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# Removal of Methylene Blue From Aqueous Solutions by Adsorption on Alum Sludge: Effect of Operating Parameters

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**Abstract**
In developing countries, high capital, operational costs, limited space are the main challenges to implement appropriate sludge treatment system. Thus, research about reutilization sludge are urgently needed. In this study, alum sludge from sedimentation basin of water treatment plant was dried and used as adsorbent for removal methylene blue from aqueous solutions. Alum sludge was charachterized by SEM-EDX, XRD, FTIR, particle size analyzer, surface area analyzer and $pH_{PZC.}$ The effect of various parameters such as: contact time (5-360 min), initial MB concentrarion (5-100 mg/L), adsorbent dosage (0.25-3.0 g/L), pH (4-10), temperature (20-60 $^0$C) and water matrix (deionized, sedimentation basin effluent, river water, wastewater) were assessed. The result showed that removal efficiency has directly proportional with contact time, adsorbent dosage and pH, while an inverse relationship with initial MB concentration was observed. Removal efficiency up to 98% was achieved at MB 50 mg/L, AS 3 g/L, temperature 30$^0$, and pH 6.8. Isotherm and kinetic models fitted Langmuir model with capacity of 37.45 mg/g and pseudo-second-order model kinetic. Applicability test in various water matrix indicated that the utilization of low-cost adsorbent from non-modified dried alum sludge has potential for removal of methylene blue from low to moderate contaminated aqueous solution.
**Keywords:** *sludge reuse, wastewater treatment, adsorption, operating parameters, water matrix*

**Abstrak**
Di negara-negara berkembang, modal yang tinggi, biaya operasional, keterbatasan lahan merupakan tantangan utama untuk menerapkan sistem pengolahan lumpur yang tepat. Dengan demikian, penelitian tentang pemanfaatan kembali lumpur sangat dibutuhkan. Dalam penelitian ini, lumpur alum dari bak sedimentasi instalasi pengolahan air dikeringkan dan digunakan sebagai adsorben untuk menghilangkan metilen biru dari air. Lumpur tawas dikarakterisasi dengan SEM-EDX, XRD, FTIR, penganalisa ukuran partikel, penganalisa luas permukaan dan pHPZC. Efek dari berbagai parameter seperti: waktu kontak (5-360 menit), konsentrasi MB awal (5-100 mg/L), dosis adsorben (0,25-3,0 g/L), pH (4-10), suhu (20-60 0C) dan matriks air (deionisasi, efluen bak sedimentasi, air sungai, air limbah) dinilai. Hasil penelitian menunjukkan bahwa efisiensi penghilangan berbanding lurus dengan waktu kontak, dosis adsorben dan pH, sedangkan hubungan terbalik diamati dengan konsentrasi awal MB. Efisiensi penghilangan hingga 98% dicapai pada MB 50 mg/L, AS 3 g/L, suhu 300, dan pH 6,8. Model isoterm dan kinetik sesuai dengan model Langmuir dengan kapasitas 37,45 mg/g dan model kinetik pseudo orde kedua. Uji penerapan dalam berbagai matriks air menunjukkan bahwa pemanfaatan adsorben murah dari lumpur tawas kering yang tidak dimodifikasi memiliki potensi untuk menghilangkan metilen biru dari larutan berair yang terkontaminasi rendah hingga sedang.
**Kata Kunci:** *pemanfaatan kembali lumpur, pengolahan air limbah, adsorpsi, parameter operasional, matrik air*

## 1. Introduction

Urbanization, population and economic growth drive a significant increase of municipal drinking water supply. Provision of urban drinking water is carried out at the Water Treatment Plant (WTP) through the treatment of raw water (lake water, rivers, etc.) into drinking water. The treatment process aims to separate various solids, organic and inorganic compounds, bacteria, andviruses found in raw water to produce drinking water in accordance with applicable regulations. In conventional WTP, removal of various contaminants is performed by a series of unit operation and process namely coagulation, flocculation, sedimentation, filtration and disinfection [1]. Removed contaminants will accumulate in the sedimentation and filtration units in the form of sludge which must be managed before being discharged into landfills. It is estimated, a conventional WTP generates sludge residues of around 40,000-100,000 tons / year [2]. Previous works reported that sludge production from WTPs in Depok City reached up to 4854 kg/day or 0.47 kg sludge/m$^3$ treated water [3,4].

Besides the large quantity, Fresh slurry sludge also contains high concentrations of solids and organic compounds (TSS ~ 2,500-52,000 mg/L and COD ~ 9,600 mg/L) as well as various inorganic compounds including heavy metals [1,3]. The characteristic and composition of sludge vary depending on the condition of raw water and the type of coagulant used during treatment process. Aluminium sulfate ($Al_2(SO_4)_3$) is the most common coagulant used in WTP therefore the sludge formed usually called as alum sludge (AS). Unfortunately, in developing countries such as Indonesia and India, alum sludge is often discharged directly into rivers without adequate treatment [1,3,4]. Some factors such as high capital and operational costs and limited space are probably the main obstacles to implement appropriate sludge treatment system [5–7]. Hence, alternative treatment or utilization of alum sludge is needed to reduce the volume of sludge that must be treated and discharged into rivers or landfills.

