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Batch Adsorption of Manganese (II) Ions Using Fruit Peels: Equilibrium and Kinetic Studies

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ABSTRACT

Manganese (Mn) is recognized as a potent neurotoxin, known to elicit severe health implications such as hallucinations, learning impairments, and Mn-induced parkinsonism in both adults and children. In 2021, the World Health Organization (WHO) established a provisional guideline of 0.08 mg/L Mn compared to 0.4 mg/L in 2004 due to mounting evidence underscoring Mn neurotoxicity. Over the years, numerous conventional methods have been explored to address this pressing concern. This study, however, explores an innovative approach focusing on the adsorption process employing langsat, orange and lemon-activated carbons (LOLAC), to remove Mn (II) ions. The primary objective is to determine the optimal conditions for manganese adsorption utilizing LOLAC. Parameters influencing Mn (II) ions biosorption, including pH, biosorbent dosage, initial adsorbate concentration, and contact time, were investigated. Batch mode was adopted for its inherent controllability, while UV-Vis spectrophotometry facilitated initial and final Mn (II) ion concentration measurements. Subsequently, the percentage of Mn (II) ions removed was computed to elucidate adsorption equilibrium and kinetics. Experimental outcomes revealed orange peels as the most efficient, achieving a remarkable 96.3% removal of manganese under conditions of pH 6, 0.8 g/100ml biosorbent dosage, 100 ppm initial adsorbate concentration, and a contact time of 90 minutes. The Langmuir isotherm model and pseudo-second-order kinetics exhibited commendable fitting with the data of Mn adsorption onto LOLAC, indicating a monolayer adsorption mechanism with a second-order reaction rate on the adsorbent. Consequently, activated carbon from orange peel emerges as a promising, cost-effective biosorbent for the remediation of manganese ions from wastewater, outperforming its langsat and lemon peel counterparts.

1.0 Introduction

Water pollution is currently a notable global issue that is mainly caused by the rapid growth of industrial activities. It is found that about 300 to 400 million tonnes of contaminants, including toxic sludge, solvents and heavy metals, have been released into the water bodies every year (Singh, Nagpal & Agrawal, 2018). Due to limited human resources in local government bodies and institutions to monitor the regulation of waste disposal and effluent discharge from industry, the number of polluted water sources in Malaysia continues to increase gradually (Marsidi, Hasan & Abdullah, 2018). The presence of heavy metals, including Manganese (Mn), in water sources, even in a trace amount, is detrimental to aquatic life, human health and the environment (Singh, Nagpal & Agrawal, 2018). The discharge of Mn-rich effluents into the river has alarmed the local authorities and waterwork industries (Rudi et al., 2020).

Mn is an easily oxidized and chemically active metal which is widely used in steel production (Rudi et al., 2020) and can exist as Mn (II), Mn (III) and Mn (IV) ions. Despite possessing an important role in steel production, its presence in water sources could lead to the formation of oxide layers in corroded pipes and lead to deteriorating water quality as well as an increase in the maintenance costs for water distribution (Alvarez-Bastida et al., 2018). Mn also has a vast potential to be present in the consumer's drinking water. According to Gerke, Little, and Maynard (2018), the consumption of Mn into the human body could lead to neurological abnormalities and affect intellectual and cognitive development. Hence, it is essential to identify effective methods to remove Mn from industrial effluents.

Many treatment methods have been employed to treat wastewater containing Mn such as chemical precipitation, ultraviolet irradiation, filtration, ion exchange and oxidation (Rudi et al., 2020; Rahman A., 2022; Du et al., 2019; Ahmadi & Ganjidoust, 2021). However, these treatments suffer serious limitations, such as space consumption, low treatment capacity, high amount of sludge production, complex processes and high operational and maintenance costs (Rudi et al., 2020; Jawed & Pandey, 2019). One of the alternative methods that can aid in the treatment of Mn in water is the adsorption process. This method is less toxic and utilizes low-cost materials used as adsorbent that are also less toxic (Rudi et al., 2020; Shahraki et al., 2023).

