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#### paper text:

Article Volume 12, Issue 5, 2022, 5845 - 5859 <https://doi.org/10.33263/BRIAC125.58455859> Synthesis and Characterization of Methionine- Functionalized Boehmite with Enhanced Removal of Methyl Orange

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Received: 25.08.2021; Revised: 29.09.2021; Accepted: 2.10.2021; Published: 24.10.2021  
Abstract: Methionine, an amino acid with thioether, carboxyl, and amino functional groups, was used to enhance the adsorption capacity of boehmite toward methyl orange (MO). An environmentally friendly synthesis method was performed to prepare the methionine-functionalized boehmite (MFB) at 70°C using water as the solvent. The MFB has prominent XRD characteristic peaks at  $2\theta = 14.5^\circ, 28.6^\circ, 38.4^\circ$ , and  $48.4^\circ$ . The addition of functional groups from methionine was indicated by the appearance of FTIR bands at 2094, 1424, and 1220  $\text{cm}^{-1}$  corresponding to carboxyl, amino, and thioether groups, respectively. The N2 isotherm curve indicates the mesoporous structure of MFB, with surface area, pore-volume, and mean pore width of 287  $\text{m}^2 \text{g}^{-1}$ , 0.996  $\text{cm}^3 \text{g}^{-1}$ , 13.85 nm, respectively. The kinetic adsorption data showed a good fitting with the pseudo-first-order model, where the equilibrium can be achieved within 50 min. The adsorption of MO by MFB was better correlated

5with the Langmuir model with a maximum adsorption capacity of 167.4075 mg g<sup>-1</sup>, which was achieved at 323 K. The

thermodynamic study reveals

19that the adsorption of MO on MFB was an endothermic and spontaneous

process. Keywords: aluminum; boehmite, methionine; methyl orange; adsorption. © 2021 by the authors.

9This article is an open-access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>). 1. Introduction The constant population growth and

rapid industrial/societal development consequently escalate the use of synthetic dyes in textile, medicinal, food, plastics, and cosmetics manufacturing [1-3]. For these coloring purposes, over 700,000 tons of synthetic dyes were created per year. Up to 15% of these dyes were reportedly leached out due to unsystematic disposal where it ended in the water bodies [4-10]. The dye-contaminated water is postulated to trigger various toxicological and carcinogenic effects on humans and the ecosystems [11- 14]. Therefore, appropriate treatment is essential to accommodate the demand for clean water globally [15]. Dye removal through physical processes (such as adsorption) is preferable to the chemical approach, which often involves harmful chemicals. Adsorption is a reliable, simple physical technique for dye removal attributed to its low cost, low energy requirement, high feasibility, high removal efficiency, high reusability, and minimal by-products [16]. The selection of adsorbent with a high adsorption capability emerges as one of the most crucial factors in guaranteeing the successful removal of hazardous pollutants [6, 17, 18]. In the past decade, metal oxides were postulated to possess a good adsorption affinity towards many ionic pollutants and had been widely utilized to remove various hazardous substances from the water. Their frequent application as adsorbent was contributed by the low- cost, straightforward synthesis and modifiable properties [19,20]. Boehmite, a dimorphous material containing diasporic  $\gamma$ -AlOOH and  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, is a class of metal oxide with hydroxide moiety. Boehmite is known to have a sorption ability toward organic and inorganic compounds, besides its potential in other applications such as catalysts [21,22], ceramics [23,24], abrasives [25], and filters [26]. There have been some reports that demonstrated the enhanced adsorption capacity of boehmite by the functionalization technique. The addition of tetraethylenepentamine (TEPA) during the preparation of amino-functionalized boehmite gave a nearly two-fold increase in the adsorption capacity toward Cr [27]. In another study, the boehmite modification through the addition of chitosan reportedly shows up to 3-fold enhanced Cr adsorption compared to the unmodified boehmite [28]. Herein, functionalization of boehmite was conducted by introducing methionine, a compound containing multiple functional groups. Methionine with amino (—NH<sub>2</sub>), carboxylic (—COOH), and thioether (R—S—R) functional groups [29,30] can provide additional binding sites to increase the number of active adsorption sites of boehmite. Synthesis of boehmite was conventionally conducted using a high temperature (100 to 240 °C) hydrothermal process and a long reaction time (5 to 24 h) [31-33]. Metal salt such as AlCl<sub>3</sub>, Al(NO<sub>3</sub>)<sub>3</sub>, Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> was commonly used as the metal precursor in the synthesis, using acid or base solvent such as acetic acid, NH<sub>4</sub>OH, CO(NH<sub>2</sub>)<sub>2</sub>, NaOH, ethylenediamine, and triethanolamine [31-34]. In a typical-conventional procedure, boehmite particles are produced from a two-steps process: (1) dropwise addition of acid/base solution to aluminum salt solution, and (2) hydrothermal reaction at a specific high temperature with a long reaction time [31,32,34]. In this work, a green synthesis procedure of boehmite was demonstrated to substitute the conventional method. The boehmite particles were synthesized in the aqueous phase at a low temperature of 70 °C. This work

