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Enhanced removal efficiency of Methylene blue and water hardness using NaOH-modified Durian and Passion fruit peel adsorbents

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Abstract

This study evaluated the effectiveness of NaOH-modified adsorbents prepared from durian and passion fruit peels on removal of methylene blue (MB) and water hardness from aqueous solution. The removal efficiencies (adsorbent dose and contact time) of sodium hydroxide (NaOH) modified adsorbents were compared to the raw adsorbents. MB and water hardness removal efficiencies have been greatly enhanced after the durian and passion fruit peels are modified by NaOH. NaOH-modified durian and passion fruit peel adsorbents at 0.05 g dose enhanced the removal of MB from aqueous solution up to 21.56% and 7.30% in comparison to raw adsorbents. This showed that NaOH-modified durian peel is a better adsorbent for MB removal with low dose but able to achieve high removal efficiency. Meanwhile, removal of MB reached maximum at 0.20 g dose using NaOH-modified passion fruit peel adsorbent. NaOH-modified passion fruit peel adsorbent showed better performance in water hardness removal as compared to NaOH-modified durian at 1.40 g dose.

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1 Introduction

Water is essential to the living beings for surviving. Sufficient supply of clean water to fulfil the need for the growing population, marine and plant life has become a major concern recently. However, water has been polluted by impurities and contaminates from various sources nowadays. Wastewater discharge from industries contained high level of pollutants such as heavy metal, high calcium and magnesium, dyes, depending on the process nature of the industries. Long term industrial wastewater discharge has contaminated water sources. Hence, water is no longer safe for direct consumption. A series of water treatment processes, therefore, has been developed to remove these pollutants. Adsorption is one of the intensively studied method for removal of these pollutants.

To ease environment burden, fruit peels and agricultural wastes have been explored as alternative materials for adsorbents in removal of heavy metal, dyes, and water hardness from aqueous solution recently. Durian peels have been attempted as adsorbents to remove methylene blue (MB) dye [1,2], water hardness [3,4], zinc [5], and bisphenol [6]. Passion fruit peels have been found their potential used in removing MB [7], water hardness [4], and lead [8]. Other fruit peels such as rambutan [9], jackfruit [10], pomelo [11,12], dragon fruit [13], citrus limetta [14] and agro-wastes of pinecone [15] and rejected tea [16] have been utilized as adsorbents for removal of MB. For water hardness removal, adsorbents prepared from pineapple peel [17], jackfruit peel [18], cactus materials [19], wheat straw [19], rice husk [19,20], canola stalk [20], and coconut shell [21] have been attempted.

Durian and passion fruit peels have been reported comprise high percentile of peel (~50 to 60%) of the whole fruit [22]. These fruit peels are normally disposed of as waste and long-term disposal would

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burden the environment. Therefore, this study aimed to investigate their potential as low-cost bio-sorbents for removal of MB and water hardness. The removal efficiency of durian and passion fruit peel adsorbents (raw and NaOH-modified) of MB and water hardness was investigated by subjecting in different dose and contact times into aqueous solution.

2 Materials and Method

2.1 Preparation of Raw and NaOH-Modified Adsorbents from Peels of Durian and Passion Fruit

Peels of durian and passion fruit were collected from local fruit market located at Cheras, Malaysia. The collected peels of durian and passion fruit were washed with distilled water for several times in order to remove dirt. The peels were cut into small pieces before drying in a hot air oven at 80 °C for 24 hours (Fig. 1). The dried samples were ground using a blender and segregated to 1mm particle size using a kitchen sieve. The ground fruit peel samples were stored in airtight containers and labelled accordingly.

For preparation of NaOH-modified adsorbent, 40 g of dried durian and passion fruit peel powders were immersed in 400 mL of 0.1 M NaOH solution for 24 hours, respectively. 0.1 M NaOH solution was prepared by dissolving 3.2 g of NaOH pallets into 800 mL of distilled water. After 24 hours of immersion time, fruit peel powder was filtered and washed with distilled water for several times. Rinsed powders were then dried in an oven at 60 °C for 24 hours. The dried NaOH-modified samples were segregated using 1-mm particle size kitchen sieve (Fig. 1). The NaOH-modified adsorbents were stored in an air-tight container and labelled. The efficiency NaOH-modified adsorbent on removal methylene blue (MB) and water hardness is investigated and compared to raw (unmodified) adsorbent.

