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## The Adsorption Study of Methylene Blue onto MgO from Various Preparation Methods

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### ABSTRACT

The investigation of Methylene Blue (MB) removal in solution by Magnesium Oxide (MgO) from various preparation methods was studied. Then, these adsorbents were characterized for the surface morphology using Scanning Electron Microscopy (SEM). Moreover, the effect of preparation parameters in MB adsorption onto resulted MgO such as aging time, drying temperature and calcinations temperature was also examined. The MgO was the precipitation of  $\text{Mg}(\text{OH})_2$  by using  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  as Mg metal source and using Oxalic acid, Urea- $\text{NH}_4\text{OH}$ , Polyvinly alcohol (PVA)/ $\text{NH}_4\text{OH}$  and only  $\text{NH}_4\text{OH}$  as precipitant which then assigned as MgO-Oxalic, MgO-Urea, MgO-PVA, MgO- $\text{NH}_4\text{OH}$ , respectively. The results indicated that MgO-Urea showed the highest MB adsorption capacity when compared to other MgO adsorbents. The SEM micrograph exhibited the different particle size and resulted that the finer particle size showed the higher MB adsorption capacity. The optimize conditions to prepare MgO-Urea for MB adsorption were aging time at  $60^\circ\text{C}$  for 6 h, dried at  $100^\circ\text{C}$  and calcined at  $300\text{-}500^\circ\text{C}$ .

**Key words:** MgO, methylene blue, adsorption, scanning electron microscopy, surface morphology

### INTRODUCTION

Now-a-days, the technology is rapidly developing which is the main reason in environmental problem. Dyes have been applied in many industries such as textiles, printing and publication, pulp mills, leather, food and plastics, etc. The effluent from these industries normally contains some of dyes. The estimation of 10,000 different commercial dyes and pigments exist and more than  $7 \times 10^5$  tons are produced per year world-wide. It has been about 10-15% of these dyes are released after dyeing process (Al-Degs *et al.*, 2008). Since many organic dyestuffs are harmful to human being and toxic to animal and microorganism. The dye removal has been in considerable attention over the past decades for dye treatment in the wastewater before release to natural stream. Various dye removal process have been used such as coagulation, chemical oxidation, membrane separation, electrochemical process, biological treatment (Durai and Rajasimman, 2011) and adsorption techniques. All techniques have their own advantages and limitations such as the adsorption process, the cost and recycling are the limitation. However, adsorption was recognized to be an effective process for removal of dyes from waste effluents (Orthman *et al.*, 2003) which are the easiness in separation after process and high effective in dye removal. Many kinds of adsorbents have been developed for various applications such as activated carbon (Iqbal and Ashiq, 2007), active carbon from Pyrolysis of Bagasse (Lori *et al.*, 2008), spent activated clay

(Weng and Pan, 2007), sand (Rauf *et al.*, 2008), soil (Cheng *et al.*, 2008), kaolin (Nandi *et al.*, 2009), biomass from *Calotropis procera* leaf (Ali and Muhammad, 2008), natural resin (Clinoptilolite) (Moazed, 2008) and Mg-Al-CO<sub>3</sub> layers (El-Gaini *et al.*, 2009).

MgO has been widely used in many applications such as catalyst, catalyst support and destructive adsorbent for many kinds of pollutants. The structure of MgO is a simple sodium-chloride structure and is very stable thermally and stoichiometrically. The highly active MgO nanoparticles with high surface area have been obtained with specific preparation method and the MgO prepared from different routes has different morphologies and properties. These could lead to different activities and selectivity of resulting MgO. Other factors such as heating temperature, treatment times, pH, gelling agent (precipitant) and atmosphere during preparation could also affect the properties of final product (Liang and Gay, 1985; Klabunde *et al.*, 1996).

Different synthetic method could provide different nanoscale MgO such as sol gel (Chadwick *et al.*, 1998), hydrothermal (Ding *et al.*, 2001) flam spray pyrolysis (Bickmore *et al.*, 1996) laser vaporization (El-Shall *et al.*, 1998) chemical gas phase deposition (Matthews *et al.*, 2000) combustion aerosol synthesis (Helble, 1998) aqueous wet chemical (Bhargava *et al.*, 1998) and surfactant method etc. Moreover, high surface area MgO could promote from simple and conventional method which is the decomposition of magnesium salt and precipitated by various base (Khairallah and Glisenti, 2007). In this study, the MgO from various precipitation methods which were PVA template deposition, urea gelation and deposition by NH<sub>4</sub>OH and oxalic acid was studied by MB adsorption. The characterization of various MgO was also examined by SEM. Then, the parameters during the MgO-Urea preparation were also investigated; including aging time, drying temperature and calcinations temperature.