Various studies have shown the potential of alum sludge valorization such as; (i) coagulant aid in water and wastewater treatment process, (ii) co-conditioner in sludge treatment, (iii) co-substrate for composting, (iv) constructed wetland substrate, (v) soil ameliorant agent, (vi) landfill cover, (vii) substitute naterials for brick, concrete and pavement, and (viii) adsorbent for water and wastewater treatment [4,7–9]. Among these, the latter is interesting due to their simplicity and possibility to reuse directly in-situ. Indeed, utilization of alum sludge-based adsorbent have been reported for the removal of various organic and inorganic contaminant [10–13] especially dyes [7,9,14,15]. However, most studies used themodified or activated alum sludge as adsorbent while the efficiency non-modified alum sludge itself was rarely studied.

In this paper, efficiency of alum sludge as adsorbent (AS) was evaluated for removal of methylene blue (MB), one of the most widely utilized dyes. Various operating parameter including adsorbent concentration, pollutant concentration, pH, temperature and water matrix were assessed.

## 2. Materials and Methods

### *Preparation of synthetic wastewater and adsorbent*

MB powder was supplied by Sigma-Aldrich, and synthetic dye wastewater was prepared by diluting appropriate amount of MB powder into deionized water. The raw AS was provided by PDAM Tirta Asasta, the Regional Drinking Water Company of Depok City. It was taken from sludge drainage pipe of sedimentation basin at Citayam WTP. The water content was reduced by filtering through cloth filter, air-dried for 5 days and dried in an oven at 105 $^0$C for 1 day. The cooled AS was then crushed into powder and stored in a sealed container.

### *Experimental setup*

Experiments were conducted in a 1 L beaker glass equipped with hot plate magnetic stirrer. The working volume of 500 mL was used. Unless otherwise stated, the experimental conditions were as follows: MB 50 mg/L, AS 1 g/L, temperature 30$^0$, pH 6.8 (not-modified) and agitation speed 300 rpm. The experiment was started by the addition AS into MB solution. Several samples were withdrawn during reaction and separation solid-liquid was performed by centrifuge. Some experiments were duplicated to check for the reproducibility and the observed deviation was always less than 5%.

### *Analytical methods*

The physico-chemical characterisric of AS was first evaluated. Morphology and elemental composition of AS were analyzed by Scanning electron microscope (SEM) (Hitachi SU-3500 equipped by Horiba EDX detector). The structure of crystalline materials and surface functional groups were assessed by X-Ray Diffraction (XRD) (PANalytical:X'PertPro) and Fourier-transform infrared spectroscopy (FTIR) (Thermoscientific Nicolet iS-10). Particle size analyzer (Malvern, Zetasizer Nano ZS) and surface area analyzer (Quantachrome NovaWin) were used to identify particle size distribution and surface area of AS, respectively. Moreover, identification of pH point of zero charge ($pH_{PZC}$) was conducted based on previous study [16]. Concerning liquid sample, quantification of MB in water was performed by DR 5000 UV-Vis Spectrophotometer at wavelength of 663 nm. In addition, COD analysis was also perfomed for some experiment in real water matrix.

## 3. Results and Discussion

### *Characterization of AS*

SEM analysis revealed the morphological structure of AS was porous and had uneven structure which should favorable for adsorption process (**Fig. 1**). Moreover, complementary elemental analysis by EDX detector indicated the presence of oxygen (56.94%), silica (23.22%), aluminium (10.97%), iron (5.33%), and others elements (3.52%).

