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Optimization of preparation conditions for activated carbons from coconut husk using response surface methodology

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Abstract

Coconut husk was used to prepare activated carbon using physiochemical activation method, consisted of potassium hydroxide (KOH) treatment and carbon dioxide (CO_2) gasification. The effects of the preparation variables which were activation temperature, activation time and chemical impregnation (KOH:char) ratio on the adsorption capacity on methylene blue dye and carbon yield were investigated. Based on the central composite design (CCD), a two factor interaction (2FI) model and a quadratic model were respectively developed to correlate the preparation variables to the adsorption capacity and yield. From the analysis of variance (ANOVA), the most influential factor on each experimental design response was identified. The predicted adsorption capacity and yield after process optimization was found to agree satisfactory with the experimental values. The optimum conditions for preparing activated carbon from coconut husk were found as follows: activation temperature of 816 °C, activation time of 1 h and KOH:char ratio of 3.9.

Keywords: Coconut husk; Activated carbon; Methylene blue; Central composite design; Optimization

1. Introduction

The presence of dyes in effluents is a major concern due to their adverse effect to many forms of life. The discharge of dyes in the environment is worrying for both toxicological and esthetical reasons [1]. Industries such as textile, leather, paper, plastics, etc., are some of the sources for dye effluents [2]. It is estimated that more than 100,000 commercially available dyes with over 7×10^5 tonnes of dye-stuff produced annually [3]. Methylene blue (MB) is the most commonly used substance for dying cotton, wood and silk. Though MB is not strongly hazardous, it can cause some harmful effects where acute exposure to MB will cause increased heart rate, vomiting, shock, Heinz body formation, cyanosis, jaundice, quadriplegia and tissue necrosis in humans [4]. Therefore, the treatment of effluent containing such dye is of interest due to its harmful impacts on the receiving waters.

In general, dyes are poorly biodegradable or resistant to environmental conditions [5]. Therefore it is necessary to treat the wastewaters or industrial effluents containing dyes before being discharged into the waterways. A number of chemical and physical processes such as flocculation, chemical coagulation, precipitation, ozonation and adsorption have been widely used to treat dye bearing wastewaters [6]. However, the adsorption onto activated carbon has been found to be superior compared to other techniques for wastewater treatment in terms of its capability for efficiently adsorbing a broad range of adsorbates and its simplicity of design. However, commercially available activated carbons are still considered expensive. This is due to the use of non-renewable and relatively expensive starting material such as coal, which is unjustified in pollution control applications [7,8]. Therefore, in recent years, this has prompted a growing research interest in the production of activated carbons from renewable and cheaper precursors which are mainly industrial and agricultural by-products, such as palm seed coat [9], rubber seed coat [10], jute fiber [11], rubber wood sawdust [12], pecan shell [13], corncob [14], bamboo [15], rattan sawdust [16] and oil palm fiber [17].

Coconut husk is the mesocarp of coconut and a coconut is consists of 33–35% of husk. In Malaysia, about 151,000 ha of

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land was being used for coconut plantation in year 2001. It was estimated that 5280 kg of dry husks were become available per hectare per year. At present, coconut husks are used as fuel for coconut processing, as a domestic fuel and as a source of fiber for rope and mats.

To make better use of this cheap and abundant agricultural waste, it is proposed to convert coconut husk into activated carbon. Conversion of coconut husk into activated carbon will serve a double purpose. First, unwanted agricultural waste is converted to useful, value-added adsorbents and second, the use of agricultural by-products represents a potential source of adsorbents which will contribute to solving part of the wastewater treatment problem in Malaysia. However, not many studies have been done on converting coconut husk into activated carbon. Some of the relevant studies found in the literature were preparation of copper impregnated coconut husk carbon for adsorption of arsenic [18], preparation of activated carbon from digested sewage sludge with the additive coconut husk using $ZnCl_2$ as activating agent [19] and production of activated carbon from coconut fiber using CO_2 and phosphoric acid activation for removal of phenol, acid red 27 dye and Cu^{2+} ions [20]. Kavitha and Namasivayam [21] has carried out a study on adsorption of methylene blue using coir pith carbon which was only subjected to carbonization and they reported that the adsorption capacity of the prepared carbon was 5.87 mg/g. The above studies have shown that coconut husk is a feasible starting material which can be used to produce activated carbons applicable in adsorbing various pollutants.