## **MATERIALS AND METHODS**

The present study was conducted at Department of Chemistry, Faculty of Sciences, Maharakham University during the academic year 2010.

### **MgO preparation**

**PVA template precipitation:** It has been proposed by Meshkani and Rezaei (2009) to obtain nano-scaled MgO.

**Precipitation by NH<sub>4</sub>OH:** It was also called as aqueous wet chemical method (Khairallah and Glisenti, 2007).

**Urea gelation precipitation:** Urea gelation precipitation had also been used. In this case aqueous solutions of Mg(NO<sub>3</sub>)<sub>2</sub>.6H<sub>2</sub>O and urea were mixed and heated up to 60°C under continuous stirring. An aqueous solution of NH<sub>4</sub>OH was added dropwise and the resulting precipitate was aged at 60°C for 3 h.

**Deposition by oxalic acid in ethanol:** The synthesis of MgO with oxalic acid was followed the route in Kumar and Kumar (2008).

**Adsorption measurement:** The process of adsorption measurement of MB onto MgO was carried out as follow method. Firstly, different concentration of MB was prepared (0.4×10<sup>-5</sup>-6×10<sup>-5</sup> M). All MB solution was then adjusted pH to 5 by using either 1 M HCl or 1 M NaOH. Then, 250 mL of

each MB solution was distributed into 500 mL round bottle under well stirring at 45°C. The MgO was weighted and loaded into adjusted pH of MB solution in ratio 0.2 g L<sup>-1</sup>. The mixture was stirred for one h to obtain the saturation adsorption and then the mixture was centrifuged to obtain the clear blue solution. This supernatant was determined the residual MB concentration by UV-V is spectrophotometer (Perkin-Elmer; Lambda 25) at wave number 650 nm. In order to study the effect of preparation method, the different MgO was used in the adsorption measurement which followed the similar method as mention above.

**Calculation:** The initial dye concentration, C<sub>0</sub> (M) and equilibrium dye concentration, C<sub>t</sub> (M) were calculated from peak height at wave number 650 cm<sup>-1</sup> of UV-vis spectra by using standard curve of MB. The dye removal per unit weight of MgO at time t, q<sub>t</sub> (mg g<sup>-1</sup>) were obtained from Eq. 1:

$$q_t = (C_0 - C_t)V/M \quad (1)$$

where, V is the volume of dye solution in adsorption process (l) and M is the mass of MgO (g).

## RESULTS

**The effect of preparation method:** The surface morphology of 4 different MgO was studied by using SEM as showed in Fig. 1. The MgO-Urea sample clearly appears in smaller particle size than MgO-NH<sub>4</sub>OH and MgO-PVA whereas the particle size of MgO-Urea is not much different from MgO-Oxalic but the distribution of particle is higher. The SEM scan of MgO-PVA contains 2 different shapes which could be large particle of MgO and some maintained PVA in sample.

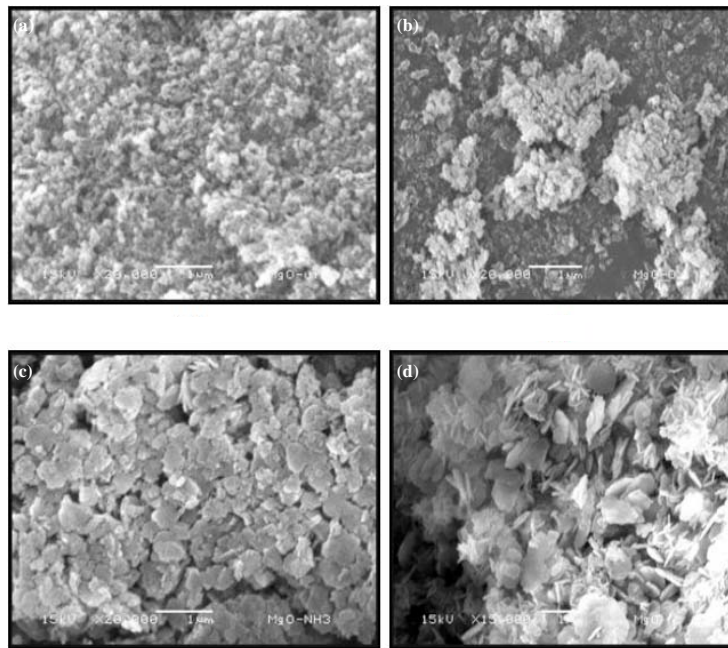


Fig. 1(a-d): SEM micrograph of (a) MgO-Urea (b) MgO-Oxalic (c) MgO-NH<sub>4</sub>OH and (d) MgO-PVA

