



Research paper

# Formulation study for softening of hard water using surfactant modified bentonite adsorbent coating

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## ABSTRACT

As an adsorbent coating of surfactants, sodium dodecylbenzene sulfonate (SDBS) modified bentonite (SMB) has been developed for the removal of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions from hard water. The adsorbent was characterized using SEM-EDX, Zeta-meter and FTIR analyses. It has the potential to replace expensive conventional softening treatment techniques as well as reduce the usage of excess chemicals (chemical precipitation). Hard water can be easily treated using SMB by applying the adsorbent coating to the treatment area. Testing of adsorbent was carried out in terms of effect of surfactant ratio, effect of different types of binder as well as binder ratio. The best formulation of SMB was achieved by using polyvinyl acetate (PVAc) and bentonite in the ratio of 0.75:1.0 (w/w). Langmuir, Freundlich and Temkin isotherm models were tested to describe the optimum adsorption of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions on SMB adsorbent coating. Thermodynamic and kinetic parameters were also examined for the adsorption of metal ions at different temperatures. SMB demonstrated the highest metal ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) removal efficiency (29.27  $\text{mg g}^{-1}$ ) in 90 min from 120  $\text{mg L}^{-1}$  hardness. On the basis of good removal capacity for  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions, SMB can be effectively used for treatment of metal ions in industrial wastewater, as well as softening of hard water.

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## 1. Introduction

Water is one of the most important sources of living beings and is used in daily life (Archimede, 2016). Excess minerals content in water can create lots of problems to human health (Nabi et al., 2011a, 2011b; Shahadat et al., 2015). Calcium and magnesium are two major hardening cationic elements that cause the formation of hard water. Water can be considered hard when the concentration of calcium and magnesium is found to be above a permissible limit (120  $\text{mg L}^{-1}$ ) (Joseph Cotruvo, 2011). According to Malaysia Water Quality Standards, the maximum acceptable value for hard water is 500  $\text{mg L}^{-1}$  (Ministry of Health, 2010). It contains minerals ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) which can be easily deposited on the surfaces of equipment such as kettles, coffee makers, and heaters in the form of scale. The formation of scale causes clogging of a piping system thus lowering the water flow (Reddiroot'r, 2016). While causing pipe clogging, however, it tends to protect their surfaces by coating to prevent corrosion (Mcfarland et al., 2015). Besides this, it also decreases the foamy nature of soap and detergent soapy, leading to the use of additional soap to wash utensils and clothes,

as well as in bathing (Heidekamp and Lemley, 2005; Cameron, 2011). Moreover, hard water also affects the health and environment. Potential health problems caused by hard water are kidney stones, dermatitis, reproductive health issues and pancreatic cancer (Sengupta, 2014; Chiu et al., 2010). Atopic dermatitis and chronic inflammatory diseases have been found due to the intake of hard water. The United States, Canada, and the Netherlands, as well as the state of Perlis in Malaysia, all face the problem of hard water (Issa et al., 1998). The degree of water hardness at Titi Tinggi (Perlis) was found to be 530  $\text{mg L}^{-1}$ . In order to overcome the problem of water hardening, a number of commercial available methods for hard water treatment methods such as chemical precipitation, ion exchange (Bushra et al., 2012), adsorption (Moreira et al., 2015), electrolysis (Agostinho et al., 2012), electrochemical (Gabielli et al., 2006) and nanofiltration (Izadpanah and Javidnia, 2012) have been applied. Ion exchange treatment is considered one of the most commonly used methods. Salt of sodium chloride (NaCl), Amberlite, titanate nanotube (TiONTs), and zeolite are potential ion exchangers that have been used to exchange  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions (Cetin, 2014; Madarasz et al., 2014; Skipton, 2008; Xue et al., 2014). Although ion exchange is very effective in use, it is an expensive treatment technique. Adsorption is one of the promising methods that can be effectively used for softening hard water. A few studies have shown that natural sand may be used as an adsorbent due to its

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potential ability to soften hard water (Jayalakshmi et al., 2014; Sivasankar and Ramachandramoorthy, 2011). Activated carbon from agricultural waste, mainly coconut and cashew shell, is also used as a low cost adsorbent for the treatment of metal ions (Rolence et al., 2014). Development of the new inexpensive adsorbent has become a primary motivation for researchers. In this regard, bentonite has been discovered to be a potential adsorbent for softening of water because of its ability to allow adsorption of cationic species such as  $Pb^{2+}$  and  $Cu^{2+}$  (Melichová and Hromada, 2013) and acid dye (Alshammari, 2014). Native form of bentonite does not achieve high removal efficiency; therefore, to improve its adsorption capacity, bentonite is typically modified by the addition of other chemicals. Sodium dodecyl benzene sulfonate (SDBS), a surfactant with a negative charge, has potential to attract positive charge ionic component. It has been widely used in detergent due to the presence of alkylbenzene sulfonate which is a major constituent of synthetic detergent (Taffarel and Rubio, 2010). Although the addition of SDBS improves hard water softening properties, the presence of surfactant in water will cause the large-scale formation of foam which is harmful to the aquatic environment (Li et al., 2011). Thus, considerable amount of surfactant should be used to reduce the pollution effect. The present article deals with the fabrication of a new formulation of adsorbent coating using bentonite clay modified with SDBS in softening hard water.