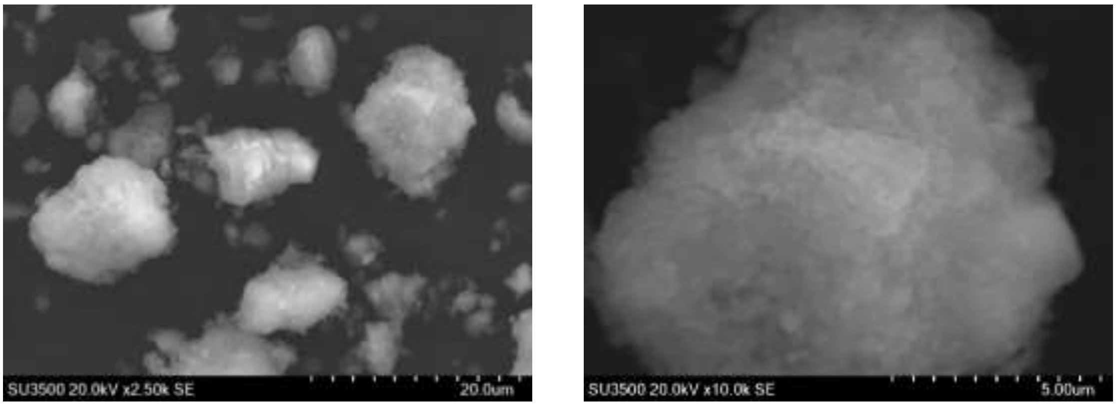



**Fig. 1**. SEM images of AS adsorbent

Furthermore, evaluation by XRD analysis showed that the main minerals composition of AS are quartz and kaolinite, followed by alumina and hermatite in small quantities (**Fig. 2**). Silica oxide is a common material found in AS which comes from river, while the alum content in form of kaolinite and alumina should be originated from the coagulant alum sulfate used by Citayam WTP.

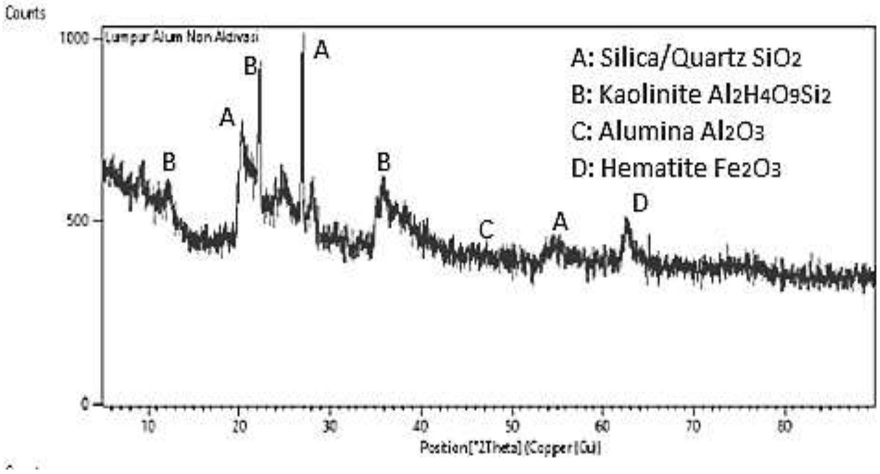



**Fig. 2**. XRD pattern of AS adsorbent

**Figure 3** showed the FTIR analysis of AS recorded at wavenumber ranging from 400 to 4000 $cm^{-1}$. It was observed that AS had strong bands around 3400-3700 $cm^{-1}$ due to stretching vibration of the Al-OH group. Peak around 1600-1700 $cm^{-1}$ indicates the double bond C = O and bending vibration of water (H-O-H) absorbed in the alum sludge. Then, a sharp peak between 1010-850 indicated stretching vibration in the double bond between metal and oxygen O=Al-O-Al=O. In addition, strong peaks in the range 1130-1000 $cm^{-1}$ and 480-170 $cm^{-1}$ indicated asymmetric stretching and bending vibrations from Si-O-Si[3].

Analysis of particle size distribution showed that AS was medium-polydispersed (PDI=0.493) with more than 90% of particle ranging from 1.1 to 2.6 µm. The Brunauer– Emmett–Teller (BET) surface area was found to be 66 $m^2$/gr, comparable with previous reported studies [7,10,17]. Last, evaluation of $pH_{PZC}$ was found to be 6.25.

Jurnal Serambi Engineering JSE Volume IX, No.4, Oktober 2024 Hal 10520 - 10530 p-ISSN : 2528-3561 e-ISSN : 2541-1934