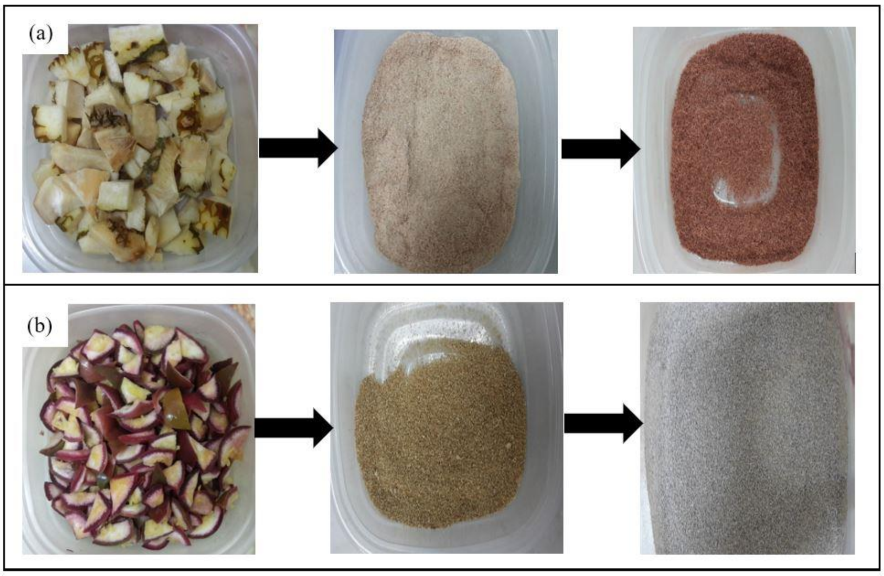

Fig. 1 Peels of (a) durian and (b) passion fruit cut into pieces, ground to powder form and NaOH modified.

2.2 Removal of Methylene Blue (MB) using Raw and NaOH-Modified Adsorbents Prepared from Peels of Durian and Passion Fruit

For preparation of 100 mg/L MB stock solution, 0.1 g of MB was dissolved into 1 L distilled water in a volumetric flask. The stock solution was diluted to concentrations of 5, 10, 15, 20, 25, 30 and 35 mg/L. The absorbance of each concentration was measured at wavelength 668 nm using an UV-Vis spectro-photometer [10]. A calibration curve of MB concentration against absorbance value was plotted (R^2 = 0.910). MB solution of 35 mg/L concentration was used as adsorbate.

The efficiency of durian and passion fruit peel adsorbents (raw and NaOH-modified) on MB removal was investigated by subjecting varying dose (0.05, 0.2, 0.4, 0.6 and 0.8 g) into 100 mL MB aqueous solution with initial concentration of 35 mg/L in a beaker. The mixture was stirred at 350 rpm for 60 min. Effect of contact times of 12, 24, 36, 48 and 60 min on MB removal was investigated as well. Adsorbent dosage of 0.8 g was added into 100 mL of MB solution of 35 mg/L concentration and stirred at 350 rpm. Experiments were conducted in duplicate. Mixture was then filtered using filter paper to separate MB aqueous solution (filtrate) from the adsorbent. The absorbance of filtrate was measured using UV-Vis spectrophotometer at 668 nm. Four absorbance values were captured at each experiment. The concentration of filtrate MB solution was determined by referring to the MB calibration curve. Removal efficiency is calculated using Eq. (1) [21].

$$\%\ \text{Removal} = \frac{C_i - C_f}{C_i} \times 100\% \tag{1}$$

where the C_i and C_f (mg/L) is the initial and final concentrations of MB aqueous solutions.

2.3 Removal of Water Hardness using Raw and NaOH-Modified Adsorbents Prepared from Peels of Durian and Passion Fruit

The efficiency of raw and NaOH-modified adsorbents prepared from peels of durian and passion fruit was evaluated by varying dosage (0.2, 0.6, 1.0, 1.4 and 1.8 g) and contact time (15, 30, 45 and 60 min) in a beaker containing 100 mL of synthetic hard water with concentration of 314.16 mg $CaCO_3$/L. The synthetic hard water was prepared by dissolving 0.3028 g of anhydrous calcium chloride and 0.139 g of magnesium sulphate into 200 mL distilled water. The solution was stirred using stirring rod until all the powder was dissolved completely. The solution was transferred into a 1 L volumetric flask and distilled water was top-up to the 1 L mark. For dosage study, the mixture at each dosage was stirred at 350 rpm for 60 min. Meanwhile, for contact time study, a fixed amount of adsorption dose of 0.50 g was used and stirred at 350 rpm for each contact time. The mixture was then filtered to separate the filtrate from the adsorbent. Total water hardness of the filtrate was determined using EDTA titration analysis. 20 mL of filtrate was added with 2 mL of ammonia buffer solution (operate at fume hood) to obtain the pH between 9 and 10, followed by a pinch of Eriochrome Black T (EBT) indicator into the 250 mL Erlenmeyer flask. The filtrate was titrated with 0.01 M EDTA titrant until the colour changed from wine red to blue (end point). Total water hardness was calculated using Eq. (2) [21].