aimed to characterize and investigate the adsorption properties of methionine-functionalized boehmite (MFB), which was synthesized using the one-step-green preparation technique, i.e., low temperature (70 °C) aqueous phase synthesis. The adsorption capacity enhancement due to additional functional groups from methionine was investigated toward methyl orange (MO) as the model dye.

## 22. Materials and Methods 2.1. Materials. Aluminum sulfate

-18-hydrate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ , MW: 666.410 g/mol) was obtained from JT Baker, Mexico. Sodium hydroxide (NaOH, MW: 40.00 g/mol) was provided by Fisher Scientific, England. DL-Methionine ( $\text{C}_5\text{H}_{11}\text{NO}_2\text{S}$ , MW: 149.21 g/mol, 99.5%) was purchased from Sigma-Aldrich, USA. MO (C.I. 13025,  $\text{C}_{14}\text{H}_{14}\text{N}_3\text{NaO}_3\text{S}$ , MW: 327.334 g/mol) was supplied by Merck, Germany.

20All chemicals used in this experiment were analytical grade and directly used without any further treatment. All solutions were prepared in deionized (DI) water

, which was filtered through a NANO Pure

7Ultrapure water filtering system with a resistance of 18.1 MΩ cm-1

. 2.2. Synthesis of MFB. Prior to the MFB synthesis, two solutions were prepared: (i) deprotonated methionine solution was prepared by adding 0.0237 mmol (3.53 g) methionine powder into 25 mL DI water containing 0.0713 mmol (2.85 g) NaOH; (ii) 0.0079 mmol (5.26 g)  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$  in 25 mL DI water. These two solutions were mixed and allowed to react at 70°C for 1 h under constant stirring. The resultant white solids (MFB) were collected by centrifugation (Hsiangtai CN-350) at 6000 rpm for 15 min and repeatedly washed with DI water until neutral pH. Those collected MFB were then dried and activated (to evaporate the  $\text{H}_2\text{O}$  bound within the material)

19in a vacuum oven at 100°C for 10 h. The unmodified boehmite was

prepared similarly but

6without the addition of methionine and later was used as a control. 2.3. Characterization

. The MFB crystal pattern was recorded using a Bruker D2 Phaser-Xray Powder Diffraction equipped with a LynxEye detector and a Cu-K $\alpha$  radiation source with  $\lambda=1.5406\text{ \AA}$ . The diffraction measurement was carried out at an angle range of 5° to 60°, with a scan step size of 0.051°. The scan time at each increment was 0.5 s. The XRD spectra were analyzed by the EVA software. Fourier transform infrared (FTIR) spectroscopy using an FTS-3500 Bio-Rad FTIR was performed to characterize the functional groups of the synthesized MFB. KBr

5was used as the background, and the measurements were conducted at a wavenumber range of 4000-400 cm-1

. The nitrogen adsorption-desorption experiments were carried out using a Quantachrome instrument (ASiQwin) to determine the specific surface area, pore diameter, and pore volume of MFB. Prior to the nitrogen sorption experiment, the sample was degassed, the outgassing process was operated at 100°C for 2 h. The

**1** surface topography was observed using a JSM-6500F field emission Scanning Electron Microscope

(FE-SEM). Before the imaging, the

**2** samples were sputter-coated with an ultra-thin layer of conductive platinum. The

backscattered electron detector

**14** at an accelerating voltage of 10 and 15 kV

and a working distance (WD) of 9.4 and 11.1 mm were used for the imaging.