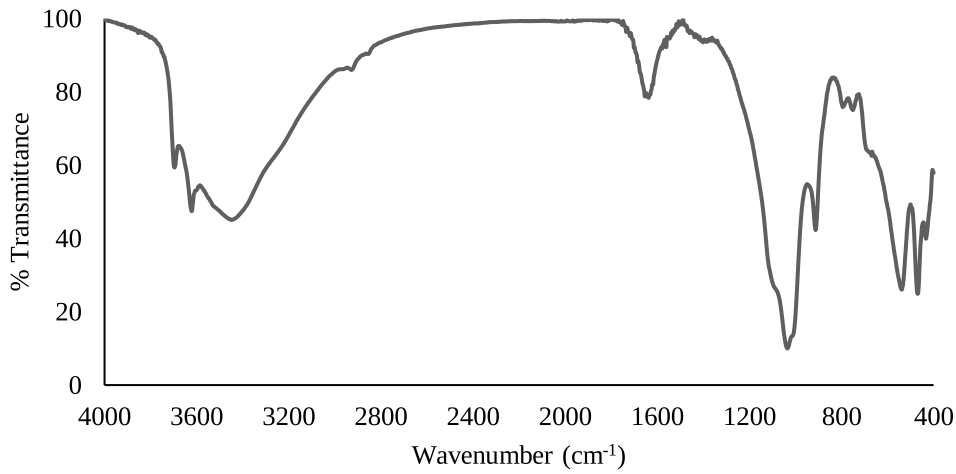



**Fig 3.** FTIR spectrum of AS

***Determination of contact time***

Adsorption capacity of the adsorbent initially increase with an increase in contact time until the the equilibrium is reached and no more adsorption occur. The duration of contact time should be determined experimentally. As shown in **Fig. 4**, it is clear that removal of MB by adsorption process increased with the increasing of contact time. MB removal was notable during the initial 60 min and the plateu was achieved around 180 min. Thus the contact time of 180 min was chosen for the following experiments.

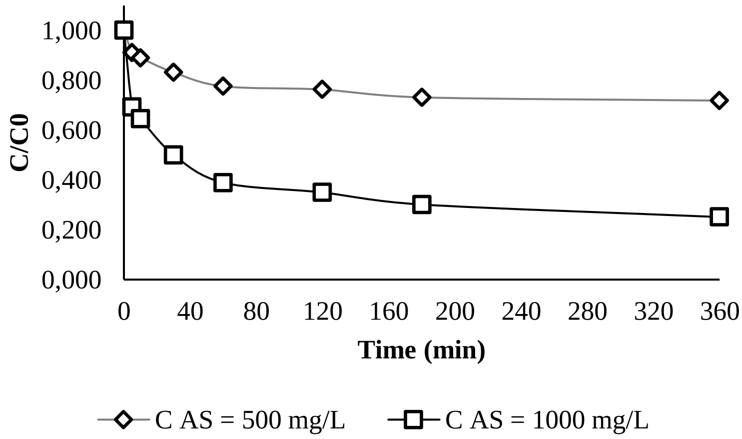
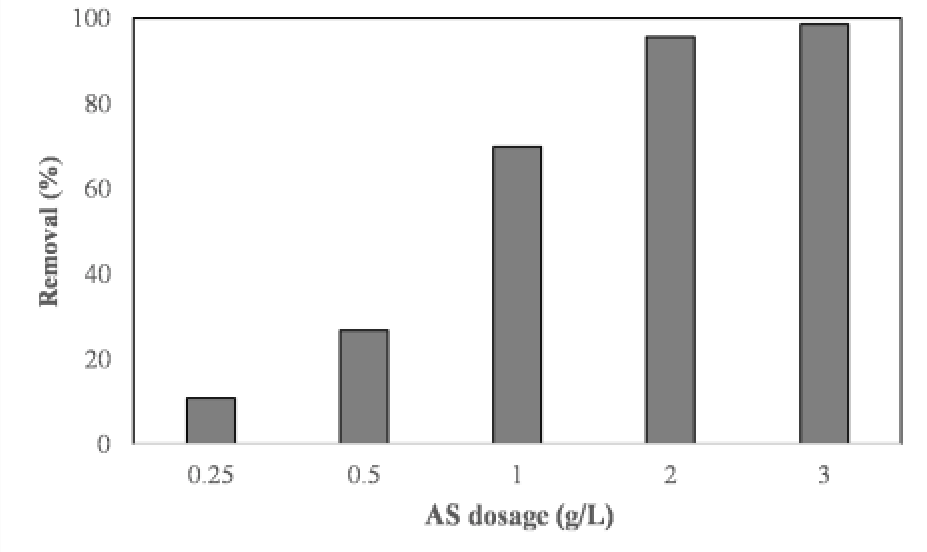



**Fig 4.** Effect of contact time

***Effect of adsorbent concentration***

From **Fig. 5**, it can be seen that MB removal efficiency increases with increase adsorbent concentration. AS dosage of 0.25 g/L; 0.5 g/L; 1 g/L; 2 g/L; and 3 g/L promoted removal of 10.82%; 26.86%; 69.70%; 95.57%; dan 98.85%, respectively. It can be explained by the availability of more number of the binding sites for the adsorption of the pollutants on the adsorbent surface resulting in high removal efficienc [7]. However, an optimum AS dosage was found around 2 g/L. This finding could be explained by the unsaturation of the adsorption sites through the sorption process, or because that particle interaction would result in the diffusion of the dye into the sludge [18,19].

Jurnal Serambi Engineering JSE Volume IX, No.4, Oktober 2024 Hal 10520 - 10530 p-ISSN : 2528-3561 e-ISSN : 2541-1934




***Effect of MB concentration***

The removal efficiency usually depends on the relationship between the initial pollutant concentration and the available active sites on the adsorbent surface. MB concentration was varied between 5 mg/L and 100 mg/L at fixed adsorbent concentration of 1 g/L. The increase of pollutant concentration from 5 mg/L to 25 mg/L did not affect MB removal efficiency (around 96%) because sufficient adsorption site are still greater than the amount of MB to be adsorbed (**Fig. 6**). However, the increasing in MB concentration to 100 mg/L lower the removal efficiency of MB to 37% due to the saturation of adsorption sites [20,21].