$$\text{Water Hardness (mg CaCO}_3\text{/L)} = \frac{\text{mL of EDTA solutio used} \times 100}{\text{Volume of water samples taken}} \tag{2}$$

2.4 Statistical Analysis

Data were analysed using Microsoft Excel Data Analysis's single factor ANOVA (Analysis of Variance) for significant difference by setting 95% confidence level ($p < 0.05$). Result is expressed as mean value ± standard deviation ($n = 8$).

3 Results and Discussion

3.1 Effect of Adsorbent Dose and Contact Time on Removal of Methylene Blue (MB) using Raw and NaOH-Modified Adsorbents Prepared from Durian and Passion Fruit Peel

Fig. 2 presents the final MB concentrations and percent removal after subjecting different doses (0.05, 0.2, 0.4, 0.6 and 0.8 g) of raw and NaOH-modified adsorbents prepared from durian and passion fruit peel into 100 mL MB aqueous solution of initial concentration of 35 mg/L for 60 min. It was observed that the MB concentration was reduced significantly with increasing dose using adsorbents prepared from raw durian (Fig. 2a) and passion fruit (Fig. 2b) peels. Raw passion fruit peel (81.39%) has higher removal efficiency in comparison to durian peel (73.82%) at 0.05 g adsorbent dose. There was no difference between the two raw adsorbents in removal efficiency at adsorption dose of 0.20 g. However, raw passion fruit peel showed higher ($p < 0.05$) removal efficiency in comparison to durian peel at 0.40 g, 0.60 g, and 0.80 g adsorbent dose.

NaOH-modified durian and passion fruit peel adsorbents enhanced the percent removal of MB from aqueous solution up to 21.56% and 7.30% compared to raw adsorbents at 0.05 g dose. This showed that NaOH-modified durian peel is a good potential adsorbent in MB removal from aqueous solution with low dose but is able to achieve high removal efficiency. Percent removal of MB from aqueous solution using NaOH-modified passion fruit peel adsorbent increased from 88.69% to 95.15% at dose from 0.05

g to 0.20 g. However, there was no significant increment on percent removal for adsorbent doses above 0.20 g (Fig. 2b). This implied that 0.20 g of NaOH-modified passion fruit peel adsorbent is an optimum dose for maximum removal of MB from aqueous solution.

