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## Mechanism of Turbidity and Hardness Removal in Hard Water Sources by using *Moringa oleifera*

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**Abstract:** The objective of this study was to describe the mechanism of simultaneous removal of hardness and background turbidity in hard water for drinking water purpose by using *Moringa oleifera* (MO) that extracted with 1.0 M sodium chloride solution (MO-NaCl). Raw water containing high concentration of hardness from Timah Tasoh Dam (Perlis, Malaysia) and synthetic water (turbid, hard, turbid and hard) were used in this study. The optimum turbidity removal efficiency of Timah Tasoh raw water, synthetic turbid water and synthetic turbid and hard water were approximately 80% which were attained at MO dosage of 15, 30 and 10 mg L<sup>-1</sup>, respectively. However, this optimum MO dosage was not applicable for hardness removal. The mechanism for turbidity removal by MO-NaCl was supposed to be adsorption and charge neutralization, with the adsorption isotherm following the Freundlich adsorption model. In addition, the mechanism for hardness removal in hard water seems to be adsorption that conforms to both Freundlich and Langmuir adsorption models. Therefore, for hard water such as Timah Tasoh raw water and synthetic turbid and hard water, the MO-NaCl tends to adsorb hardness to form a netlike structure followed by turbidity removal through sweep coagulation mechanism. Hence, hardness and turbidity can be removed simultaneously by using MO-NaCl.

**Key words:** Coagulation, adsorption, active agent, softening, synthetic water

### INTRODUCTION

It is well known that Aluminium Sulphate (Alum) and Polyaluminium Chloride (PAC) are coagulants that widely used in conventional water treatment processes for tap water production (Amirtharajah and Mills, 1982). The average percentage removal of turbidity by these coagulants ranged from 75.1 to 97.8% after filtration (Okeyo *et al.*, 2011). However, many studies reported that aluminum which is the major component of Alum and PAC, may induce adverse health effect such as Alzheimer's disease to the consumers (Crapper *et al.*, 1973; Martyn *et al.*, 1989; Suet-Mei and Wing-Shiu, 1992). In addition, the problem of reaction of alum with natural alkalinity present in the raw water leading to the reduction of pH and low efficiency in coagulation of cold water. Therefore, there has been considerable interest in the development of new natural coagulants that is more cost effective, less adverse health effect and environmentally friendly (Prasad, 2009).

MO is a tropical plant that has been shown to be one of the most promising natural coagulants for the removal

of turbidity in raw water for drinking water purposes, with potential usage on a large scale in tropical developing countries (Gassenschmidt *et al.*, 1995; Muyibi and Okufu, 1995; Ndabigengesere and Narasiah, 1998). Oluduro and Aderiye (2007) reported that MO seed can remove suspended particles in underground and surface water samples efficiently. The MO seed in diverse extracted and purified forms has also proved to be effective in reducing sludge volumes produced during coagulation process in comparison to alum (Ndabigengesere *et al.*, 1995), soften hard waters (Muyibi and Evison, 1996) and act as effective adsorber for heavy metals in water (Mataka *et al.*, 2006; Sharma *et al.*, 2006; Bhuptawat and Folkard, 2007). The mechanism of coagulation with MO that extracted with distilled water appears to consist of adsorption and neutralization of the colloidal charges. The active agents of the MO seeds have been determined to be cationic peptides that has molecular weight between 6-16 kDa (Jahn, 1986; Ndabigengesere *et al.*, 1995). Previous study reported that MO seed in dry form contained a significant amount of the active agent for coagulation (Anhwange *et al.*, 2004; Yameogo *et al.*, 2011).

Some studies reported that coagulation efficiency of MO can be improved by extraction of its active agents with sodium chloride solution (Okuda *et al.*, 1999; Prasad, 2009; Madrona *et al.*, 2010). This improvement was apparently due to the salting-in mechanism in proteins wherein a salt increases protein-protein dissociations, leading to increasing protein solubility as the salt ionic strength increases (Okuda *et al.*, 2001a). However, the active agents in this extraction method were predominantly caused by the lower molecular weight compound (e.g., 3 kDa). In addition, coagulation mechanism of purified MO-NaCl seems to be an entrapment by the insoluble matters formed by the coagulation active component (Okuda *et al.*, 2001b).