Figure 2 showed the effect of preparation method of MgO by using urea-NH<sub>4</sub>OH, Oxalic acid, PVA-NH<sub>4</sub>OH and only NH<sub>4</sub>OH. The pH of solution during the preparation was about 9-10 by adding NH<sub>4</sub>OH drop wise. The resulting slurry was then filtrated, dried and calcined at 100 and 500°C, then tested the MB adsorption as the results exhibited in Fig. 2. The resulting MgO obtained by using urea-NH<sub>4</sub>OH (MgO-Urea) is the most effective material to removal MB in solution and followed by MgO-Oxalic, MgO-NH<sub>4</sub>OH and MgO-PVA, respectively.

**The effect of aging time of MgO-Urea:** The aging time during the stirring precipitation mixture was suspected to affect the MB removal capacity of resulted MgO. Figure 3 showed the influence of aging time during urea gelation preparation. When slurry mixture was completely obtained, the mixture was aged for 1-9 h. The results showed that the optimum of aging time was 6 h, followed by 3, 1 and 9 h. The adsorption was reached equilibrium at about 0.06 mmol g<sup>-1</sup> for 6 h aging time sample whereas only 0.025 and 0.03 mmol g<sup>-1</sup> for the sample aged for 1 and 9 h, respectively.

**The effect of drying and calcination temperature over MgO-Urea:** The urea gelation precipitation was the selected preparation to prepare the soaked Mg(OH)<sub>2</sub> for studying the effect of drying method. After the precipitation was completed, the mixture was washed with DI water and filtrated to obtain the soaked Mg(OH)<sub>2</sub>. Then the sample was dried at different temperature; 40, 70, 100 and 130°C for overnight. These dried samples were calcined at 500°C for 4 h and then these resulted 5 samples were test the efficiency of MB adsorption. Figure 4 showed the effect

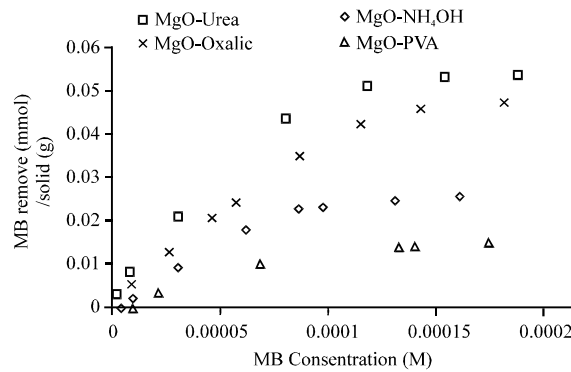


Fig. 2: Effect of preparation method of MgO in MB removal

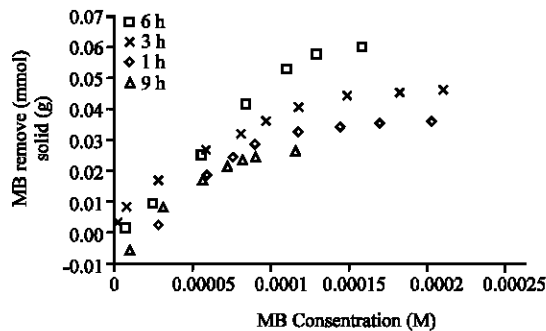


Fig. 3: Effect of aging time during MgO-Urea preparation in MB removal

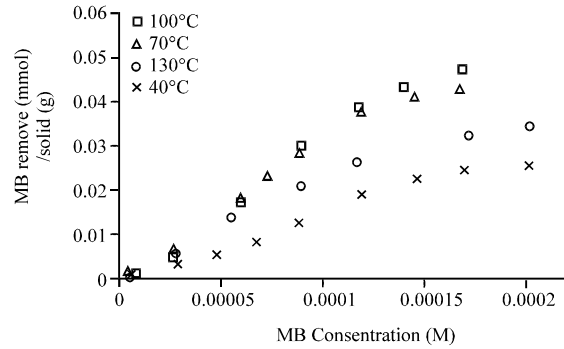


Fig. 4: Effect of drying temperature of MgO gel from urea gelation precipitation in MB removal