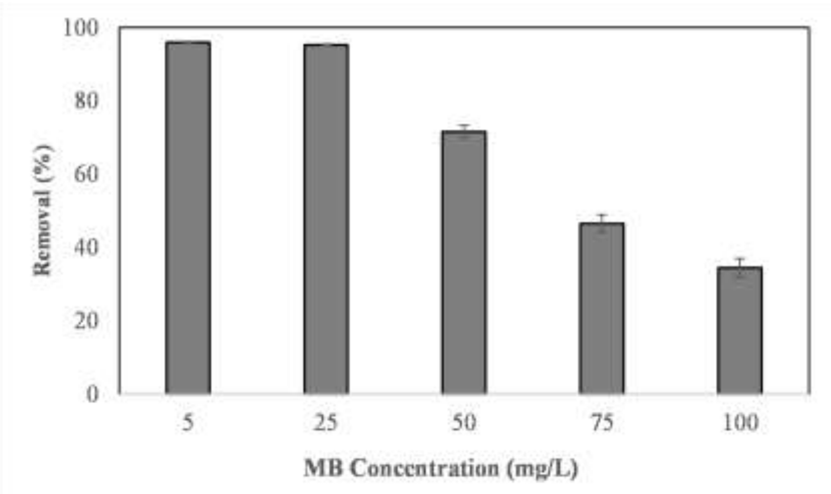



**Fig 6.** Effect of MB initial concentration

***Effect of temperature***

According to literature, temperature may affect adsorption process by: (i) altering the viscocity and solubility of dyes in solution, (ii) swelling the internal structure of the adsorbent, or (iii) damaging the active sites of adsorbent [7,22,23]. Based on **Figure 7**, it also can be seen that the adsorption of MB on AS was relatively independent from the variation of temperature ranging from 20$^0$C to 60$^0$C.

Jurnal Serambi Engineering JSE Volume IX, No.4, Oktober 2024 Hal 10520 - 10530 p-ISSN : 2528-3561 e-ISSN : 2541-1934

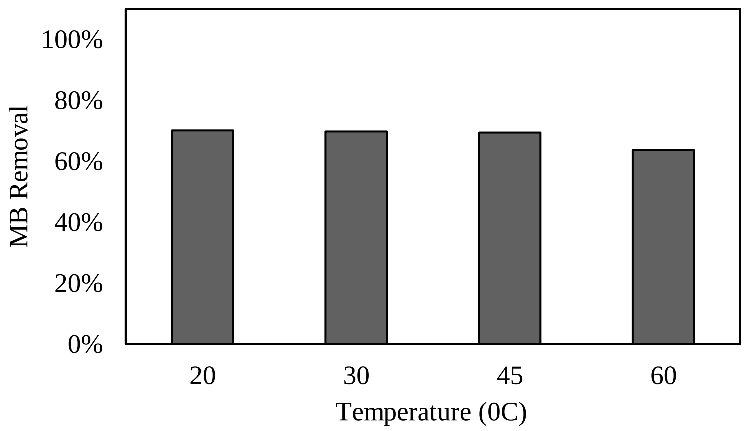



**Fig 7.** Effect of temperature

### *Effect of initial pH solution*

Theoretically, adsorption is a pH-dependent process. Alteration of pH can affect the surface charge and functional groups of the adsorbent (expressed as $pH_{PZC}$) as well as the ionization capacity of the adsorbate (expressed as *pKa*) [7]. The investigation of pH effect was carried out by blocking the assays in acid, neutral and alkaline condition (Table 1.). Under acid condition (2 < pH < 4), solution pH was lower than $pH_{PZC}$ (6.25). This condition causing positive charge on AS adsorbent surface which repelling cationic dyes such as MB. On the contrary, the removal efficency of MB increased with the increasing of pH. It can be explained that at neutral-alkaline pH condition, AS surface was negatively charge, thus attracting ionic form of MB in solution. In addition, comparison of buffer and non-buffered condition showed AS adsorbent might be able to lower the solution pH.