Several studies have also investigated the performance of MO for hardness removal in hard water for drinking water purpose. Sani (1990) and Fahmi *et al.* (2010) observed that in addition to turbidity reduction, the hardness was also reduced after coagulation with MO and settling. It was also reported that MO which is extracted with distilled water can be used for softening synthetic hard water, naturally hard surface water and groundwater from tube wells (Muyibi and Evison, 1995b). Higher dosages were required to achieve equivalent residual hardness for water samples with the same initial hardness but higher number of hardness-causing species in the water (Muyibi and Evison, 1995a). However, minimal studies have been directed towards simultaneous sorption behavior for the removal of hardness and turbidity by using MO-NaCl from water bodies. The objective of this study is to describe the mechanism of simultaneous removal of hardness and background turbidity in hard water for drinking water purpose by using MO-NaCl.

## MATERIALS AND METHODS

**Water sources:** Two sources of water (synthetic and surface waters) were used for the laboratory based experimental study. A surface water samples were collected in the lake of Timah Tasoh Dam (Perlis, Malaysia), during the month of January 2010. An in-situ parameters such as pH, temperature, conductivity and Oxidation Reduction Potential (ORP) were measured at site by using water quality multi-probe. This instrument was calibrated in the laboratory prior conducting the in-situ measurement. On the other hand, a 1 L of water samples were taken back to the laboratory for the determination of turbidity and hardness. Table 1 summarized the raw water quality characteristic for Timah Tasoh Dam. The hardness and turbidity in the raw water were ranged from 314-362 mg L<sup>-1</sup> as CaCO<sub>3</sub> and 12.1-113.4

Table 1: Water characteristic for Timah Tasoh Dam

Parameters	Unit	Range
pH	-	8.2-10.4
ORP	mV	361-372
Conductivity	mS cm <sup>-1</sup>	0.18-0.19
Temperature	°C	28.6-32.1
Turbidity	NTU	12.1-113.4
Calcium hardness	mg L <sup>-1</sup> as CaCO <sub>3</sub>	189-267
Magnesium hardness	mg L <sup>-1</sup> as CaCO <sub>3</sub>	86-119
Total hardness	mg L <sup>-1</sup> as CaCO <sub>3</sub>	314-362

ORP: Oxidation reduction potential

Nephelometric Turbidity Unit (NTU), respectively. High level of hardness in Timah Tasoh Dam was possibly due to the geological formation of the dam that consists of mainly alluvium, gravel and sand layers and limestone hard rock. The total hardness from this study was higher than 300 mg L<sup>-1</sup> as CaCO<sub>3</sub> which was classified as very hard water (Al-Salamah and Nassar, 2009).

**Preparation of synthetic water samples:** The stock solutions for synthetic calcium and magnesium hard waters were prepared by adding 2.6 g of hydrated calcium chloride (CaCl<sub>2</sub>.2H<sub>2</sub>O) and 4.1 g of hydrated magnesium sulphate (MgSO<sub>4</sub>.7H<sub>2</sub>O) into 1 L of distilled water, respectively. Both stock solutions containing calcium and magnesium hardness concentrations about 2000 mg L<sup>-1</sup>. The concentrated synthetic hard waters were diluted approximately to the hardness level of the Timah Tasoh raw water. The concentrations of synthetic calcium and magnesium hard water were prepared at 250 and 100 mg L<sup>-1</sup> as CaCO<sub>3</sub>, respectively.

Stock solution for synthetic turbid water was prepared by adding 10 g of Kaolin powder into 1 L of distilled water. The suspension was stirred for 1 h in order to ensure uniform mixing of Kaolin particles. The solution was left for 24 h to allow complete hydration of particles. The synthetic turbid stock solution was diluted immediately with distilled water to 21±1 NTU before the coagulation experiment which nearly the same as turbidity level of the natural raw water collected from Timah Tasoh Dam. The synthetic turbid and hard water was prepared by adjusting the synthetic hard water with Kaoline stock solution up to 21 NTU and total hardness up to 350 mg L<sup>-1</sup>.