Many studies have been conducted on the adsorption treatment process, but there are limited studies on the potential adsorbent from langsat, orange and lemon peels. Therefore, this study aims to explore the use of langsat, orange and lemon peels as adsorbent materials and determine their effectiveness in treating water containing Mn (II) ions. Their efficiency as adsorbent material was analysed through adsorption and kinetic studies. Besides, the effect of different operating conditions for the removal of Mn ions was also studied.

2.0 Literature review

Recent advancements in environmental science and technology have emphasized the importance of sustainable solutions for water treatment, especially in mitigating heavy metal contamination. Fruit peel adsorbents have gathered attention as a potential eco-friendly alternative due to their abundance, low cost, and inherent adsorption properties. In recent years, there has been growing interest in exploring the adsorption properties of fruit peels for the removal of heavy metals from aqueous solutions. The effectiveness of different fruit peel as adsorbents is shown in Table 1.

Table 1: The effectiveness of fruit peel adsorbents

Adsorbent	Manganese removal (%)	References
Banana peels	97.4	(Mahmoud, 2014)
Grafted banana peels	94	(Ali, 2017)
Orange peel	96	(Surovka & Pertile, 2017)
Modified tangerine peel	92.48	(Abdić *et al.,* 2018)

In a study conducted by Mahmoud (2014), it was demonstrated that low-cost banana peels as adsorbents exhibited significant efficacy in removing manganese from aqueous solutions, achieving a remarkable percentage removal rate of 97.4%. Moreover, the adsorption capacity was observed to be contingent upon several factors, including pH, adsorbent dosage, initial metal ion concentration, and contact time.

In a comprehensive investigation by Ali (2017), the adsorption capacity of chemically modified banana peels for the removal of manganese from water was thoroughly explored. The study illuminated the exceptional efficiency of the adsorbent, a testament to its rich composition of functional groups such as hydroxyl groups, carboxylic acids, and phosphates. These multifaceted functional groups were identified as pivotal active centres, orchestrating the effective adsorption of metal ions from aqueous solutions.

Surovka & Pertile (2017) highlighted the practical efficacy of orange peel as a versatile adsorbent for manganese removal from wastewater, achieving an impressive efficiency rate of 96%. The investigation meticulously optimized several crucial parameters, including the solution's pH, the initial concentration of the adsorbent, contact time, and temperature, to maximize removal efficiency.

Abdić et al. (2018) investigated the efficacy of tangerine peel adsorbent for removing manganese from water. Their study revealed that increasing the amount of tangerine peel adsorbent corresponded to heightened efficiency in manganese removal. This phenomenon can be attributed to the augmented surface area and enhanced availability of metal binding sites facilitated by the higher adsorbent dosage.

Despite the efficiency demonstrated by banana, orange, and tangerine peel adsorbents in removing manganese from water, there remains a notable gap in the literature regarding the comprehensive assessment of fruit peel adsorbents specifically for manganese removal. While individual studies have highlighted the potential of various fruit peels, there is a lack of detailed comparative analyses across different types of fruit peels to determine their relative effectiveness and suitability for manganese remediation.

Thus, this study aims to address this gap by evaluating the efficiency of langsat, orange, and lemon peels as adsorbent materials for the removal of Mn (II) ions from water. By conducting adsorption and kinetic studies, this research seeks to provide insights into the potential of fruit peel adsorbents as a viable and sustainable solution for mitigating Mn contamination in water sources. This study contributes to the existing literature by assessing the efficacy of langsat, orange, and lemon peels as adsorbents for Mn removal, thereby advancing our understanding of sustainable approaches to water treatment.

3.0 Methodology

3.1 Adsorbate solution

To prepare a stock solution of manganese ions with a concentration of 1000 ppm of manganese, 3.077 g of $MnSO_4.H_2O$ was weighed and dissolved into 1 litter of distilled water. Working solutions were then derived from the stock solution by using dilution method to the desired volumes.