**Table 1.** Effect of solution pH on MB removal

| No | Chemicals added | $pH_0$ | $pH_F$ | pH vs $pH_{PZC}$ | pH vs pKa | % Removal |
|---|---|---|---|---|---|---|
| A | Acid | | | | | |
| A.1 | $H_2SO_4$ | 2.1 | 2.2 | pH < $pH_{pzc}$ | pH < pKa | 54.60% |
| A.2 | $H_2SO_4$ | 4 | 3.9 | pH < $pH_{pzc}$ | pH > pKa | 56.16% |
| A.3 | Buffer pH 4 | 4.4 | 4.5 | pH < $pH_{pzc}$ | pH > pKa | 56.04% |
| B | Neutral | | | | | |
| B.1 | None | 6.8 | 6.8 | pH > $pH_{pzc}$ | pH > pKa | 69.87% |
| B.2 | Buffer pH 7 | 7.4 | 7.5 | pH > $pH_{pzc}$ | pH > pKa | 69.22% |
| C | Alkaline | | | | | |
| C.1 | NaOH | 10 | 7.2 | pH > $pH_{pzc}$ | pH > pKa | 81.14% |
| C.2 | Buffer pH 10 | 8.5 | 8.1 | pH > $pH_{pzc}$ | pH > pKa | 81.49% |

### *Adsorption isotherms*

Adsorption isotherms express the theoretical adsorption capacity for a specific adsorbent for a particular contaminant. In this paper, the adsorption isotherm were evaluated using Langmuir, and Freundlich. From **Fig. 8**, it can be seen that the Langmuir isotherm model fitted the data better than Freundlich and Temkin models, indicating that the monolayer of MB molecules covered AS surface. Indeed, the maximum adsorption capacity of 37.45 mg/g ($K_L$ = 1.103 L/mg ; $R_L$= 0.017 ; $R^2$ = 0.999) was smaller than previous similar studies [14,21] using modified sludge as adsorbent.

Jurnal Serambi Engineering JSE Volume IX, No.4, Oktober 2024 Hal 10520 - 10530 p-ISSN : 2528-3561 e-ISSN : 2541-1934

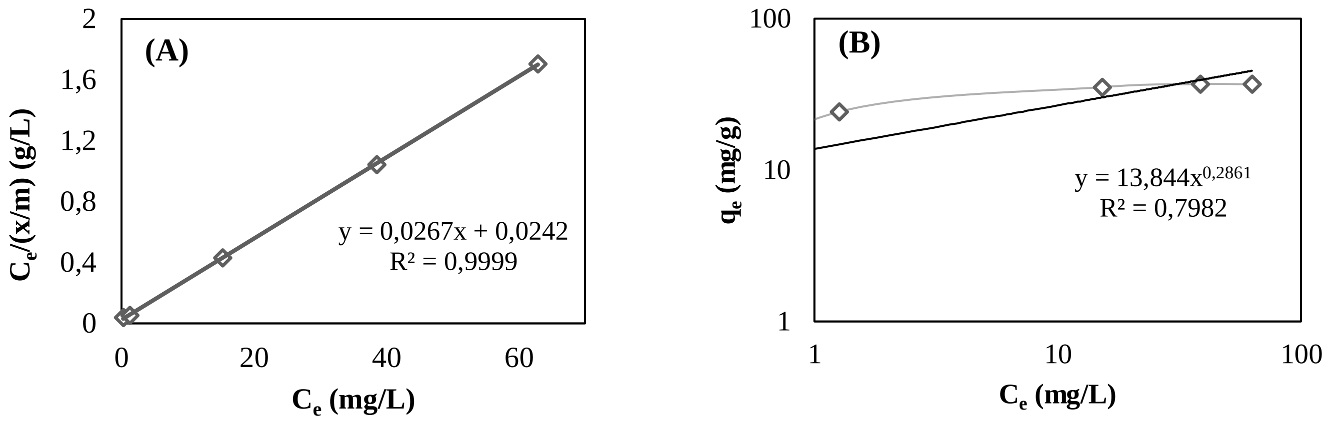



**Fig 8**. Isotherm adsorption using (A) Langmuir, and (B) Freundlich

***Adsorption kinetic***

In an adsorption system, equilibrium is established between the adsorbent and the adsorbate in the bulk phase. The adsorption kinetics is the rate of approach to equilibrium. The adsorption of dye from aqueous solution on adsorbent consists of several stages such as transport in the solution, external diffusion or boundary layer diffusion, internal diffusion or intraparticle diffusion, adsorption or desorption on the surface of the interior sites [24].

In this study the adsorption kinetic study were evaluate using pseudo first order, pseudo second order, and Evolich model. As depicted in **Fig. 9**, it was found that pseudo-second-order kinetic model described well the adsorption of MB on AS. The pseudo-second-order kinetic model assumes that the adsorption rate might be controlled mainly by chemisorptions although the mechanism supposed to be complex and includes concomitant physical adsorption [19,23]. Based on the pseudo-second-order model kinetic, the values of *qe* and $k_2$ were 38.91 mg/g and 0.0018 g/mg.min, respectively.