**7** Thermogravimetric analysis (TGA) was performed on a TA Instruments TGA 550 under N<sub>2</sub> flow at a constant heating rate of 10°C

/min. The elemental analysis (EA) and inductively coupled plasma - optical emission spectrometry (ICP-OES) to determine the C, H, O, N, S, Na, and Al content was performed on Elementar Vario EL Cube and Thermo iCAP 7000.

**18** The point of zero charges (pHPZC) was determined using

a PALS

**14** Zeta Potential Analyzer (Brookhaven Instruments). The Smoluchowski model was used to obtain the

zeta potential values. A series of scintillation vials containing 50 mL DI water was prepared for this analysis at different pH values (2 – 12) by adding 0.05 N NaOH or 0.05 N HCl solution. Subsequently, 10

**1** mg of adsorbent was added to each scintillation vial and

shaken with a sonicator for 1 h, and the zeta potential of each solution was measured. The pHPZC was determined by plotting zeta potential values versus pH values.

## 72.4. Adsorption experiment. 2.4.1. Effect of pH

on MO adsorption. Before the detailed adsorption kinetic and adsorption study, the effect of pH on the adsorption efficiency of MFB for MO removal was assessed. The analysis was done according to the previous reports.[35] The initial and equilibrium concentration of MO was determined by a UV-visible spectrophotometer (UV-2600 UV-VIS Shimadzu) at 460 nm. 2.4.2. MO adsorption kinetics. A batch adsorption kinetics experiment was conducted by adding a fixed amount of adsorbent (5 mg) into a series of scintillation vials containing 5 mL of MO solutions at various initial concentrations (9 to 60

3mg L<sup>-1</sup>). The adsorption was carried out in an orbital shaker incubator

(DENG YNG Incubator E600L) at 200 rpm and 298 K., The amount of MO adsorbed was measured at

4a specific time interval. The remaining MO concentration

was then determined using a spectrophotometer

11at a maximum wavelength of 460 nm. The

4amount of MO adsorbed at a specific time was determined according to Eq. (1).  
 $q_t$

$= (C_0 - C_t) \times V / m$  (1) where  $C_m$

4is the initial concentration of MO and  $C_t$  (mg L<sup>-1</sup>) is the concentration of MO  
at a specific time  $t$ .  $q_t$  (mg g<sup>-1</sup>) is the amount of

MO adsorbed per g of adsorbent at a specific time.  $V$  (L) and  $m$  (g) are the volume of the adsorption system and the mass of the adsorbent, respectively. 2.4.3. MO

4adsorption isotherm study. The adsorption isotherm study was conducted at  
various initial MO concentrations

(

1750 to 2000 mg L<sup>-1</sup>). The pH of the MO solution was

adjusted to pH 3 since the best MO adsorption occurred at this pH (see the discussion section 3.3.1). The adsorbent (5

16mg) was then added to the MO-containing solution. The

adsorption isotherm

1 was conducted at three different temperatures (30, 40, 50 °C) for 6 h. The amount of

MO adsorbed by MFB at the equilibrium condition was calculated using Eq. (2).  $q_e = (C_0 - C_e) \times V / m$  (2)

2 where  $C_e$  (mg L<sup>-1</sup>) is the concentration of MO at equilibrium and  $q_e$  (mg g<sup>-1</sup>) is the amount of MO adsorbed per g of