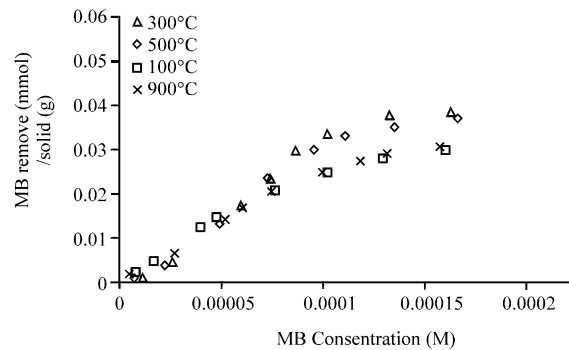


Fig. 5: Effect of calcination temperature of MgO from urea gelation precipitation in MB removal

of drying temperature in MB removal of sample which dried at 40, 70, 100 and 130°C for overnight. The results clearly showed that the drying temperature at 70-100°C was the sufficient temperature to dry the resulting cake. The sample that dried at 40°C showed the lowest MB adsorption capacity which only 0.025 mmol g<sup>-1</sup> at saturation stage.

Moreover, the effect of calcinations temperature was also carried out. All five samples were dried at 100°C for overnight and then calcined at different temperature; 100, 300, 500, 700 and 900°C for 4 h before studied the MB removal capacity. Figure 5 illustrated the effect of calcination temperature over the dried Mg(OH)<sub>2</sub> at 100°C which prepared from urea gelation precipitation. The MB adsorption activity of calcined MgO was not much different. The sufficient temperature to calcine the dried MgO was between 300-500°C and the adsorption capacity was about 0.05 mmol g<sup>-1</sup> at saturation stage.

## DISCUSSION

**The effect of preparation method:** The different chemicals were used in the preparation method with different hypothesis. In the case of metal oxide preparation using water soluble metal salt with urea, during addition of base solution such as NaOH or NH<sub>4</sub>OH was known as homogeneous precipitation (Subrt *et al.*, 2006). At initial step, the solution of metal salt and urea was in acid condition. Then, when starting to adding the precipitant (NaOH or NH<sub>4</sub>OH), the pH was slowly increasing whereas without the urea in solution, pH was increase more quickly. When pH of

mixture increased slowly, this resulted in the fine particle was occurred. This could explain by the decomposition of urea ( $\text{CH}_4\text{N}_2\text{O}$ ) in aqueous solution is accompanied by slow and controlled supply of ammonia ( $\text{NH}_3$ ) and carbon dioxide ( $\text{CO}_2$ ) into solution. The smooth pH increase obtained by the degradation of urea in synchrony with the active release of  $\text{OH}^-$  and  $\text{CO}_3^{2-}$  ions, usually pH around 8-10 leads to the precipitation of metal hydroxide particles. This phenomena during metal hydroxide precipitation resulted in better crystallinity, regular shape and nano-size of particles (Sohn *et al.*, 2004). The addition of oxalic acid in ethanol to forming the  $\text{Mg}(\text{OH})_2$  gel was expected that the oxalic acid solution could reduce the sintering of MgO during drying and calcination step or stabilized the resulting MgO in thermal treatment (Kumar and Kumar, 2008). This could result in lower agglomeration MgO which enhanced by thermal and achieved the finer particle of MgO. However, when using only  $\text{NH}_4\text{OH}$  (no-urea adding), the pH during the preparation was increased faster and resulted in particle formation occurred faster. It related to larger particle of MgO as also found in SEM micrograph (Fig. 1). It was also clearly that the smaller particle of MgO was obtained in MgO-Urea sample. Therefore, the particle size could be reason to explain the higher adsorption capacity of MgO-urea in MB removal as found in Fig. 2. Moreover, PVA was also used in MgO preparation in this study. Meshkani and Rezaei (2009) suggested that PVA could act as polymeric surfactant which dispersed the precipitant  $\text{Mg}(\text{OH})_2$  in polymer matrix of PVA during precipitation. This could correlate to the fine particle and reduce the agglomeration during drying and calcination step.

From Fig. 1, the particle size of MgO-Oxalic was finer than MgO- $\text{NH}_4\text{OH}$  and MgO-PVA, respectively. Moreover, the MB removal capacity over MgO-Oxalic was also higher than MgO- $\text{NH}_4\text{OH}$  and MgO-PVA, respectively (Fig. 2). Therefore, this could be up to the fine particle of MgO-Oxalic sample which resulted from the protection of high thermal over the sample by using Oxalic acid in preparation. This could enhance the adsorption of MB over MgO-Oxalic sample.

However, Fig. 1 showed that this sample was the lowest MB adsorption efficiency. It could be explained that the adsorption of MB might not suitable with the surface of resulted MgO-PVA from this method or other preparation parameter using in this study might not the optimization condition for MgO-PVA route which resulting in unpreferable surface sites for MB adsorption.