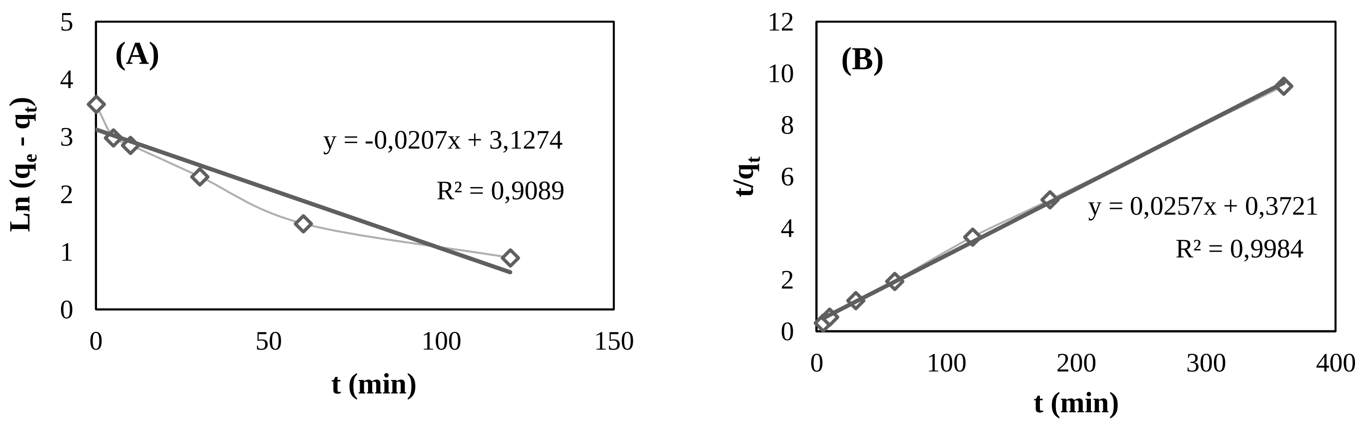



**Fig 9.** Adsorption kinetic using (A) Pseudo-first-order; and (B) Pseudo-second-order

***Reusability***

Despite the numerous available literature on adsorption of dyes,there are still several gaps which needed further studies such as reusability and effect of water matrix or real wastewater [22]. The reusability of AS adsorbent was evaluated using used AS and the result was compared to the fresh one (**Fig. 10**). It was clear that the adsorption capacity of reuse AS was much lower than the fresh one suggesting the reuse AS was already saturated.

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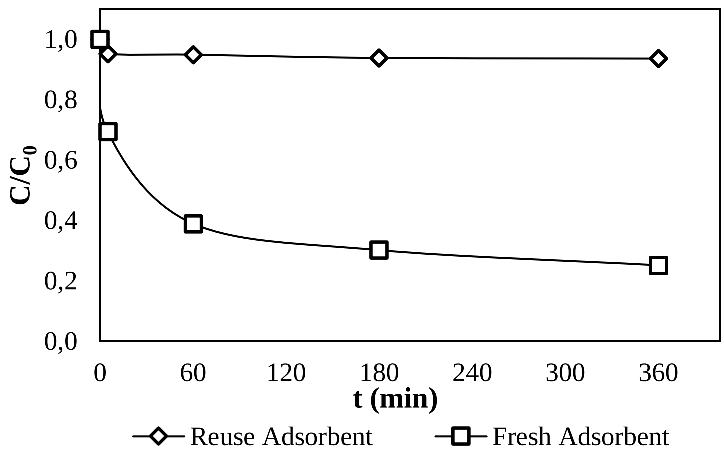


**Fig 10.** Comparison of fresh and reuse adsorbent

***Effect of water matrix***

As per the litterature, effect of water matrix is still rarely studied. Nevertheless, real water matrix or wastewater contains various impurities that may affect the performance of adsorption process by diffusion hidrance, pore blockage or competition for sorption sites [22,25,26]. Adsorption of MB on AS adsorbent was compared in four (4) different water matrix namely deionized water (DW), effluent of sedimentation basin (SW), river water (RW) and canteen wastewater (WW), which are corresponding to non, low, medium and heavy polluted water, respectively. It was found that the adsorption of MB on AS adsorbent was marginally affected by SW (Fig. 11.A). Moreover, the MB removal efficiency was even higher, probably due to the presence of readily adsorbed compounds in RW matrix (Fig. 11.B). On the contrary, adsorption MB on AS adsorbent was significantly hampered in wastewater matrix, where MB removal efficiency reduced from 70% in DW to 30% in WW (**Fig. 11.C**).