adsorbent at equilibrium state. 3. Results and Discussion 3.1. The formation mechanism of MFB. The formation of MFB was initialized by the deprotonation of methionine (Figure 1a). The deprotonation of methionine was triggered by OH<sup>-</sup> ions from NaOH, which created an alkaline condition. The addition of NaOH in this step caused the increase of solution pH to 10. The deprotonation of methionine occurred at the —COOH and —NH<sub>3</sub><sup>+</sup> groups, with pKa1 and pKa2 values of 2.16-2.28 and 9.12-9.21, respectively. The deprotonation of methionine facilitated its dissolution and allowed the formation of negatively charged methionine species [36]. Subsequently, the trivalent Al ions were added to the solution containing the deprotonated methionine. The charge difference between Al and methionine allowed the electrostatic interaction, leading to the formation of coordinated Al/methionine complexes (Figure 1b). The excess OH<sup>-</sup> ions attacked the Al metal and induced hydroxide species formation, that is, the boehmite. The methionine, initially coordinated with Al, stayed bonded with the boehmite particle, thus creating the MFB particles. (a) pKa1 protonated methionine pKa2 1st deprotonation 2nd deprotonation (b) Alkaline pH Methionine-functionalized Boehmite (MFB) R = methionine Al/methionine complex Figure 1. Schematic formation of MFB. (a) Stepwise deprotonation of methionine. (b) The formation of Al/methionine complexes, followed by the formation of methionine-functionalized boehmite (MFB) at alkaline conditions. 3.2. Characterization and compositional analysis of MFB. 3.2.1. Characterization of MFB. The XRD crystallinity pattern of MFB particles is presented in Figure 2a. The un-sharped and broad diffraction peaks of MFB indicate the amorphous nature of these particles [16].

4 A Rietveld refinement was conducted on the XRD pattern of

MFB, and it showed good convergence with the XRD pattern of boehmite (JCPDS 21-1307). The goodness of fit was also indicated by the low Bragg R-factor (12.1) and Chi-square value (5.2) of the Rietveld refinement. The XRD pattern of the synthesized-MFB shows typical characteristic peaks at 2 $\theta$ : 14.5°, 28.6°, 38.4°, and 48.4°, corresponding to the (020), (021), (130), and (150) crystal plane, respectively [37]. This result demonstrates that  $\gamma$ -AlO(OH) is the major crystal component in the MFB compound. The FTIR spectra bands of MFB are shown in Figure 2b. The asymmetric and symmetric stretching vibrations of the —OH group were indicated by the

15 appearance of bands at 3390 and 3102 cm<sup>-1</sup>, respectively. Meanwhile, the

symmetric bending vibration of —OH was indicated by the band at 1068 cm<sup>-1</sup>. The presence of coordinated Al-O was indicated by bands at 747, 646, and 486 cm<sup>-1</sup> [38]. Evidence of methionine binding in MFB was



indicated by the appearance of methionine characteristic peaks [39], such as CH<sub>3</sub> stretching vibration at 2922 and 2835 cm<sup>-1</sup>, C=O stretching at 2094 and 1687 cm<sup>-1</sup>, C-N stretching at 1424 cm<sup>-1</sup>, and CH<sub>2</sub>-S wagging vibration at 1220 cm<sup>-1</sup>. (a) MFB Intensity AIO(OH) Ref 10 20 30 40 50 60 2? (o) (c) 700 600 Adsorption (cc/g STP) 500 Desorption 400 300 200 100 0 0.0 0.2 0.4 0.6 0.8 1.0 (b) 100 1424 % Transmittance 90 ?C-N 80 2094, 1687 ?C=O 70 2922, 2835 1220 ?CH<sub>3</sub> ?CH<sub>2</sub>-S 60 3102 1068 ?sOH ?sOH 20 3390 ? asOH 747, 646, 486 0 ?Al-O 4000

113000 2000 1000 Wavenumber (cm<sup>-1</sup>) (d) **Figure 2.** Characterization of MFB. (a

) XRD pattern of MFB. The XRD reference pattern of  $\gamma$ -AlO(OH) was adopted from JCPDS 21-1307 (gray plot). (b) FTIR chromatogram of MFB with characteristic peaks of  $\gamma$ -AlO(OH) and methionine are labeled in red and blue color, respectively. (c) N<sub>2</sub> adsorption-desorption isotherm of MFB. (d) SEM image of MFB. The N<sub>2</sub> sorption isotherm curves of MFB are shown in Figure 2c.