A challenge in activated carbon production is to produce very specific carbons which are suitable for certain applications. The most important characteristic of an activated carbon is its adsorption capacity which is highly influenced by the preparation conditions. In assessing the effect of treatments on quality attributes, the use of an adequate experimental design is particularly important. Response surface methodology (RSM) has been found to be a useful tool to study the interactions of two or more variables [22]. Optimization of experimental conditions using RSM was widely applied in various processes, however, its application in activated carbon production is very rare. Some of the previous studies found in applying RSM in preparation of activated carbons were using precursors such as olive-waste cakes [23], Luscar char [24] and Turkish lignite [22]. As far as known, no study has been done on preparation of activated carbon from coconut husk with physiochemical activation method consisting of KOH treatment and CO_2 gasification, by using RSM approach.

Therefore, the focus of this research was to optimize the preparation conditions of activated carbon with high carbon yield and adsorption capacity on methylene blue (MB) dye. A central composite design (CCD) was selected to study simultaneously the effects of three numerical activated carbon preparation variables: activation temperature, activation time and chemical impregnation (KOH:char) ratio, on the two responses. Empirical models correlating the MB adsorption capacity and the activated carbon yield to the three variables were then developed.

2. Materials and methods

2.1. Preparation of activated carbon

Coconut husk used for preparation of activated carbon was obtained locally. The precursor was first washed to remove dirt from its surface and was then dried overnight at 105 °C. The dried husk was crushed to desired mesh size (1–2 mm) and then carbonized at 700 °C under purified nitrogen (99.995%) flow of 150 cm^3/min for 2 h in a stainless steel vertical tubular reactor placed in a tube furnace. The heating rate was fixed at 10 °C/min. The char produced was then soaked in potassium hydroxide (KOH) solution with different impregnation (KOH:char) ratio. The mixture was then dehydrated in an oven overnight at 105 °C to remove moisture and then activated under the same condition as carbonization, but to a different final temperature. Once the final temperature was reached, the nitrogen gas flow was switched to carbon dioxide (CO_2) and activation was held for different period of time. The activated product was then cooled to room temperature and washed with hot deionized water and hydrochloric acid of 0.1 M until the pH of the washing solution reached 6–7 [17].

2.2. Design of experiments

The parameters for preparing the activated carbon was studied with a standard response surface methodology (RSM) design called a central composite design (CCD). This method is suitable for fitting a quadratic surface and it helps to optimize the effective parameters with a minimum number of experiments, and also to analyze the interaction between the parameters [24]. RSM is a collection of mathematical and statistical techniques that are useful for modeling and analysis of problems in which a response of interest is influenced by several variables [25]. Generally, the CCD consists of a 2^n factorial runs with $2n$ axial runs and n_c center runs (six replicates).

In this study, the activated carbons were prepared using physiochemical activation method by varying the preparation variables using the CCD. The variables studied were (i) x_1, activation temperature; (ii) x_2, activation time and (iii) x_3, KOH impregnation ratio. These three variables together with their respective ranges were chosen based on the literature and some preliminary studies. Activation temperature, activation time and chemical impregnation ratio were found to be important parameters affecting the characteristics of the activated carbons produced [19,26–29].

For each categorical variable, a 2^3 full factorial central composite design for the three variables, consisting of 8 factorial points, 6 axial points and 6 replicates at the centre points were employed, indicating that altogether 20 experiments were required, as calculated from Eq. (1) [24]:

$$N = 2^n + 2n + n_c = 2^3 + 2 \times 3 + 6 = 20 \quad (1)$$

where N is the total number of experiments required and n is the number of factors.