Chapter 19
Adsorption of Dyes in Aqueous Medium Using RHA and CFA: Effect of Preparation Methods and Process Optimization

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ABSTRACT

The preparation of adsorbent from the mixture of rice husk ash (RHA) and coal fly ash (CFA) has been investigated for adsorption of acid violet 7 (AV7) and brilliant green (BG) dyes. The RHA-CFA adsorbents were prepared using three different methods, i.e. reflux, magnetic co-precipitation, and magnetic template. Five different additives were used in reflux method. The results showed that RHA-CFA adsorbent prepared through reflux methods using NaOH and Na2CO3 shows higher dyes adsorption removal as compared to other methods. From zeta potential analysis, the electric charge of the outer layer of prepared adsorbent shows no effect towards adsorption of AV7 and BG dyes. By using a 3-factor, 3-level factorial design, the relationship between all variables was studied. From the response surface models, the optimum adsorbent preparation variables could be obtained by using RHA-CFA adsorbent prepared by refluxing 3:1 ratio of RHA to CFA in 1.21 M NaOH solution. The results indicated that the optimized values agree reasonably well with the validated experimental results.

INTRODUCTION

Environmental pollution control has been a crucial concern to every country. In Malaysia, water pollution and solid waste disposal from many industries have become a major problem over the last decade. Water pollution usually caused by the effluent that comes from industrial and domestic sewage. One of the concerns in wastewater pollution is the presence of coloured compounds such as dyes. For industrial

DOI: 10.4018/978-1-7998-0369-0.ch019

usage, synthetic dyes are usually preferable because of its complex aromatic structures that provide resistance to light, biological activity, ozone and another degradative environment condition (Kaushik & Malik, 2014; Holkar *et al*., 2016). Acid violet 7 (AV7) and brilliant green (BG) dyes are the example of synthetic dyes that are being used in textile and leather, paper, automotive, plastic, soap, wood, medicine and cosmetics of dyeing as well as in biological dyeing (Kumar *et al*., 2012; Mansour *et al*., 2009; Tavlieva *et al*., 2013). Both dyes have complex and stable structure with carcinogenic, neurotoxicity and mutagenic properties. Therefore, the removal of these two types of dyes become important as the toxic compounds can endanger water organisms while the excess colourant can change the natural colour of water bodies.

Adsorption is one of the methods that has been used for removal of AV7 and BG dyes. Despite the widely used of commercial activated carbon (CAC) for removal of dye-containing wastewater, studies on low-cost materials such as industrial and agricultural solids wastes have been extensively performed to replace the CAC. These including rice husk ash (RHA), palm oil fuel ash (PFA) and coal fly ash (CFA) which are abundantly available through the combustion process of rice mill, palm oil mill and coal-fired power plant industries, respectively. Typically, for every 100 kg of paddy milled, about 20 kg (20%) of husk is being produced and after the husk being burnt, around 18% of ash (RHA) will be generated (Bronzeoak, 2003). From this conversion, it was estimated that more than 27.7 million tons of RHA is produced annually worldwide, of which Malaysia alone produces more than 104.4 thousand tons of RHA annually (FAO, 2019). While, PFA is a by-product produced by countries having palm oil industry such as Indonesia and Malaysia. This waste ash is generated by the incineration of palm oil fibers, empty fruit bunches and shells as fuel in palm oil mill boilers. By this incineration process, about 5% by weight of the PFA is produced. The quantity of PFA produced in Malaysia alone is about 4 million tons a year (Awang *et al.*, 2014). Meanwhile, coal fired thermal power plants have generated huge amount of coal bottom ash (CBA) and coal fly ash (CFA). Typically, CBA forms up to 25% - 90% of total ash generated while the remaining are CFA (Sadon *et al.*, 2017). Since 1988, Malaysia produces about 8.5 million tons of coal ash (Rafieizonooz *et al.*, 2016; Alhokabi & Ing, 2019).

Even though RHA, PFA, and CFA have been used (with or without modification) and possess good adsorption capacity towards dyes, however nearly all of studies only use one form of ash. Previously, we had reported the characteristics/properties of these ashes and adsorption capacity of adsorbent prepared from the mixture of rice husk ash (RHA), palm oil fuel ash (PFA) and coal fly ash (CFA). We have used this type of adsorbent (i.e. RHA/PFA/CFA sorbent) in removing heavy metals (Dahlan & Ismail, 2012; Dahlan & Razali, 2012; Dahlan *et al*., 2013; Dahlan & Zwain, 2013; Zwain *et al*., 2018) and dyes (Dahlan & Ismail, 2012; Dahlan & Noor, 2014). Nevertheless, our previous reports only dealt with activity measurement related to adsorbent preparation conditions by single method and effects of batch operating conditions. Less information is available about the effects of the various preparation methods for this kind of adsorbent. A facile one-step refluxing route in alkaline solution has been used as a method to prepare a magnetic activated carbon/$CoFe_2O_4$ composite (Ai *et al.*, 2010). This magnetic adsorbent has been used for the removal of malachite green dye from water. Their results indicated that $CoFe_2O_4$ particles deposited on the surface of activated carbon were uniform, with the particle size in the range of 14–20 nm. The preparation of adsorbent derived from pine cone has also been successfully synthesized by sulfuric acid reflux method. With sulfuric acid reflux, the pine cones undergone carbonization as well as functionalization with sulfonic acid groups. The adsorbent demonstrated high adsorption capacity for two emerging organic pollutants, *i.e.* methylene blue and tetracycline (Islam *et al.*, 2018). While Zhang *et al.* (2007) used magnetic co-precipitation method to prepare $CuFe_2O_4$/activated carbon