1A typical type-IV mannerism with a broad hysteresis loop

at a relative pressure (P/P<sub>0</sub>) between 0.4 to 1.0, indicates the mesoporous structure of MFB. The steep gradient in the hysteresis loop denotes capillary condensation, which causes a delayed desorption process [40]. Calculation using the BJH equation further confirms the mesoporous structure with an average pore diameter of 13.85 nm. The calculated

22BET surface area and total pore volume of MFB are 287 m<sup>2</sup> g<sup>-1</sup> and 0.996 cm<sup>3</sup> g<sup>-1</sup>

, respectively. The SEM image of MFB (Figure 2d) shows the morphology of the particles. No specific geometry can be observed from the particles. The particles show an irregular surface characteristic that resembles a crumpled sheet. 3.2.2. Compositional analysis of MFB. The composition and quantity of the content in the MFB were determined by TGA and EA. The result of the TGA analysis is shown in Figure 3. The decomposition of the MFB material is divided into three stages. The first stage is a sharp decrease of around 9% of mass loss from room temperature until ~100°C. At this stage, the H<sub>2</sub>O in the material has evaporated and marked the desorption of physic-sorb. Subsequently, the second stage of the decline is wider from 100 to 500°C with 20% mass loss, and this condition is the result of the transformation MFB change into  $\gamma$ -alumina. From this stage, it was explained that the amount of methionine contained in the MFB was about 20% by mass. The last stage portrayed no significant decrease of mass from 500 to 900°C with 3.3% mass loss, and this result connected to the elimination of residual hydroxyls (-OH). After 900°C, the remaining component is Al<sub>2</sub>O<sub>3</sub> [41]. The EA data (C 0.36, H 3.75, O 30.32, Al 57.14, Na 4.76, and trace amount of N, S) are consistent with the TGA result, where the major component in the MFB material is Al and O. 100 Weight (%) 90 80 70 60 0 0 100 200 300 400 500 600 700 800 900 Temperature (°C) Figure 3. TGA analysis of MFB. 3.3. Adsorption of MO by MFB. 3.3.1. Compares Adsorption of MO by Boehmite and MFB. The success in boehmite modification by methionine also can be observed by the enhanced adsorption capacity of MFB compared to the unmodified boehmite. As shown in Table 1, the MFB showed a 1.33-fold higher adsorption capacity than the unmodified one. This suggests that the amine group of methionine may facilitate a selective binding affinity towards anionic dyes such as MO. Table 1. Adsorption of MO at 25°C. Adsorbent *qe* (mg g<sup>-1</sup>) Boehmite \* 32.68 ± 0.59 MFB 43.48 ± 0.42 \* without methionine functionalization 3.

53.2. Effect of pH. As shown in Figure 4a, maximum adsorption of

MO by MFB occurred at pH 3. Adsorption of MO decreased sharply as the pH was increased to >4.

**12**This phenomenon can be explained by the surface charge of the MFB particles and the charge of MO molecules. At acidic pH, the excess H<sup>+</sup> ions

attack the MFB particles (Al-OH and Al-NH<sub>2</sub>) and cause positive surface charges (Al-OH<sub>2</sub><sup>+</sup> and Al-NH<sub>3</sub><sup>+</sup>). The sulphonate group (SO<sub>3</sub><sup>-</sup>) of the MO are willingly dissociated to form the anion. The positively charged MFB particles at acidic pH were confirmed from the positive zeta potential value, as shown in Figure 4b. MO, which is an anionic dye, is attracted to the MFB particles due to the difference in charge. Therefore, the electrostatic interaction occurs, leading to the attachment of MO on MFB [42,43]. As the pH increases, the zeta potential of MFB decreases toward the negative value. Thus, the electrostatic attraction between the two compounds is reduced as the surface charges become more negative. (a) 10 (b) 30 (mg/g) 8 6 4 0 10 0 2 Zeta potential (mV) 20 -10 -20 -30 1 2 3 4 5 6 7 8 9 10 11 pH 2 4 6 pH 8 10 12 14 Figure 4. Effect of pH on MFB adsorption efficiency and surface property. (a) Adsorption capacity ( $q_e$ , mg/g) of MFB for MO removal and (b) Zeta potential of MFB from pH 3 to 10. 3.3.3. Adsorption kinetics. Adsorption kinetics plays an important role in providing information about the