3.2 Preparation of biosorbents

The experiment involved using peels sourced from oranges, lemons, and langsats, obtained from local markets. These peels underwent thorough washing and multiple rinses with distilled water to eliminate any traces of dirt particles. Subsequently, the cleaned peels were subjected to drying in an oven at 80°C for approximately 48 hours and then grounded using a mortar and pestle. Carbonization of the fruit peels was conducted in a furnace at 500°C for 1 hour in the presence of air. Following activation, any ash content was removed by washing with distilled water, after which the material was dried in an oven at 100°C for 24 hours (Amit et al., 2010). The obtained biosorbents were examined using Fourier-Transform Infrared Spectroscopy (FTIR) to study their functional groups.

3.3 Adsorption experiment

At room temperature, batch adsorption experiments were conducted, examining the three adsorbents (orange, lemon, and langsat) under diverse conditions. These conditions encompassed pH levels ranging from 2 to 8, with pH adjustments facilitated by solutions of HCl and NaOH, biosorbent dosages spanning from 0.2 to 0.8g per 100 ml, initial manganese ion concentrations fluctuating between 100 and 300 ppm, and contact time extending from 30 to 120 minutes. The mixtures were contained within 250 ml conical flasks and subjected to agitation on a mechanical orbital shaker operating at 200 rpm. Subsequent to agitation, filtration through filter paper (size 101) was performed, followed by the determination of manganese (II) ion concentrations via UV-Vis Spectroscopy.

The percentage removal of Mn (II) ions (%) was determined using the following equation:

$$Removal = \frac{C_o - C_e}{C_e}\, x\ 100\% \qquad [1]$$

Where, Co is the initial concentration of Mn (II) ions in ppm and Ce is the equilibrium metal ion concentration in ppm.

The equilibrium amount of metal ion adsorbed by the biosorbent (qe, mg/g) was calculated using the formula:

$$q_e = \frac{(C_o - C_e)V}{m} \qquad [2]$$

Where, V is the volume of the solution treated in litter and m is the biosorbent in grams.

4.0 Discussion of analysis and findings

4.1 Characterization of adsorbents

Examining adsorbents through FTIR is a crucial technique to determine the presence of functional groups essential for facilitating adsorption behaviour. By comparing the spectrum of LOLAC (Lemon, Orange, Langsat Activated Carbons), one can identify the functional groups responsible for adsorbing manganese (II) ions. The spectrum of LOLAC shows broad absorption peaks around 3750 - 3900 cm^{-1}, indicating O-H (hydroxyl) stretching vibrations. The adsorption band at 2945 cm^{-1} is likely associated with the stretching vibration of C-H bonds in aliphatic (saturated) hydrocarbon chains, such as those present in alkane groups. Additionally, stretching vibration bands observed at 2250 and 2400 cm^{-1} can be attributed to the presence of carbon dioxide (CO_2) gas, reflecting the stretching vibrations of the C=O bonds in CO2 molecules (Silverstein et al., 2014).

4.2 Effect of pH

The research findings, as depicted in Figure 4.1, illustrate the effect of initial pH on Mn (II) ions adsorption onto LOLAC. The results indicate a notable increase in adsorption with rising pH from 3 to 6, followed by a significant decrease beyond pH 6.0. Consequently, subsequent investigations were conducted under pH 6.0 conditions. At lower pH levels, the abundance of H$^+$ ions results in competition with Mn (II) ions onto the adsorbent's surface, hindering access to binding sites and thus leading to reduced adsorption. Conversely, as pH increases, the diminished presence of protons reduces this competition, enhancing the adsorption efficiency of Mn (II) ions. Furthermore, at higher pH, the speciation of Mn (II) ions may promote the formation of soluble hydroxide complexes, thereby diminishing the concentration of Mn (II) ions available for adsorption onto the solid surface (Wang et al., 2017).