**Preparation of MO coagulant:** The MO used in this study was obtained from Kangar, Perlis (Malaysia) and the extraction method was based on the modified procedure developed by Okuda *et al.* (1999). A qualified seeds were selected and dried in an oven at 40°C for 24 h. The kernel was separated from hulls and wings and grinded to fine powder by using domestic blender. The MO seed extracts was prepared by mixing 1 g of MO seed powder with 100 mL of 1.0 N sodium chloride solution to extract active component. The filtered solution used as a 10,000 mg L<sup>-1</sup> MO-NaCl stock solution in this study.

**Coagulation efficiency test:** A standard jar test unit equipped with 6 paddles was used in this study. The experiment were carried out in 4 different types of solutions namely synthetic turbid water (21±1 NTU), synthetic hard water (250 mg L<sup>-1</sup> Ca as CaCO<sub>3</sub> and 100 mg L<sup>-1</sup> Mg as CaCO<sub>3</sub>), synthetic hard and turbid water (21±1 NTU and 250 mg L<sup>-1</sup> Ca as CaCO<sub>3</sub> and 100 mg L<sup>-1</sup> Mg as CaCO<sub>3</sub>) and natural surface water. MO coagulant was prepared on the same day of experiment to avoid aging phenomena. The MO dosages used in this study were ranging from 3 to 50 mg L<sup>-1</sup>. The pH was set approximately to Timah Tasoh water sample at 9.3.

The jar test was performed to study the effect of coagulant dosage on the removal efficiency of turbidity and hardness as well as the competitive removal efficiency of turbidity and hardness. All five 1000 mL beakers were initially filled with water samples and were placed in the slots of jar tester and were subsequently agitated. During this process, 3, 5, 10, 15, 20, 30, 40 and 50 mg L<sup>-1</sup> of MO coagulant were added to each beaker and the agitation process were conducted in two stages; (1) 150 rpm for 2 min (2) 30 rpm for 30 min. After sedimentation for 1 h, 30 mL of water sample was collected from the middle section of the beaker and the residual turbidity of each coagulated water samples were measured using a turbidity meter. At the same time some suspended flocs were observed in the water samples and this may take a longer time for the flocs to settle. Hence, a filter paper was used to obtain the product water for the measurement of hardness water quality.

**Simultaneous removal of hardness and turbidity:** In order to clarify the mechanism of simultaneous removal of turbidity and hardness in natural and synthetic water, the Langmuir and Freundlich isotherms were employed to describe the adsorption equilibrium. Theoretically derived Langmuir model assumes adsorption energies are uniform and independent of surface coverage and complete coverage of the surface by a monolayer of adsorbate indicates maximum adsorption. On the other hand, Freundlich model is an empirical equation encompasses the heterogeneity of the adsorbent surface, the exponential distribution sites and their energies. The linear form of Langmuir isotherm is expressed as follows:

$$\frac{1}{q_e} = \frac{1}{Q_0} + \frac{1}{Q_0 b C_e} \quad (1)$$

where,  $q_e$  is the ratio of hardness or turbidity adsorbed to the dosage of the MO,  $C_e$  is the equilibrium concentration of adsorbate (residue hardness, mg L<sup>-1</sup> as CaCO<sub>3</sub>),  $Q_0$  is the adsorption capacity of MO and  $b$  is a constant related

to the energy of adsorption. The values of Langmuir constants (slope) and adsorption capacity (intercept) were obtained from the linear correlation plots between  $1/q_e$  with  $1/C_e$  (Fig. 4b, 5b). With Freundlich isotherm, the linear equation is expressed as the follows:

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (2)$$

where, as  $K_f$  is the adsorption capacity of MO and  $n$  is a constant related to the intensity of adsorption. The values of Freundlich constants (slope) and adsorption capacity (intercept) were obtained from the linear correlation plots between  $\ln q_e$  with  $\ln C_e$ .