**21**time required to reach equilibrium [44]. As

shown in Figure 5, rapid uptake of MO occurred in the first 15 min. A deceleration point before the plateau occurred at  $t = 30$  min. The kinetics data were fitted with the pseudo-first-order by Lagergren [45] and pseudo-second-order [46] models to determine the adsorption rates. The corresponding mathematical expression of the two models is presented as Eq. (3) and (4), respectively.  $qt = qe(1 - e^{-k_1t})$  (3)  $qt = 1 + \frac{k_2 qe^2 t}{1 + k_2 qe t}$  (4) where

**10** $k_1(\text{min}^{-1})$  and  $k_2(\text{g mg}^{-1} \text{ min}^{-1})$  are the first- and second-order rate constants, respectively

.  $C_0 = 9.5 \text{ mg/L}$   $C_0 = 21 \text{ mg/L}$  (a)  $C_0 = 33 \text{ mg/L}$  (b) 50 50 40 40 (mg/g) 30 20 (mg/g) 30 20 10 10 0 0 50 100 150 200 Time (min) 0 0 50 100 150 200 Time (min) Figure 5. Adsorption kinetics of MO onto MFB. Solid lines represent (

**2a)** pseudo-first-order and (b) pseudo- second-order fitting curves. The

calculated kinetic parameters obtained from the data fitting are shown in Table 2. The values of  $q_e$  from the pseudo-first-order model are very close to the experimental data ( $q_{e, \text{exp}}$ ), indicating that this model can satisfactorily represent the kinetics data. The goodness of fit of the pseudo-first-order model is also indicated by their correlation coefficients value ( $Q^2$ ) which close to 1. The effect of increasing initial MO concentration ( $C_0$ ) on the parameters  $q_e$  and  $k$  was also observed. A high  $C_0$  induces higher MO uptake, as indicated by the higher  $q_e$ . This is since a higher number of adsorbate molecules can provide a higher driving force to overcome the mass transfer resistance. In contrast, the increase in  $C_0$  is followed by a decrease in adsorption rate ( $k$ ). The decline of  $k$  at higher  $C_0$  is attributed to the higher probability of collisions between molecules which cause an increase in mobility of the molecules and prevent the adsorbates from attaching to the adsorbent surface [35,47]. Table 2. Summarize of kinetic parameters of MO onto MFB material at 25°C The initial concentration of MO (mg/L) 9.5 21 33 44 56  $q_{e, \text{exp}}$  (mg/g) 7.5128 16.1943 26.7279

39.0902 45.8017 Pseudo-first order  $qe_1$  (mg g<sup>-1</sup>) 7.3690 14.5701 25.1250 37.1854 45.5491  $k_1$  (min<sup>-1</sup>) 0.1118 0.1120 0.1105 0.1668 0.0888  $Q_2$  0.9922 0.9715 0.9856 0.9944 0.9965 Pseudo-second order  $qe_2$  (mg g<sup>-1</sup>) 7.7335 15.3584 26.5935 38.9599 48.0305  $k_2$  (g mg<sup>-1</sup> min<sup>-1</sup>) 0.0322 0.0153 0.0084 0.0069 0.0038  $Q_2$  0.9981 0.9741 0.9966 0.9882 0.9835 3.3.4. Adsorption isotherms and thermodynamic. The Langmuir and Freundlich models were used to represent the adsorption equilibria of MO on MFB.

**16**The Langmuir isotherm model has the form as follows.  $qe = \frac{K_1 L + Q K_m L C_{ee}}{(5) \text{ where } Q_m$

(

**15**mg g<sup>-1</sup>) and  $K K(L \text{ mg}^{-1})$  are adsorption capacity and adsorption affinity, respectively

. Hall et al. (1966) and Tran et al. (2017) suggested that the separation factor or equilibrium parameter ( $QK$ ) can be used to express the essential characteristics of the Langmuir isotherm. The  $QK$  value which describes the adsorption favorability, wherein  $QK < 1$  shows favorable adsorption and  $QK > 1$  shows unfavorable adsorption [44,48].  $QK$  is a dimensionless constant and is defined as follows:  $QK = 1 + K L C_o$  1 (6) The Freundlich isotherm has a mathematical form shown in Eq. (7).  $qe = K F C_e^{1/m}$  (7) where  $K F$  ((mg/g)/(mg/L)<sup>n</sup>) and  $m$  (dimensionless) are Freundlich adsorption capacity and the intensity parameter, respectively. The adsorption isotherm data and the fitting are depicted in Figure 6.