## 2. Experimental

### 2.1. Materials and methods

The materials for the development of adsorbents were bentonite, kaolin, sodium dodecyl benzyl sulfonate (SDBS), and polyvinyl (PVAL) purchased from Sigma Aldrich, Malaysia. Pumice stone and polyvinyl acetate (PVAc) were bought from Watson Health Store and Mydin Store Penang (Malaysia), respectively. Water based paint was purchased from Sisson Company, Penang, Malaysia. For preparation of synthetic water,  $CaCl_2 \cdot 2H_2O$  and  $MgSO_4 \cdot 7H_2O$  salts were supplied from R&M Chemicals, Malaysia. All experiments were conducted according to the Standard Methods for the Examination of Water and Wastewater (Federation, 1999). The samples of synthetic hard water ( $120 \text{ mg L}^{-1}$ ) were prepared by dissolving 2.6 g hydrated calcium chloride ( $CaCl_2 \cdot 2H_2O$ ) and 4.1 g hydrated magnesium sulphate ( $MgSO_4 \cdot 7H_2O$ ) in distilled water (Fahmi et al., 2015).

### 2.2. Development of SMB adsorbent coating

To determine the optimal adsorption capacity of adsorbents, bentonite, pumice stone, and kaolin were chosen for testing. All three adsorbents were used to treat a synthetic hard water of  $120 \text{ mg L}^{-1}$  using a fixed amount of each adsorbent (0.3 g) individually at the speed of 300 rpm at room temperature ( $25 \pm 2^\circ \text{C}$ ). The amount of adsorbed metal ions on each adsorbent was determined by the titration of hard water using EDTA-titration at every interval of 30 min for 4 h. Thus, on the basis of higher adsorption capacity (among three adsorbents), bentonite was selected to fabricate surfactant modified bentonite adsorbent.

Surfactant modified bentonite (SMB) was prepared by mixing a fixed amount of bentonite (0.5 g) and SDBS (0.25 g) in distilled water (5 mL). The mixture was stirred for 10 min to complete dissolution of surfactant with bentonite. By adding 1.0 g binder (PVAc) to the mixture, a slurry formed was coated on the fabric material of  $126 \text{ cm}^2$  surface area and dried in an oven at  $70^\circ \text{C}$  for 24 h. The schematic presentation for the preparation of SMB adsorbent coating is shown in Fig. 1a. The proposed scheme for the removal of  $Ca^{2+}$  and  $Mg^{2+}$  ions from synthetic hard water is shown in Fig. 1b.

### 2.3. Instruments

The morphology of adsorbent was observed using a scanning electron microscope (SEM, Crest System (M) Sdn. Bhd, Quanta Feg 450) and its composition was determined via energy dispersive X-ray (EDX, Crest System (M) Sdn. Bhd, Quanta Feg 450). The surface charge of particles in the solution was determined using a zeta meter (Malvern idb Zeta Sizer Nano Series). Fourier Transform Infrared Spectroscopy (FTIR, NICOLET i10) was used to identify the existence of functional group in the sample of adsorbent coating. The X-ray diffraction (XRD) patterns of bentonite, SDBS and SMB were measured on a Bruker D/Max-2200 X-ray powder diffractometer with  $Cu/K\alpha$  ( $k = 0.154 \text{ nm}$ ) radiation (Bruker diffrac. suite eva version 1.4) AXS, Germany.

### 2.4. Effect of surfactant ratio

To improve the adsorption capacity of SMB, a fixed amount of bentonite (0.3 g) was mixed with various amounts of the surfactant SDBS (0.15 g, 0.3 g and 0.6 g). On the basis of higher adsorption efficiency, the selected SMB adsorbent was used for further study.

### 2.5. Effect on type of binder

To examine the maximum supporting ability of a binder with bentonite, polyvinyl alcohol, polyvinyl acetate, and also water based paint were selected. The selection of binder was on the basis of their safety use and without affecting the performance of adsorbent.

### 2.6. Effect on adsorbent to binder ratio

In order to determine the best adsorbent coating, an adsorbent to binder ratio experiment was conducted. Adsorbent to binder ratios with different dosages as studied are shown in Table 1.

### 2.7. Batch experiments

All experiments were conducted using a batch method in 250 mL beakers. A solution of hard water (200 mL) was stirred for 4 h at room temperature at 300 rpm with an initial concentration of  $120 \text{ mg L}^{-1}$ . Concentrations of  $Ca^{2+}$  and  $Mg^{2+}$  ions were determined at 15 min intervals for 4 h using EDTA-titration method (Federation, 1999). The metal ions ( $Ca^{2+}$  and  $Mg^{2+}$ ) removal efficiency (RE) was determined using Eq. (1) and adsorption capacity was calculated by Eq. (2). All experiments were repeated in triplicate.

$$RE = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

$$q_t \left( \frac{\text{mg}}{\text{g}} \right) = \frac{(C_0 - C_e) \times V}{m} \quad (2)$$

where  $C_0$  is the initial concentration ( $\text{mg L}^{-1}$ ),  $C_e$  is the final concentration ( $\text{mg L}^{-1}$ ),  $q_t$  is the adsorption capacity at time ( $\text{mg g}^{-1}$ ),  $V$  is the volume of the solution (L), and  $m$  is the mass of the adsorbent (g).

### 2.8. Adsorption isotherms

The adsorption behaviour of SMB for the removal of  $Ca^{2+}$  and  $Mg^{2+}$  was described in terms of isotherm models. Three adsorption isotherms; Langmuir, Freundlich and Temkin isotherms can be used to determine nature of the adsorption.

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