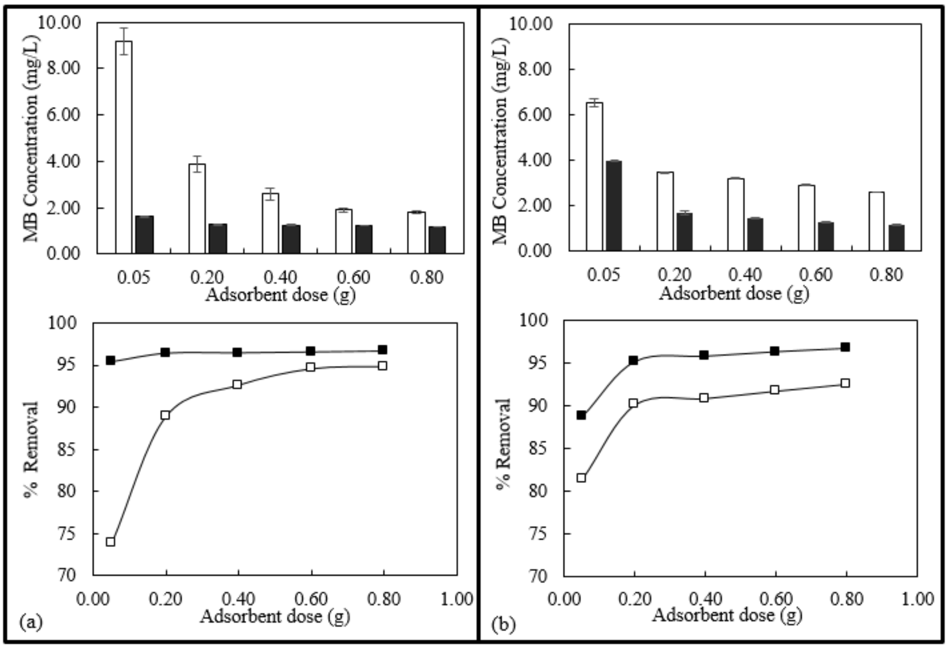

Fig. 2 Effect of adsorption dose on final MB concentration and removal efficiency using raw (□) and NaOH-modified (■) adsorbents prepared from peels of (a) durian and (b) passion fruit peel.

The increase of removal efficiency of MB with increasing dose could be linked to adsorption surface. A greater adsorption surface resulted more available sites for MB molecules to be adsorbed, leading to the uptake of more molecules from solution in high doses at fixed contact time [23]. Nevertheless, adsorption dose is varied with biomaterials. For example, Kallel et al. [23] reported that raw garlic straw dose of 0.04 g gave optimum in removal efficiency from MB aqueous solution of 50 mg/L. On the other hand, Tanzim and Abedin [11] discovered that 1.0 g dose of raw pomelo peel adsorbent gave the maximum removal efficiency of MB solution of initial concentration of 1000 mg/L. Geed et al [24] reported that the highest percent removal (62%) of MB was at 0.50 g dose from an initial concentration of 100 mg/L using coconut shell biochar.

Effects of contact times of 12, 24, 36, 48, and 60 min on final MB concentration and percent removal using fixed dose of 0.8 g of raw and NaOH-modified adsorbents prepared from peels of durian and passion fruit are illustrated in Fig. 3. Results showed that NaOH-modified of both durian and passion fruit peel adsorbents enhanced MB removal efficiencies by 3.75% and 8.42%, respectively, at 12 min contact time, in comparison to raw adsorbents. Both NaOH-modified adsorbents achieved approximately 96% removal efficiency at all contact times studied. There was no significant trend in percent removal of MB with increasing contact time. This was most probably due to high adsorbent dose used in conducting this experiment. High dose provides enough adsorption surface for binding and uptake of MB molecules from aqueous solution even in a short contact time. Therefore, for evaluation of contact time on MB removal, low doses of 0.05 g and 0.20 g of adsorbents prepared from peels of durian and passion fruit respectively, are recommended.

Mallampati et al. [13] reported that avocado and dragon fruit peel adsorbents showed maximum removal efficiency of 63% and 60% at equilibrium condition within 3 hours and 5 hours contact time, respectively. Kallel et al [23] observed that the contact time curve shows that the MB removal was rapid in the first 15 min using garlic straw adsorbent for a period of 300 min. There was no significant variation in MB concentration detected after 100 min [23]. An optimum contact time of 80 min achieved removal percentage to nearing 93% was found when 1.0 g pomelo peel adsorbent was used with MB solution concentration of 100 mg/L [11]. Almost all the active sites of the adsorbent might have been saturated after 80 min of contact time, hence, increasing the contact time to 120 min resulted a decrease in percent removal.