**7**According to the classification by Giles et al. (1960), the

**5**adsorption isotherm curve can be classified as an L- curve with subclass

1 [49]. The L-type curve indicates that the adsorption occurs mainly due to the weak forces, i.e., van der Waals forces. Subclass 1 suggests that the adsorption sites were not fully occupied. Due to MFB have large capacity: surface area and total pore volume (287 m<sup>2</sup> g<sup>-1</sup> and 0.996 cm<sup>3</sup> g<sup>-1</sup>, respectively). 30 oC (a) 150 (mg/g) 120 90 60 40 40?0CoC 303?0CoC 505?0CoC 40 oC 50 oC 50 oC Model (b)M1o5d0el Col 12 v s Col 13 Model

**13**Col 12 vs Col 13 Col 14 vs Col

15 Col12104 v s Col 15

**13**Col 12 vs Col 13 Col 14 vs Col

15 90 (mg/g) 60 Model

**13**Col 12 vs Col 13 Col 14 vs Col

15 30 40 50 30 30 0 0 500 1000 1500 2000 0 500 1000 1500 2000 (

1mg/L) (mg/L) Figure 6. Adsorption isotherm of MO onto

MFB at different temperatures. Solid lines represent (a) Langmuir and (b) Freundlich fitting curves. All constants obtained from the two isotherm models are summarized in Table 3. Based on the correlation coefficient ( $R^2$ ), the Langmuir model is more suitable than the Freundlich model. In this study, the temperature positively influenced the amount of MO adsorbed by MFB. Increasing

6temperature from 303 to 323 K resulted in the increase of  $Q_m$  from 104.9140 to

167.4075 mg g<sup>-1</sup>. Based on the separation factor or equilibrium parameter ( $R_L$ ), this adsorption is favorable because of  $R_L < 1$  [44]. Table 3. Summary of the temperature effects on adsorption isotherm parameters of MO onto MFB material Temperature Langmuir Freundlich (

12K)  $Q_m$  (mg/g)  $K$  (L/mg)  $Q$   $QK$   $K$

(mg/g)(mg/L)<sup>1/n</sup>  $F$   $1/m$   $Q_1$  303 104.9140 0.0019 0.9613 0.21-0.84 2.0902 0.4932 0.9346 313 125.5011 0.0036 0.9595 0.12-0.73 5.7638 0.3997 0.9492 323 167.4075 0.0022 0.9827 0.19-0.90 4.0792 0.4763 0.9361 The thermodynamic adsorption parameters were determined based on Eq. (8) and (9).  $\Delta G = -QK \ln(KC)$  (8) where  $\Delta G$  (kJ mol<sup>-1</sup>),  $Q$  (8.3144 J mol<sup>-1</sup> K<sup>-1</sup>),  $Q$  (K), and  $KC$  (dimensionless) are the change in

14Gibbs free energy, the ideal gas constant, the temperature

of the process, and the equilibrium constant, respectively. The  $KC$  value can

1be obtained by multiplying  $K$  (L mg<sup>-1</sup>) value with the molecular weight of the adsorbate (g mol<sup>-1</sup>) and by

1855.5, which corresponds to the number of moles of pure water per liter

[50]. The van't Hoff equation, Eq. (9), was then used to determine the change in enthalpy ( $\Delta H$ ,

17kJ mol<sup>-1</sup>) and entropy ( $\Delta S$ , J mol<sup>-1</sup> K<sup>-1</sup>

).  $\Delta G = \Delta H - Q\Delta S$  (9) Table 4. Thermodynamic properties of adsorption of MO onto MFB. Temperature  $\Delta H$   $\Delta S$   $\Delta G$  (K) (