**Analytical method:** The pH of samples in laboratory experiment was measured using the Hanna H98150 pH meter. Turbidity value was determined using the turbidimeter model HACH 2100N series with USEPA Method 180.1. The hardness of calcium and magnesium was determined using HACH DR2800 Spectrophotometer.

## RESULTS AND DISCUSSION

### Turbidity and hardness removal in natural surface water:

The residual turbidity and hardness in Timah Tasoh raw water is shown in Fig. 1. In the initial stage with the absence of MO, the hardness and turbidity of the raw water were 350 mg L<sup>-1</sup> and 21 NTU, respectively. The residual turbidity was significantly decreased to 4.1 NTU at MO dosage of 15 mg L<sup>-1</sup>. At this point onwards the residual turbidity was observed varying over MO dosage ranging between 4.3 to 7.0 NTU. The behavior of total residual hardness was found similar to residual turbidity. In the early stage, the total residual hardness was observed decreasing with MO dosage. A sharp concentration decreased to 109.8 mg L<sup>-1</sup> was found at

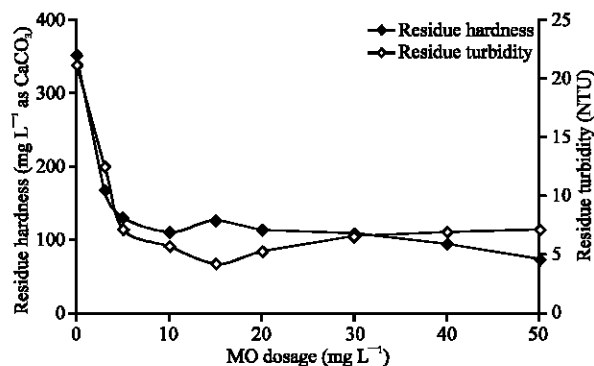


Fig. 1: Residual turbidity and hardness in Timah Tasoh raw water at various MO dosages

MO dosage of 15 mg L<sup>-1</sup>. The concentration of residual hardness continues to decrease gradually from this point onwards. The behaviors of both compounds suggest that the optimum removal efficiency for turbidity and hardness may achieve at different MO dosage. The optimum turbidity removal efficiency was 80%, whereas the percentage removal for hardness tends to increased with higher MO dosage.

In previous study, Okuda *et al.* (1999) reported that the optimum turbidity removal efficiency for synthetic turbid water with initial turbidity of 35 NTU was approximately 97% using MO-NaCl. In another study, Muyibi and Evison (1995a) documented the surface water with initial hardness of ca. 1000 mg L<sup>-1</sup> as CaCO<sub>3</sub> was decreased gradually to 280 mg L<sup>-1</sup> as CaCO<sub>3</sub> when 2400 mg L<sup>-1</sup> of MO that extracted with distilled water was applied. The differences in turbidity and hardness removal might be due to difference in experimental setting employed in respective studies, such as initial concentrations, settling time, MO extraction method, mixing velocity gradient and mixing durations (Katayon *et al.*, 2005). The use of difference species of MO might also influence the efficiency of hardness removal contained in the raw water.

**Removal of turbidity in synthetic and natural surface water:**

The removal of turbidity in natural surface water, synthetic turbid water and synthetic hard and turbid water by using MO that extracted by sodium chloride is shown in Fig. 2. The initial concentration of turbidity and hardness in synthetic water has been set based on Timah Tasoh raw water data at 21 NTU. Initially, the turbidity concentration for all water samples tend to be decreased with MO dosage until the optimum turbidity removal efficiencies approaching 80% (Fig. 2). Nonetheless, each water samples has its specific removal trend. Although, Timah Tasoh raw water and synthetic turbid and hard water exhibit similar removal characteristic, the synthetic turbid and hard water reached its optimizing point at lowest MO dose approximately 10 mg L<sup>-1</sup>. Moreover, the residual turbidity in the synthetic turbid water was relatively higher which imply that it has a lower removal capacity in compare to other water samples. The optimum MO dosage for Timah Tasoh raw water, synthetic turbid water and synthetic turbid and hard water were attained at 15, 30 and 10 mg L<sup>-1</sup>, respectively. The difference in the optimum MO dosage suggested that hardness and other water quality parameter could possibly affect the coagulation activity of MO.