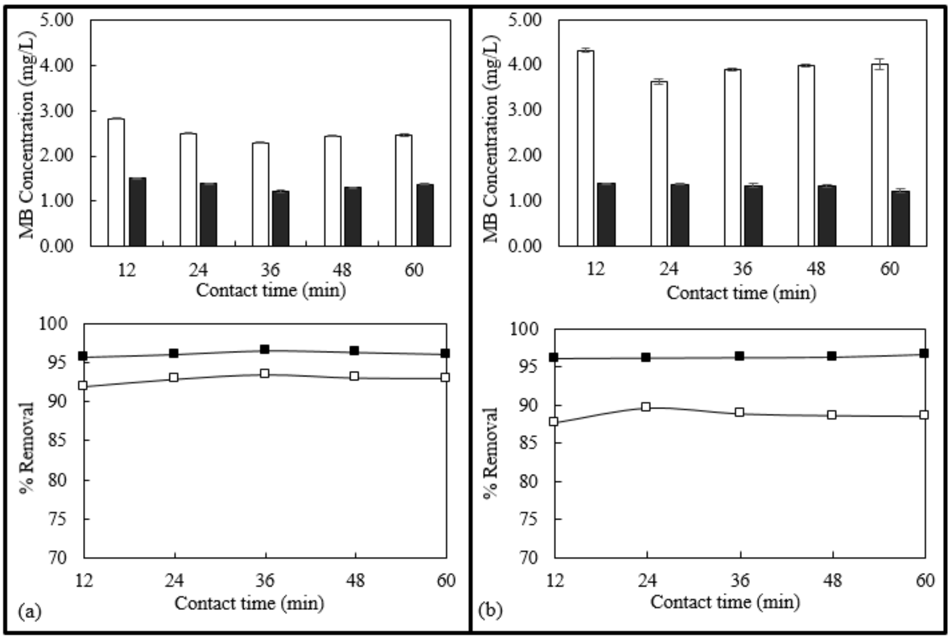

Fig. 3 Effect of contact time on final MB concentration and removal efficiency using raw (□) and NaOH-modified (■) adsorbents prepared from peels of (a) durian and (b) passion fruit peel.

3.2 Effect of Adsorbent Dose and Contact Time on Removal of Methylene Blue (MB) using Raw and NaOH-Modified Adsorbents Prepared from Durian and Passion Fruit Peel

Fig. 4 presents the efficiency of raw and NaOH-modified adsorbents prepared from peels of durian and passion fruit evaluated at varying doses of 0.20, 0.60, 1.00, 1.40 and 1.80 g for 60 min in a beaker containing 100 mL of synthetic hard water of 314.16 mg $CaCO_3$/L initial concentration. Percent removal of water hardness was increased with increasing dose from 0.20 g to 1.80 g for both raw adsorbents prepared from durian and passion fruit peels. For raw durian peel adsorbent, no significant difference ($p > 0.05$) was observed in water hardness removal between dose 0.20 g and 0.60 g. However, in higher dose of 1.00 g, 1.40 g and 1.80 g, percent removal of water hardness was increased significantly (Fig. 4a). Result revealed that for raw durian peel adsorbent, optimum dose of 1.80 g exhibiting the best water hardness removal efficiency at which 36.34% percent removal with final total water hardness of 201.25 mg $CaCO_3$/L achieved. On the other hand, raw passion fruit peel adsorbent recorded a maximum removal of 36.74% with final water hardness of 198.75 mg $CaCO_3$/L at higher dose of 1.80 g (Fig. 4b). Removal efficiency of water hardness using raw durian peel absorbent observed in this study has similar trend but different extent with previous study done Payus et al. [3]. This study demonstrated removal efficiency of water hardness increased gradually from 9.28% to 36.34% with increasing adsorbent dose from 0.20 to 1.80 g (Fig. 3a). Meanwhile, Payus et al. [3] observed that there was a significant rise of removal efficiency from 7 to 18% with the increase of the adsorbent dosage from 0.5 to 2.5 g/l by the durian husk. Similar trend of water hardness removal was observed although onto different adsorbents. Mgombezi et al. [18] reported that the percentage removal of water hardness increased as cactus adsorbent dosage increasing from 0.5 g to 3.5 g. They recorded the maximum removal efficiency of 80.7 % at 3.5 g adsorbent dosage.

Water hardness removal efficiency was greatly enhanced after the durian and passion fruit peels modified by NaOH, in comparison to raw adsorbents. At 1.40 g dose, percent removal of water hardness was enhanced by 8.76% and 42.89% using NaOH-modified durian and passion fruit peel adsorbents. For NaOH-modified durian peel adsorbent, it showed the same trend as raw adsorbent in water hardness removal but in greater extent. The highest removal efficiency of 45.89% with final water hardness of 170.00 mg $CaCO_3$/L was obtained using dose of 1.80 g (Fig. 4a). The present study has slight contradict observation with previous study done by Tan et al. [4]. They reported that NaOH-citric acid (CA) modified durian peel adsorbent has lower water hardness removal efficiency than the raw adsorbent. They explained this was most probably during the NaOH-CA modification process, most of the durian peel residues were washed away when rinsing with distilled water as durian peel fibre consists of 48.6% of cellulose, 10.2% of hemicellulose, and 10.1% of lignin. Hence, adsorbent produced with less amount of cellulose, hemicellulose and lignin resulted lesser hydroxyl groups that could be modified, therefore, led to less efficiency of water hardness removal [4].