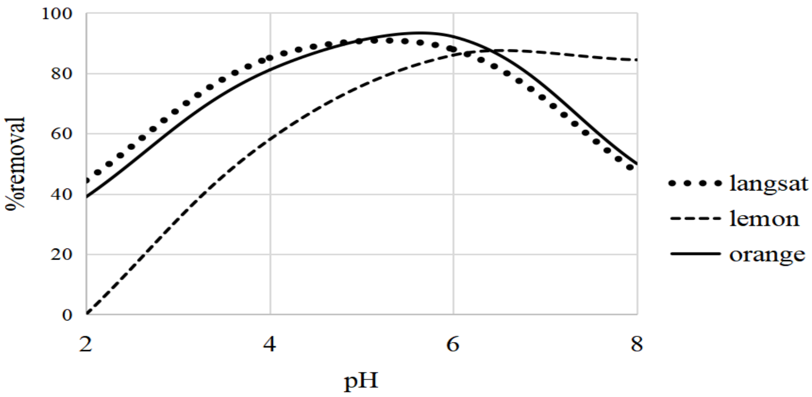

Figure 4.1: Graph of percentage removal of Mn (II) ions against initial pH at 25 ℃ (0.8 g of LOLAC dosage, 100ml of 100ppm Mn (II) ions solution, 120 min)

4.3 Effect of adsorbent dose

In Figure 4.2, the impact of varying adsorbent doses on the removal of Mn(II) ions is illustrated. This parameter is of considerable significance as it directly influences the adsorption capacity of the biosorbent. The investigation revealed a positive correlation between removal efficiency and the increase in adsorbent dosage at room temperature across all three types of adsorbents. Particularly noteworthy is the observation that the utilization of 0.8g of orange biosorbent resulted in the highest removal efficiency, reaching up to 92.13%. This improvement can be attributed to the increased availability of adsorption sites resulting from the higher adsorbent dosage.

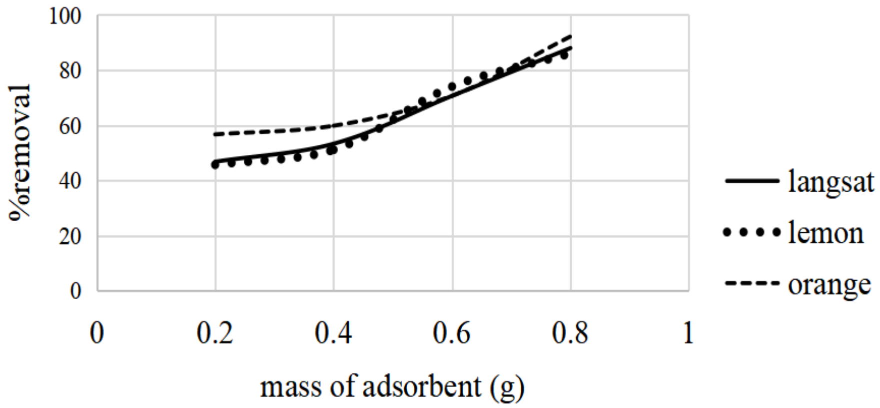

Figure 4.2: Graph of percentage removal of Mn (II) ions against mass of adsorbent at 25℃ (100ml of 100ppm Mn (II) ions solution at pH 6, 120 min)

4.4 Effect of contact time

Equilibrium time plays a pivotal role in the optimization of cost-effective wastewater treatment systems as it indicates when an adsorbent has reached its maximum capacity, facilitating process optimization. Increasing contact time initially enhances removal rates until saturation occurs, after which additional time provides diminishing returns. The comparative analysis of percentage Mn(II) ion removal using various adsorbents over time is presented in Figure 4.3. The results indicate a direct correlation between agitation time and Mn(II) ion removal, with equilibrium achieved within 90 minutes for all three biosorbents. This behaviour suggests rapid sorption initially, primarily on the adsorbent's external surface due to significant surface area of adsorbent, followed by equilibrium once the active sites of the adsorbents become fully saturated with adsorbate molecules.