Okuda *et al.* (2001b) reported that the coagulation activity was higher in water sources containing bivalent cations such as Mg<sup>2+</sup>, Ca<sup>2+</sup> or Ba<sup>2+</sup>. The occurrence of

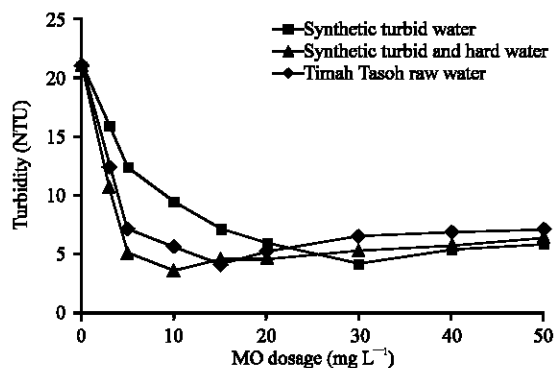


Fig. 2: Residual turbidity in Timah Tasoh raw water, synthetic turbid water and synthetic turbid and hard water

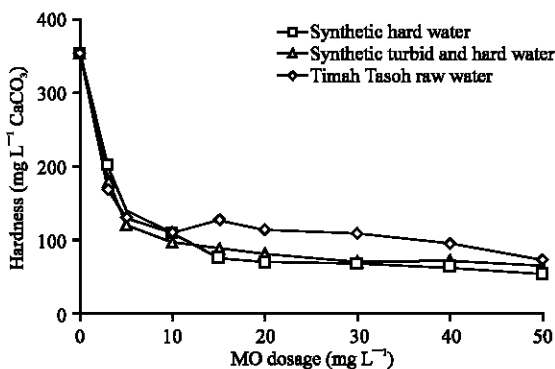


Fig. 3: Residual hardness in Timah Tasoh raw water, synthetic hard water and synthetic turbid and hard water

matter to form flocs and eventually increase of coagulation activity of MO-NaCl. Therefore, the faster turbidity removal in Timah Tasoh raw water and synthetic turbid and hard water could possibly due to the presence of Mg<sup>2+</sup> and Ca<sup>2+</sup> ion in the water.

**Removal of hardness in synthetic and natural surface water:**

The removal of residual hardness of Timah Tasoh raw water, synthetic hard water and synthetic turbid and hard water is shown in Fig. 3. The removal efficiency of hardness was observed to increase with increasing dosage of MO. The highest MO dosage (e.g., 50 mg L<sup>-1</sup>) in this experiment indicates the highest removal of hardness to less than 90 mg L<sup>-1</sup>. This pattern is suggested to decline with the increase in MO dosage. At MO dosage between 10 to 30 mg L<sup>-1</sup>, the removal of hardness in all samples ranging between 60 to 130 mg L<sup>-1</sup> CaCO<sub>3</sub>. The same dosage was found to be the most efficient for turbidity removal as shown in Fig. 2.

However, the residual hardness at these concentrations is considered as moderate classes (Sincero and Sincero, 2003). While at high hardness concentrations up to more than 1000 mg L<sup>-1</sup> as CaCO<sub>3</sub>, a relatively higher MO dosage at approximately 2400 mg L<sup>-1</sup> is required to efficiently remove the hardness in the water (Muyibi and Evison, 1995a). This indicates the potential advantage in the use of MO seeds extracted with sodium hydroxide solution in softening hard waters.