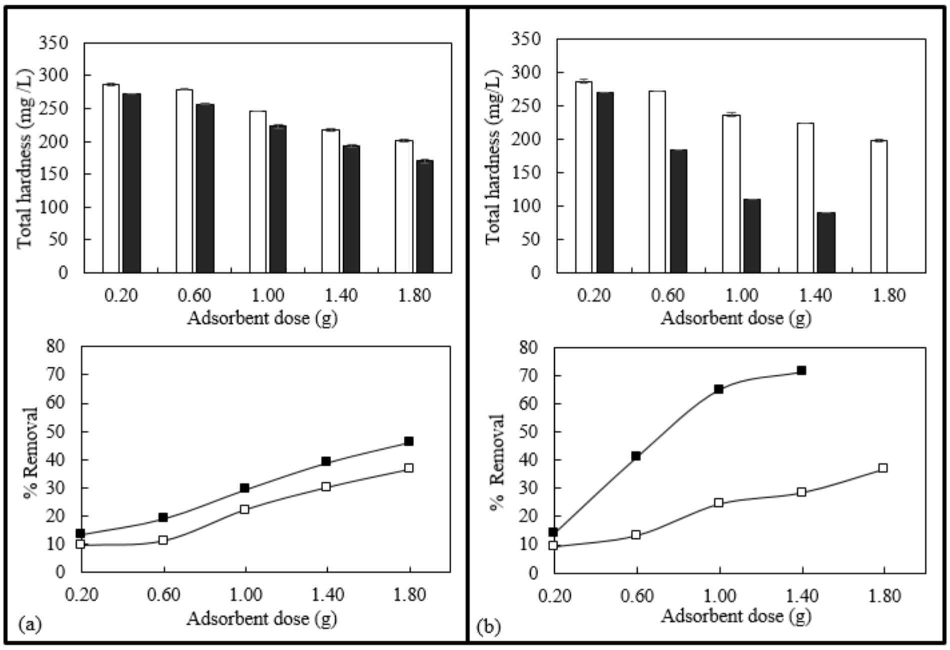

Fig. 4 Effect of adsorption dose on total water hardness and removal efficiency using raw (□) and NaOH-modified (■) adsorbents prepared from peels of (a) durian and (b) passion fruit peel.

The removal efficiency increased greatly up to 42.89% at 1.40 g dose using NaOH-modified passion fruit peel adsorbent, in comparison to raw adsorbent (Fig. 4b). This result is in agreement with Tan et al. [4]. They observed that NaOH-CA modified passion fruit peel adsorbent enhanced water hardness removal greater than raw adsorbent after 60 min at 1.0 g dose. This study noticed that water hardness could not be quantified using EDTA titration after the synthetic water sample was treated for 60 min at dose of 1.80 g using NaOH-modified passion fruit peel adsorbent. This was probably the trace amount of calcium and magnesium ions left in the treated water were not sufficient to react with EBT indicators to produce wine red colour complexes [19].

Both NaOH-modified durain and passion fruit peel adsorbents have been found more effective in water hardness removal than the raw adsorbents. This can be explained that higher surface area increases the probability of ions migration to the surface of the adsorbent [20]. According to Feng et al. [25], adsorbent modified with NaOH solution enhanced the capacity of ions uptake due to more irregular and porous structure leading to more specific surface area compared to raw fruit peel.

The effect of contact time was studied at every 15 min interval to 60 min using 0.50 g of adsorption dose (Fig. 5). The initial water hardness was 314.16 mg/L. Water hardness removal efficiency increased merely 1.60% from 15 min to 30 min using both raw durian and passion fruit peel adsorbents. Decline of water hardness removal efficiencies of 6.37% and 3.18% were observed for raw durian and passion fruit peel adsorbents, respectively, from 30 min to 60 min contact time. Both NaOH-modified durian and passion fruit peel adsorbents showed the same trend as raw adsorbents in water hardness removal, but at higher efficiency. Water hardness removal efficiency was about 16.71% enhanced at 30 min contact time using NaOH-modified durian and passion fruit peel adsorbents as compared to raw adsorbents. A decline of 13.53% water hardness removal efficiency was recorded from 30 min to 60 min using NaOH-modified durian peel adsorbent (Fig. 5a). For NaOH-modified passion fruit peel adsorbent, an increase of 5.57% was observed from 30 to 45 min contact time, however, a slight decline of 2.39% was shown from 45 to 60 min (Fig. 5b). Results suggested that the optimum contact times for NaOH-modified durian and passion fruit peel adsorbents achieving maximum removal of water hardness were 30 min and 45 min, respectively.

