



Thermally processed sewage sludge for methylene blue uptake



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ARTICLE INFO

Article history:

Received 27 September 2012

Received in revised form

5 December 2012

Accepted 6 December 2012

Available online 11 January 2013

Keywords:

Adsorption

Activated carbon

Methylene blue

Sewage sludge

ABSTRACT

Sewage sludge, with zero recycling rate, poses landfill problems as Semakau island, the dumping site, is rapidly occupied in within a short period of time. Dried sewage sludge was heated at different temperatures ranging from 200 °C to 700 °C over various heating times of 2–5 h. The heated dried sludge (DS) was equilibrated with methylene blue (MB) solutions at different concentration to determine the optimum adsorption time, effect of different heating temperatures of sludge, effect of pH and effect of adsorbent concentration on the percentage of MB adsorbed. The results were compared to a commercial activated carbon (AC) in adsorbing the same MB concentration. The higher heating temperatures the more the DS was able to adsorb MB. DS, which was heated at 700 °C, showed the highest percentage of MB adsorbed of 95%, compared to that of AC, which were 91.20% (Grade 17) and 97.99% (Grade 19), when adsorbing the same initial MB concentration of 10 μmol l⁻¹. However, DS heated at 200 °C for 5 h (DS-200-5) was found to have a ratio of MB adsorbed per power consumption of 21.568 (μmol l⁻¹ kW), the highest ratio among the other sludge samples, making it the most cost effective type of sludge. Overall, DS was able to adsorb MB between 90% and 98% at a wide pH range of 3 < pH < 12, showing the potentiality of DS to remove contaminant from industrial discharge. In addition the reuse of sludge for other environmental application can help to prolong the lifespan of landfill dumping site.

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1. Introduction

Discharge from industrial plants usually consists of many unwanted effluents, and part of it comes from organic dyes. Textile, leather, food processing, dyeing, cosmetics, paper and dye manufacturing industries are contributing sources of dye pollution Sarioglu and Atay (2006). At the moment, approximately 8000 chemical problems are known to be associated with the dyeing process and 100,000 are commercially available. Annually, 700,000 tons of dye related stuff is produced in the world Prasad and Rao (2010). MB has been found to be one of the common dyes used in the textile industry Lazaro et al. (2011). Conventional methods, such as activated sludge treatment and membrane filtration, have been unsuccessful in removing these dyes from industrial wastes and although granular activated carbon (AC) has been proven to be able to remove these contaminants effectively, its main drawback lies in its high energy cost. The price of granular AC ranges between US\$ 1500–US\$ 3000 per ton (Alibaba, 1999–2012). Several low cost adsorbent materials were also researched upon and tested for MB adsorption capacities, such

examples includes pistachio shells Vasconcelos et al. (2010), coffee ground Reffas et al. (2010) and wood waste Djati Utomo et al. (in press). They are often commonly regarded as waste in society but they can be alternative low cost methods in addressing dye pollution. Adsorbent of pistachio shells and wood waste have shown good adsorption capacity and are effective in reducing MB significantly, with 74% and 98% removal efficiency respectively (Vasconcelos et al., 2010; Djati Utomo et al., in press). The latter has also shown to surpass AC extensively.

Of all the waste generated around the world, it is estimated that 46 billion metric tonnes of sewage sludge, which is the natural by-product of the wastewater treatment process, are produced globally annually Lux Research (2009). In 2011, Singapore was reported to have produced about 6.9 million tonnes of natural waste with a recycling rate of 59% NEA (2011). Out of the total natural waste in Singapore, 153 kt of sewage sludge were produced annually and it is usually incinerated before being transported to the only landfill at Pulau Semakau, where it is buried under the earth at the landfill Spinosa (2007). Semakau landfill, an offshore landfill located 8 km south of mainland Singapore, covers a land area of 350 ha with a landfill capacity of 63 million m³ NLB (2012). As sludge is the only waste with 0% recycling rate NEA (2011), this non-existent recycling rate poses landfill problems as land would be rapidly occupied in within a short period of time i.e. 30 years NLB (2012). In comparison

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to countries like Japan, sewage sludge is used as a raw material for Portland cement Onaka (2000), and as fertilisers in agriculture in the US National Research Council (1997). Hence, instead of just disposing them, this study requires and focuses on the utilisation of sludge in Singapore through recycling which can help prolong the lifespan of Pulau Semakau.

The objectives of this study is to investigate sewage sludge, obtained from a water reclamation plant, to remove MB in a water medium which could turn this natural waste into a useful value-added product due to its abundance as it is readily available in water reclamation plants, and low processing cost. At the same time, it can prolong the lifespan of Pulau Semakau before incineration and land filling of the sludge.

2. Materials and methods

A bag containing 10 kg of sludge sample, 90%–95% dried, was obtained directly from Changi Water Reclamation Plant (CWRP), Singapore. Twenty of 500 g–600 g of sludge samples were wrapped with aluminium foil and weighed accurately. Aluminium foil was used so as to trap heat within the sludge for more effective heating. The samples were then placed in a furnace (Carbolite Asphalt Binder Analyser Model ABA 7/35) and heated at different temperatures, 200 °C, 300 °C, 400 °C, 500 °C and 700 °C, over 2–5 h. All the dried samples were grinded and sieved through a 600 µm sieve pan (ASTM E11), to achieve a uniform size of sample of DS. The grinded sludge was then stored in labelled plastic containers. For simplicity the type of DS will be coded DS-T-t referring to the dried sludge heated at T (°C) heating temperature and t (h) heating time. The morphology of DS was analysed using scanning electron microscope (SEM JSM 7001F) following Costa et al. (2009). Commercial AC made from coconut shell was purchased from a local manufacturing company with code GR 17 and GR 19.