**The effect of aging time over MgO-Urea:** Esmaeili *et al.* (2009) also studied the effect of aging time over MgO preparation using microwave-induced process. They found that the surface area of MgO was went up to  $400 \text{ m}^2 \text{ g}^{-1}$  at aging time 20 min and then decreased when increased aging time. They explain that for longer period of time the particle may grow and Ostwald ripening may occur, resulting in larger particle grows at the expense of smaller ones. The effect of aging time during the preparation of MgO-Urea was also investigated by using MB removal as a tool. The solution of Mg nitrate salt and urea was mixed and the slurry was obtained after  $\text{NH}_4\text{OH}$  was added dropwise. This slurry was then aged for 1-9 h and the results showed that 6 h is the optimum aging time. In the present study, the similar results as found in Esmaeili *et al.* (2009) were obtained in Fig. 3 which could explain by the growth of larger particle when aged the mixture longer than 6 h whereas aging time less than 6 h could not sufficient time to allow the well particle growth in solution.

**The effect of drying and calcination temperature over MgO-Urea:** During the drying step was studied by Meshkani and Rezaei (2009). The as-prepared Mg hydroxide was heated from RT to  $800^\circ\text{C}$  and the weight loss was corrected by using TGA/DTG technique. The results showed weight loss at below  $100^\circ\text{C}$  is assigned to the loss of free water and the next weight loss at

300-450°C is correlated to the decomposition of  $\text{Mg}(\text{OH})_2$  to  $\text{MgO}$ . They also studied the effect of calcinations, it showed that  $\text{MgO}$  calcined at 700-800°C related to bigger crystallite size than sample calcined at 600°C. In term of surface area, Esmaili *et al.* (2009) showed that BET surface area of  $\text{MgO}$  gradually increase when increase calcined temperature to 400°C and then decreases when continue increase calcined temperature. The particle size or crystallite size was also affected by sintering of  $\text{MgO}$  by overheated as found in Kumar and Kumar (2008), decomposition temperature increase from 500-1000°C resulted in crystallite size increase from 6.5-73.5 nm.

The effect of drying temperature in this study, the filtrated precipitate of urea gelation precipitation was then dried at temperature between 40-130°C and followed by calcined at 500°C. The obtained  $\text{MgO}$  was then tested the MB adsorption and the results showed in Fig. 4. The drying temperature between 70 to 100°C was resulted as the sufficient temperature to dry the precipitate and exhibited high MB adsorption whereas the drying at 130°C was too high thermal and the excess thermal could sintering resulting  $\text{Mg}(\text{OH})_2$ . This is the cause of the agglomeration of  $\text{MgO}$  and the final  $\text{MgO}$  will be larger particle. The overheating was explained similarly in the calcinations temperature effect as following part (Meshkani and Rezaei, 2009; Kumar and Kumar, 2008; Esmaili *et al.*, 2009). For the sample dried at 40°C showed the lowest MB removal, this was due to the exhibited too low thermal. Since in drying step was needed the sufficient thermal to remove free water from the precipitate.

The dried sample was also tried to calcined at 100°C and it showed low MB adsorption property which could related to the small amount of  $\text{MgO}$  was formed. Moreover, the dried sample was calcined at high temperature, 900°C, the results showed similarly low MB removal as 100°C calcination. This temperature was generated high thermal enough to change all  $\text{Mg}(\text{OH})_2$  to  $\text{MgO}$ . Nevertheless, the side effect of this high thermal could enhance the sintering of  $\text{MgO}$ . Therefore, in the present study, the  $\text{MgO}$  calcined at 900°C could sintering from high thermal and affect the surface area and crystallite size which then showed the low MB adsorption capacity. The effect of calcination temperature showed in Fig. 5 which clearly illustrated that the temperature 300-500°C suitable to second heat the sample and obtain the high MB adsorption capacity. This was supported from the reason that the dried sample need the temperature at least 300°C to achieve phase transition of  $\text{Mg}(\text{OH})_2$  to  $\text{MgO}$ .

## CONCLUSION

In this study, the various  $\text{MgO}$  was used as the adsorbent for MB removal. The results showed that  $\text{MgO}$ -Urea sample showed the highest MB adsorption capacity and followed by  $\text{MgO}$ -oxalic,  $\text{MgO}$ - $\text{NH}_4\text{OH}$  and  $\text{MgO}$ -PVA, respectively. The effect of preparation over  $\text{MgO}$ -Urea sample such as aging time, drying temperature and calcination temperature was also investigated and the optimum conditions were 6 h in aging, 100°C for drying sample and 300-500°C for calcining the sample.

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