| Activation time | $x_2$ | h | 0.32 | 1.00 | 2.00 | 3.00 | 3.68 |
| Impregnation ratio (IR) | $x_3$ | – | 0.15 | 1.00 | 2.25 | 3.50 | 4.35 |