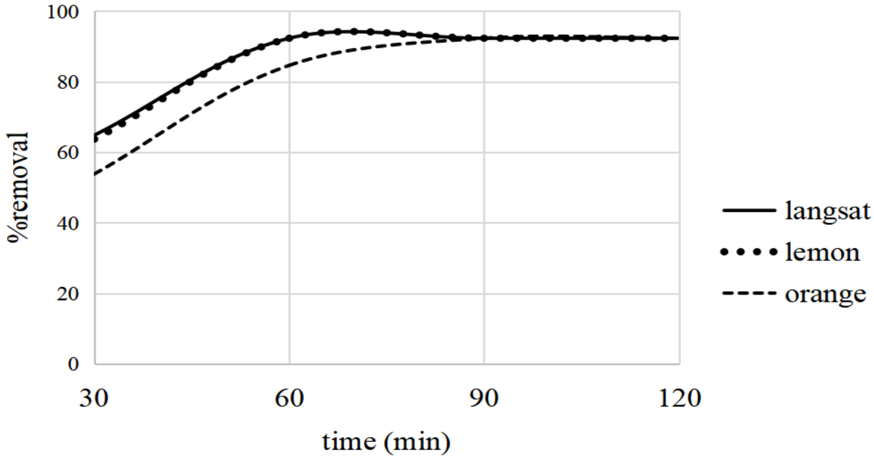

Figure 4.3: Graph of percentage removal of Mn (II) ions against contact time at 25 ℃ (0.8 g of LOLAC dosage, 100ml of 100ppm Mn (II) ions solution at pH 6)

4.5 Effect of initial concentration of adsorbate

Figure 4.4 describes the impact of the initial concentration of Mn (II) ions on their removal by LOLAC. The illustration suggests that the removal of Mn (II) ions diminishes with increasing initial concentration of the adsorbate. This decline occurs because at higher initial concentrations of Mn (II) ions, the available adsorption sites on LOLAC become limited. Consequently, the percentage removal of metal ions is contingent upon their initial concentration. Increasing the biosorbent dosage can mitigate these limitations, allowing for better utilization of available active sites and enhancing the removal efficiency of Mn (II) ions.

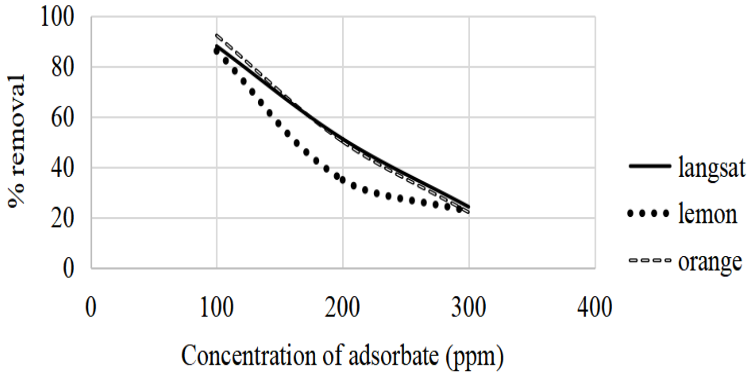

Figure 4.4: Graph of percentage removal of Mn (II) ions against initial concentration of adsorbate solution at 25 ℃ (0.8 g of LOLAC dosage, 100ml of 100ppm Mn (II) ions solution at pH 6, 120 min)

4.6 Adsorption isotherms

Adsorption isotherms are necessary for deciding the adsorption limit of the adsorbent, grasping solute-solution interactions, and surveying the level of Mn (II) particles accumulating on the outer layer of LOLAC. In this examination, two adsorption isotherm models, Langmuir and Freundlich isotherms, were analysed by fitting the adsorption data into the linear form of these isotherms. The outcomes are tabulated in Table 4.1.

Langmuir isotherm:

$$\frac{C_e}{q_e} = \frac{1}{Q_m.b} + \frac{C_e}{Q_m} \qquad [3]$$

Freundlich isotherm:

$$\log q_e = logK_f + \frac{1}{n} logC_e \qquad [4]$$

Where, q_e (mg/g) represents the amount of adsorbed metal ions per unit weight of adsorbent at equilibrium concentration C_e (mg/L). The constants Q_m (mg/g) and b (L/mg) in the Langmuir equation are related to the maximum monolayer capacity and energy of adsorption, respectively. Similarly, K_f and 1/n in the Freundlich equation represent adsorption capacity and intensity of adsorption, respectively.