2kJ mol<sup>-1</sup>) (kJ mol<sup>-1</sup>) (J mol<sup>-1</sup> K<sup>-1</sup>

) 303 -26.3243 313 -28.8562 6.4528 109.6836 323 -28.4556 The obtained thermodynamic parameters are summarized in Table 4. The negative  $\Delta G$  values of -26.3243, -28.8562, and -28.4556 kJ mol<sup>-1</sup> were obtained for adsorption at 303, 313, and 323 K, respectively. The increase in temperature followed by a decrease in  $\Delta G$  value indicates the spontaneity of the adsorption. The calculated  $\Delta H$  value is +6.4528 kJ mol<sup>-1</sup>, which shows the occurrence of an endothermic process. Meanwhile, the positive value of  $\Delta S$  (109.6836 J mol<sup>-1</sup> K<sup>-1</sup>) indicates the increase of randomness during MO uptake onto MFB [51]. 3.3.5. Adsorption mechanism. FTIR and XRD measurements were employed on the spent-MFB (MO@MFB)

10to gain insight into the adsorption mechanism of MO onto the adsorbent. As shown in Figure

7a, there is a change in the bands at about 3500 cm<sup>-1</sup>, corresponding to the —OH group vibration. The band alteration can be

2attributed to the interaction between the MO molecules and the

—OH group on the surface of MFB, which occurred through the van der Waals interactions. Another band change was observed at ~2900 cm<sup>-1</sup>, corresponding to the CH<sub>3</sub> alkyl group vibration [52]. These results suggest that there was an interaction between the MO molecules with the alkyl group. However, there is no significant change in the XRD spectra of MO@MFB from that of MFB, which implies that there is no molecular structure change due to the attachment of MO (Figure 7b). Overall, the FTIR and XRD data indicate that the adsorption of MO by MFB does not occur via anion-cation exchange [16] but through a physical process. (a) (b) %T ransmittance Intensity 4000 3000 2000 1000 10 20 30 40 50 60 Wavenumber (cm<sup>-1</sup>) 2? (o) Figure 7. Characterization of MFB after adsorption of MO (MO@MFB). (a) FTIR spectra and (b) XRD pattern of MFB (black color) and MO@MFB (orange color). Table 5 compares the maximum adsorption capacity ( $Q_m$ ) of MFB (in this work) and several functionalized-adsorbent for MO removal. The  $Q_m$  of MFB was ~20.9-folds higher than that of the unmodified boehmite ( $\gamma$ -AlO(OH)). MFB also shows much higher MO adsorption (~10.8 times) than the amino-crosslinked hypromellose. On the other hand, the  $Q_m$  of MFB was about 11% lower than the amino-MIL-101(Al). Furthermore, the use of MFB offers several advantages in terms of its synthesis procedure. MFB can be synthesized at a lower temperature within a shorter duration than the other adsorbents listed in Table 5, thus providing an economic incentive for MFB. Table 5. MO adsorption Material capacity by amine-composite materials. Reference Synthesis condition  $Q_m$  (mg/g) Amino-MIL-101(Al) 130oC, 6 h 188.00 [53] Amino-crosslinked hypromellose 80oC, 3 h 15.56 [54]  $\gamma$ -AlO(OH) 150oC, 24 h 8.00 [34] MFB 70oC, 1 h 167.41 This work 4. Conclusions A green and energy-efficient technique was demonstrated to synthesize methionine- functionalized boehmite in an aqueous phase at a low temperature of 70°C. Methionine act as the Al complexation ligand that facilitates the formation of the Al-oxide-hydroxide (boehmite). The presence of methionine provides additional surface binding sites of boehmite, which enhances its adsorption capacity. The adsorption study of the methionine-functionalized boehmite against MO showed that the adsorption between the two compounds occurred via

5van der Waals forces. The adsorption

proceeded more spontaneously at a higher temperature, indicating the endothermic nature of the adsorption process. Funding Financial support by the National Science Council of Taiwan (MOST 108-

32221-E-011-106) is appreciated. A.E. Angkawijaya and A.W. Go would like to thank the National Taiwan University of Science and Technology for the

. Acknowledgments This research has no acknowledgment. Conflicts of

**6Interest The authors declare that the research was conducted without any  
commercial or financial relationships that could be construed as a potential  
conflict of interest**

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