**Mechanism of simultaneous removal of hardness and turbidity:** Figure 4a and b shows the respective parameters of Freundlich and Langmuir Adsorption Equilibrium Model for turbidity removal. It was observed that only synthetic turbid water shows positive correlation with Freundlich and Langmuir models with R<sup>2</sup> of 0.7 and 0.5, respectively. However, these models were found not fit for Timah Tasoh raw water and synthetic turbid water and hard water with both R<sup>2</sup> of less than 0.1. This shows that both waters were not fit for turbidity removal using Freundlich and Langmuir Adsorption models. The evaluation of hardness removal by using Freundlich and Langmuir Adsorption Equilibrium Models are shown in Fig. 5a and b. Positive correlation was found for all water samples with Freundlich and Langmuir adsorption models. The R<sup>2</sup> for Timah Tasoh raw water, synthetic hard water and synthetic turbid and hard water was 0.7, 0.8 and 0.8, respectively. This indicates that the adsorption and neutralization mechanisms are most likely involved in the process.

Ndabigengesere *et al.* (1995) proposed that the mechanism of turbidity removal with MO that extracted by distilled water appears to consist of adsorption and neutralization of the colloidal charges. Furthermore, Muyibi and Evison (1995a) suggested that the mechanism for softening of hard water by MO was found to be due to adsorption with the adsorption isotherm approximating to the Langmuir type and conversion of soluble hardness-causing ions to insoluble products by precipitation reactions. On the other hand, Okuda *et al.* (2001b) suggested that coagulation mechanism for turbidity removal with MO that extracted by sodium chloride seemed to be an enmeshment by the insoluble matters formed by the coagulation active component in MO. Other coagulation mechanisms such as compression of double layer, inter-particle bridging or charge neutralization were not responsible for the coagulation by MO.

As shown in Fig. 4a and b, only synthetic turbid water shows strong correlation with adsorption phenomena for turbidity removal. Although, Timah Tasoh raw water and synthetic turbid and hard water did not fit

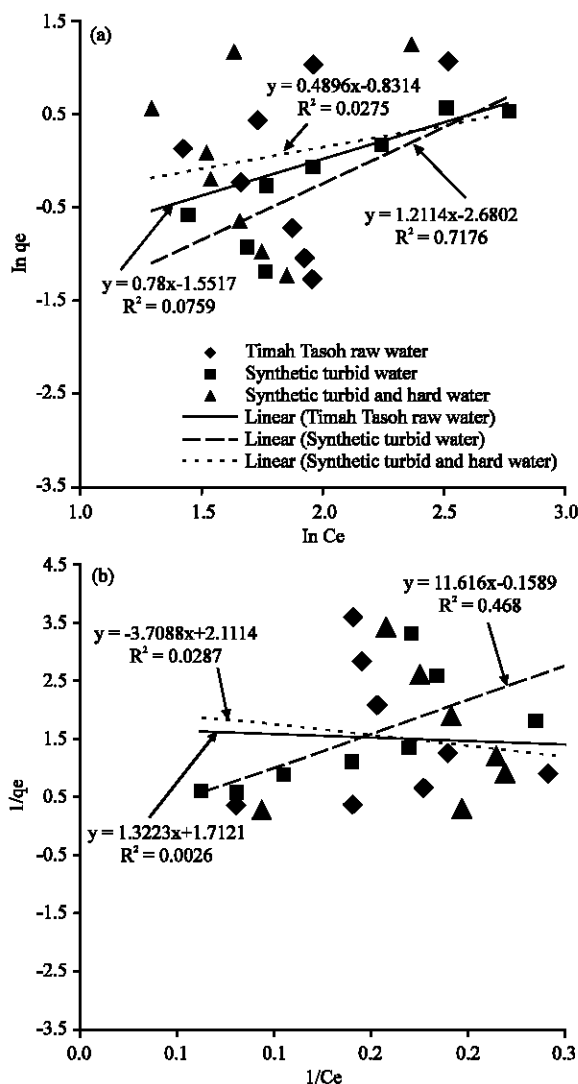


Fig. 4 (a-b): Freundlich and Langmuir adsorption isotherm for turbidity removal in Timah Tasoh raw water, synthetic turbid water and synthetic turbid and hard water. (a) Freundlich adsorption isotherm and (b) Langmuir adsorption isotherm

with Freundlich and Langmuir adsorption model, these waters exhibit higher coagulation activity for turbidity removal as shown in Fig. 2. This occurrence can be explained by the fact that MO tends to remove turbidity in soft water by adsorption and more likely to adsorb hardness in hard water to form a netlike structure followed by turbidity removal through sweep coagulation mechanism (Okuda *et al.*, 2001b). Therefore, MO that extracted by sodium chloride solution can remove both hardness and turbidity contents simultaneously.