The equilibrium adsorption isotherms are significant in determining the adsorption capacity of Mn (II) ions and analyse the nature of adsorption onto the LOLAC. In view of the correlation coefficients (R^2) in Table 4.1, it is obvious the adsorption of Mn (II) ions on LOLAC is best fitted to the Langmuir model. The adsorption capacity of Mn (II) ions by LOLAC expressed by Langmuir coefficient, Q_m, demonstrates that adsorption capacity increases in the sequence, orange < lemon < langsat. Since Langmuir addresses the equilibrium distribution of metal ions between the solid and liquid phases, this portrays quantitatively the development of a monolayer adsorbate on the external surface of the adsorbent, and no further adsorption happens after that. This model suggests uniform energies of adsorption onto the surface and no immigration of adsorbate in the plane of the surface (Dada A.O. et al., 2012).

Table 4.1 shows the calculation results of Langmuir and Freundlich model for Mn (II) ions on LOLAC.

Adsorbent	Langmuir model			Freundlich model		
	Q_m	b	R^2	K_f	n	R^2
Langsat	11.429	-0.193	0.992	19.355	-7.052	0.716
Lemon	10.989	0.252	0.995	14.887	-7.994	0.988
Orange	10.604	-0.132	0.989	54.013	-2.791	0.968

4.7 Kinetic studies

To examine the experimental kinetic data, pseudo first- and second-order kinetic equations were studied.

The pseudo first-order rate equation is expressed as:

$$log(q_e - q_t) = log(q_e) - \frac{k_1}{2.303}t \qquad [5]$$

where, q_e (mg/g) and q_t (mg/g) are the amounts of metal ions adsorbed at equilibrium and at time t (min), respectively, while k_1 (min^{-1}) denotes the pseudo first-order rate constant of adsorption.

The pseudo second-order rate can be formulated as follows:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}t \qquad [6]$$

where, k_2 (g/(mg·min)) is the rate constant of second-order adsorption.

The outcomes derived from varying contact time were computed and are displayed in Table 4.2. The R^2 values for the pseudo-second-order kinetic model surpass those for the pseudo-first-order kinetic model, suggesting a preference for the supposition of a second-order adsorption mechanism. This indicates that the overall rate of the metal adsorption process is governed by chemical mechanisms such as ion exchange or complexation, involving valency forces through the trade or sharing of electrons between the adsorbent and adsorbate (Robati D., 2013).

Table 4.2 shows the calculation results of pseudo-first and pseudo-second order kinetic model for Mn (II) ions on LOLAC.

Adsorbent	Pseudo first order			Pseudo second order		
	q_e	k_1	R^2	q_e	k_2 (10^3)	R^2
Langsat	6.25	0.018	0.600	24.038	2,859	0.986
Lemon	6.5	0.019	0.600	24.272	2.651	0.984
Orange	11.574	0.024	0.024	27.397	1.158	0.976

5.0 Conclusion and Future Research

The findings underscore the potential of LOLAC, a low-cost adsorbent, in efficiently removing manganese from aqueous solutions. Key factors such as pH, initial metal ion concentration, adsorbent dose, and contact time were identified as significant influencers on LOLAC's manganese adsorption capacity. Optimal removal occurred at pH 6.0, with adsorption rates positively correlated with higher doses of adsorbent and longer contact times, but inversely proportional to increasing metal ion concentrations. Through a series of experiments, it was found that orange peels exhibited the highest efficiency in adsorbing Mn (II) ions, achieving an impressive removal efficiency of 96.3% under specific conditions (pH 6, 0.8 g/100ml biosorbent dosage, 100 ppm initial adsorbate concentration and 90 minutes contact time), thus highlighting their potential as an effective adsorbent for manganese removal. The Langmuir isotherm and pseudo-second-order model were identified as the most suitable models for describing the equilibrium adsorption data. Future research avenues may entail investigating alternative agricultural waste for activated carbon production, potentially leading to the discovery of more effective bio-sorbents for pollutant removal.

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Author Contributions

Kartikah S.: Conceptualization, Methodology, Experimental Design, Writing- Original Draft Preparation; **Azini Amiza H.**: Data Collection, Data Analysis, Validation, Supervision; **Siti Fatimah S.**: Data Interpretation, Writing-Reviewing and Editing; **Sanjith U.**: Writing-Reviewing and Editing.

Conflicts of Interest

The manuscript has not been published elsewhere and is not being considered by other journals. All authors have approved the review, agree with its submission and declare no conflict of interest in the manuscript.

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