**Table 3.** Physicochemical properties of the tested water matrix

| **Parameters** | **SW** | **RW** | **WW** |
|---|---|---|---|
| pH | 6.8 | 6.9 | 4.8 |
| Turbidity (NTU) | 2.48 | 6.72 | 75.35 |
| TDS (mg/L) | 63.5 | 69.9 | 487 |
| COD (mg/L) | 9.0 | 97 | 2,290 |

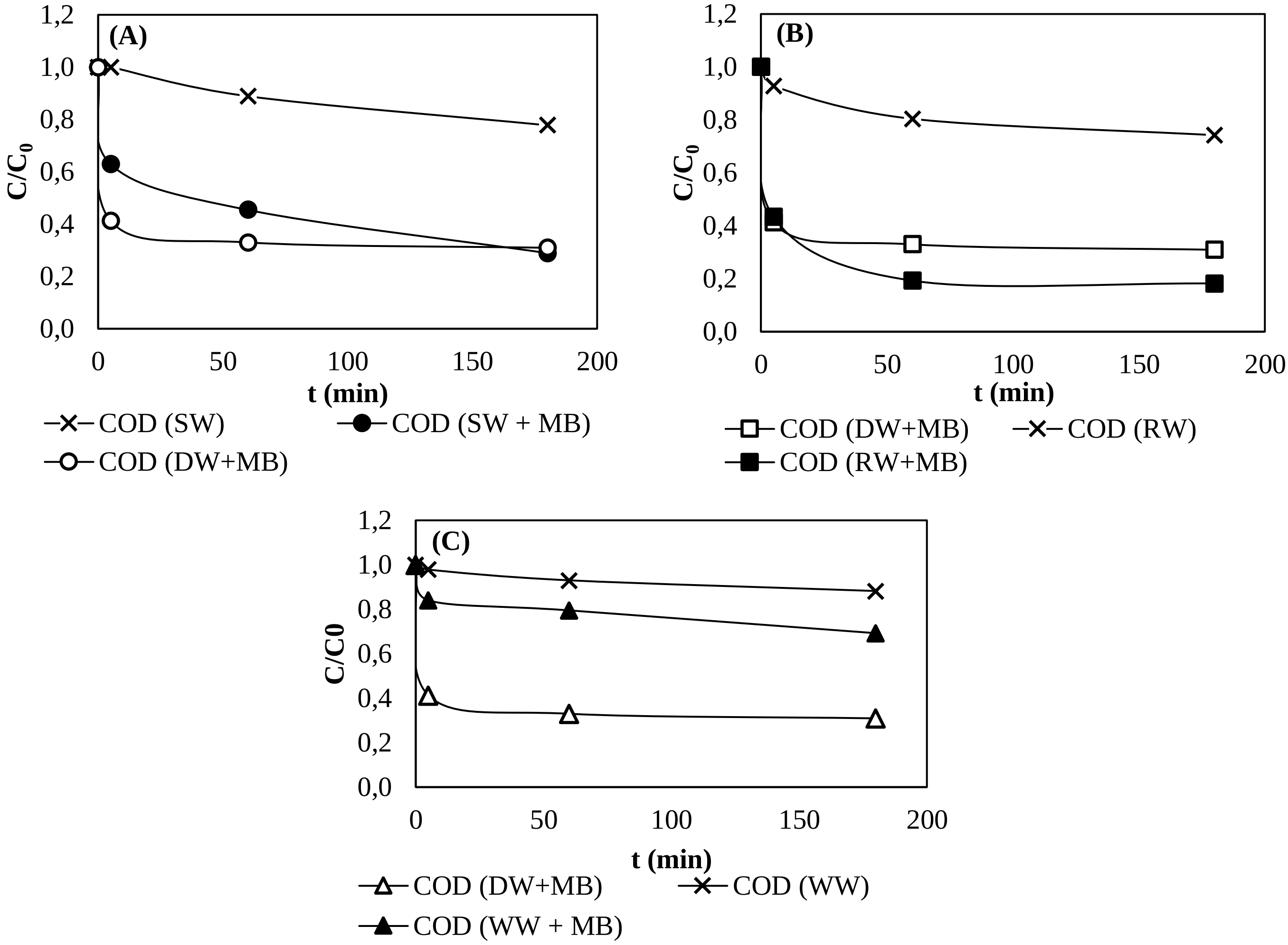


**Fig 11.** Performance of MB adsorption on AS in various water matrix: (A) Effluent of sedimentation basin (SW); (B) River water (RW); (C) Wastewater (WW)

## 4. Conclusion

The non-modified dried alum sludge (AS) from water treatment plant was used as adsorbent to remove Methylene Blue (MB) from synthetic aqueous solutions. The results showed that adsorption process was significantly affected by AS adsorbent concentration, initial MB concentration, and pH, while the effect of temperature was marginal. Adsorption MB on AS adsorbent followed Langmuir model and pseudo-second-order kinetic. Evaluation on adsorbent reusability was found that AS adsorbent was saturated after first trial. In real water matrix, the adsorption efficiency was relatively not affected by effluent of sedimentation water and river water matrix, but hampered in wastewater matrix.

## 5. Acknowledgment

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