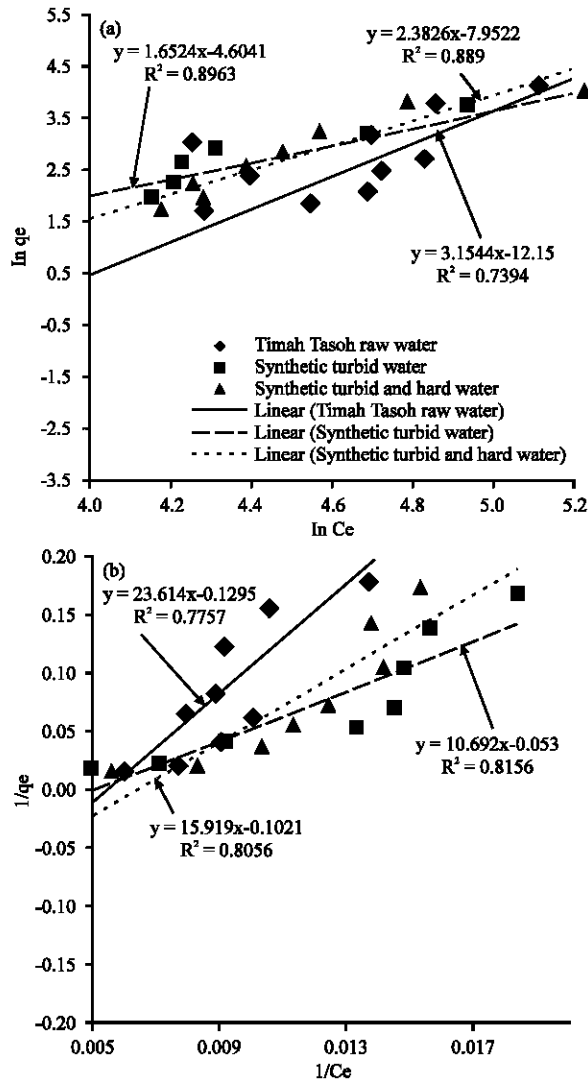


Fig. 5 (a-b): Freundlich and Langmuir Adsorption Isotherm for hardness removal in Timah Tasoh raw water, synthetic turbid water and synthetic turbid and hard water. (a) Freundlich adsorption isotherm and (b) Langmuir adsorption isotherm

**CONCLUSIONS**

The objective of this study is to clarify the mechanism of simultaneous removal of hardness and turbidity in raw water for drinking water purpose by using *Moringa oleifera* (MO). The raw water contain high concentrations of hardness from Timah Tasoh Dam (Perlis, Malaysia) and synthetic water (e.g., turbid water; hard water; turbid and hard water) were used in this study. The findings were obtained as follows:

- The optimum MO dosage for turbidity removal in Timah Tasoh raw water, synthetic turbid water and synthetic turbid and hard water were attained at 15, 30 and 10 mg L<sup>-1</sup>, with the removal efficiencies approximately 80%. However, this optimum MO dosage was not applicable for hardness removal
- The mechanism for turbidity removal by MO-NaCl for synthetic turbid water (soft water) supposed to be adsorption and charge neutralization, with the adsorption isotherm following the Freundlich adsorption model. Further, the mechanism for hardness removal in hard water seems to be adsorption that conforms to both Freundlich and Langmuir adsorption model. Therefore, for hard water such as Timah Tasoh raw water and synthetic turbid and hard water, MO-NaCl tends to adsorb hardness to form a netlike structure and followed by turbidity removal through sweep coagulation mechanism. Consequently, hardness and turbidity can be removed simultaneously by MO-NaCl

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