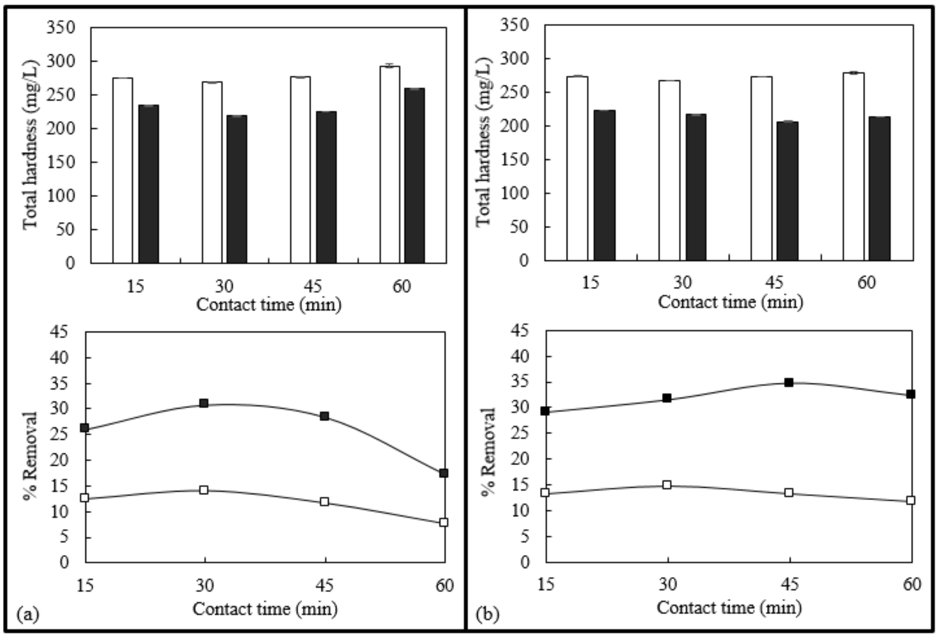

Fig. 5 Effect of contact time on total water hardness and removal efficiency using raw (□) and NaOH-modified (■) adsorbents prepared from peels of (a) durian and (b) passion fruit peel.

Further increase in contact time did not enhanced the hardness removal efficiency. This is due to reduction of availability sites for adsorption with the passage of time. There were repulsive forces between the ions on the surface which created difficulty for the ions to occupy on the adsorbent surface [21]. The porosity of the surface of adsorbent acts as the active sites for adsorption and increase surface area [8,15,17]. The data in the present study was exhibited similar trend compared with previously reported work [21]. They observed that the best contact time was 40 minutes with removal efficiency of 90% in time range from 10 min to 50 min onto coconut shell activated carbon for removal water hardness. The percentage hardness removal approached equilibrium within 10 hours with optimum removal efficiency about 68%. Water hardness removal efficiency increased rapidly at the beginning, gradually decreased and finally reached equilibrium [21].

4 Conclusion

This study revealed that both NaOH-modified durian and passion fruit peel adsorbents greatly enhanced removal of MB and water hardness from aqueous solution, in comparison to raw absorbents. NaOH-modified durian and passion fruit peel adsorbents at 0.05 g dose enhanced the removal of MB from aqueous solution up to 21.56% and 7.30% as compared to raw adsorbents. NaOH-modified durian peel adsorbent showed better performance in MB removal even though in lower dose. NaOH-modified passion fruit peel adsorbent achieved maximum MB removal at 0.20 g dose. No significant trend was observed for removal of MB with increasing contact time because high adsorbent dose of 0.8 g was used in the experiment. High dose provides enough adsorption surface for binding and uptake of MB molecules from aqueous solution even in a short contact time. On the other hand, NaOH-modified durian and passion fruit peel adsorbents have been discovered to be able to enhance water hardness removal by 8.76% and 42.89% respectively at 1.40 g dose. The optimum contact times for NaOH-modified durian and passion fruit peel adsorbents to achieve maximum removal of water hardness were 30 min and 45 min, respectively. Results of this study concluded that NaOH-modified adsorbents prepared from durian and passion fruit peels could be used as low-cost adsorbents in removal of MB and water hardness for industrial wastewater treatment.

Declaration of Conflict of Interest

The authors declared that there is no conflict of interest with any other party on the publication of the current work.

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