MB stock solution of 10 µmol l⁻¹ was prepared by accurately weighing 0.0019 g of powdered MB. It was then dissolved completely in 500 ml of deionised water and transferred into a 500 ml storage bottle to make a 10 µmol l⁻¹ MB solution. The storage bottle was wrapped with aluminium foil to prevent decolourisation of the solution affected by surrounding light.

Various volumes of 4 ml, 8 ml, 12 ml, 20 ml and 25 ml of 10 µmol l⁻¹ of MB stock solution were pipetted and poured in 25 ml volumetric flasks. The solutions were topped up with de-ionised water to obtain MB standard concentrations of 1.6 µmol l⁻¹, 3.2 µmol l⁻¹, 4.8 µmol l⁻¹, 8.0 µmol l⁻¹ and 10.0 µmol l⁻¹ respectively. The absorbance of the various MB standard solutions was measured using a UV-spectrophotometer (Tech Comp UV 1100 Spectrophotometer), at two reference wavelengths of 609 nm Bangash and Manaf (2005) and 665 nm Pasha and Narayana (2008). Both wavelengths were compared based on its regression value, and 609 nm was found to have the best regression value ($R^2 \approx 1$) Djati Utomo et al. (in press). Thus, 609 nm was selected for the study. Calibration work was conducted before the start of every batch adsorption experiment.

Various amount of sludge was weighed using a four-decimal-places mass balance and placed into 12 ml test polypropylene tubes before being filled with 10 ml of 10 µmol l⁻¹ MB solution. The mixtures were mixed thoroughly using an adjustable rotator (Digital Feedback Controlled Rotator RT-10) at 30 rpm. To separate DS from the unadsorbed MB, the mixtures were subsequently centrifuged in the centrifuge machine (Jouan Centrifuge, BB 3V) at 1000 rpm for 10 min. The supernatant liquid was then extracted using a dropper and tested for its final absorbance using UV-spectrophotometer. For the study of pH effect, required amount of concentrated HCL and NaOH were added to 10 µmol l⁻¹ of MB solutions, before the addition of 50 g l⁻¹ of sludge samples into the test tubes.

The percentage of MB adsorbed was found for each sample of adsorbent at the same equilibrium points using the formula below (Khan et al., 2005; Nameni et al., 2008):

$$\% \text{MB adsorbed} = \frac{C_i - C_f}{C_f} \times 100\% \quad [1]$$

Where,

$$C_i = \text{initial concentration of MB solution (mg L}^{-1}\text{)}$$

$$C_f = \text{final concentration of MB solution (mg L}^{-1}\text{)}$$

The adsorption data was analysed by fitting them to Langmuir and Freundlich equations. Each linearized equation is presented below:

Langmuir isotherm:

$$\frac{1}{X} = \frac{1}{bC_e X_m} + \frac{1}{X_m} \quad [2]$$

Where X (mg g⁻¹) is the amount of MB adsorbed per gram of DS-T-t and X_m (mg g⁻¹) is the monolayer capacity of DS in adsorbing MB. The constant of b indicates the equilibrium binding constant of the MB adsorption (l mg⁻¹) and C_e is the MB equilibrium concentration (mg l⁻¹).

Freundlich isotherm:

$$\log(x/m) = \log k + 1/n \log C \quad [3]$$

Where x/m (mg g⁻¹) is the amount of MB adsorbed per gram of DS-T-t, C is the MB equilibrium concentration (mg l⁻¹), whereas k and $1/n$ are equilibrium constants that indicate a measure of multi-layer adsorption capacity and intensity of adsorption respectively Khan et al. (2005). The plots of $1/X$ versus $1/C_e$ and $\log(x/m)$ versus $\log C$ were made to test the data fitness to the Langmuir and Freundlich adsorption models, respectively.

Experimental analysis of sample was setup in triplicates and relative standard deviation (RSD) was calculated for each result. Any triplicate set with an RSD greater than 5% was rejected and such experiments were repeated to get a better reliability.

3. Results and discussions

3.1. Mass difference of sludge before and after heating

The process of heating DS was conducted at different temperatures and time. Fig. 1 showed the percentage of mass loss of sludge at various heating temperatures.

A general relationship between the percentage of mass loss of sludge and the heating temperature can be observed in Fig. 1. The percentage of mass loss of sludge steadily increased from 0 °C to 300 °C. At heating temperatures between 300 °C and 500 °C, the increase in percentage loss became significant before reaching a plateau after approximately 700 °C. Some inconsistency in the percentage of mass loss with increasing heating time was inconclusively observed.

3.2. Adsorption time of MB onto heated DS

To ensure that adequate time was given for the adsorption of MB onto DS, the optimum adsorption time of MB onto DS was determined. A reasonable adsorption time will then be used for the rest of the experiment. The results of two types of DS-200-t and DS-700-t are shown in Fig. 2.

Fig. 2 showed that the rate of adsorption of MB onto heated sludge is fast as it reached approximately 80%–95% for various

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