International Journal of Integrated Engineering: Special Issue 2018:
Innovations in Civil Engineering, Vol. 10 No. 9 (2018) p. 172-177

DOI: https://doi.org/10.30880/ijie.2018.10.09.031

Chitosan ultilization in biocomposite adsorbent in Iron (Fe) removal from landfill leachate

Zawawi Daud[1*], Halizah Awang[2], Farah Nur Diyana Ibrahim[1], Nur Adila Ab Aziz[1], Mohd Baharudin Ridzuan[1], Zulkifli Ahmad[1], Mahmoud Hijab Abubakar[1], Husnul Azan Tajarudin[3]

[1]Centre of Advance Research for Integrated Solid Waste Management (CARISMA), Faculty of Civil and Environment Engineering, UTHM, Johor, Malaysia
[2]Faculty of Technical and Vocational Education, Universiti Tun Hussein Onn Malaysia, Johor, Malaysia
[3]Bioprocess Technology Division, School of Industrial Technology, Universiti Sains Malaysia, Gelugor 11800, Malaysia

Received 22 March 2018; accepted 29 December 2018, available online 31 December 2018

Abstract: Leachate are very high strength wastewaters that contain a variety of pollutants that pose a serious threat to the environment if appropriate control measure is ignored. Composite adsorbent is an emerging, interesting and attractive alternative to conventional adsorbents and having the ability to act as catalysts due to their high reactivity and excellent selectivity towards specific pollutant compounds. This study investigated the potential of biocomposite adsorbent made from a combination of chitosan, feldspar and zeolite (CFZ) for the treatment of Iron (Fe) from leachate wastewater. Leachate characterization and batch adsorption experiments was conducted to determine the optimum conditions for pH, dosage and contact time parameter in the removal of Fe. The result shows that the concentration of Fe was 15.82 which exceeded the recommended limit. The optimum conditions also occurred at pH 5 with 6 gram of biocomposite dosage and at 180 minutes contact time. The corresponding removal efficiency for Fe is 90% with 0.0127 mg/g uptake capacity.

Keywords: Biocomposite, Leachate, Adsorption, Chitosan

1.0 INTRODUCTION

Recently, advancement in technology has improved the quality of life with better facilities and infrastructures. It contributes to the rapid development of an area, resulting urbanization and high resource consumption [1]. Unfortunately, rapid urbanization gives impact towards most countries and one of the biggest impacts of rapid urbanization is the increase in a waste generation [2], especially in developing country.

Hence, in managing the increased waste, landfilling becomes prominent choice for disposing solid wastes in Malaysia [3]. The landfill is cost-effective and simple, compared to the other common disposal method such as incinerator, where this method is costly and requires technological experts to operate it. Unfortunately, the production of heavily contaminated wastewater namely leachate is one of the major drawbacks for landfilling [4]. The presence of leachate is threatening and cause detrimental effect on the survival of aquatic life form, ecology, and food chains [5]. Therefore, appropriate treatment is required to reduce the impact of discharged leachate towards the environment.

Numerous techniques were developed and it could be categorized as physical, chemical, and biological [6]. The suitability of leachate treatment technique is dependable on leachate characteristics. For instance, biological treatment has shown efficiency in eliminating organic matter in early stage when BOD/COD ratio of leachate is high. However, when the ratio decreases with the increasing of landfill age, the process become less effective [7], since biological treatment is not suitable to treat old or stabilize leachate that mainly contain recalcitrant matter and varied substances such as ammonia that hinder of biological activity [8].

Out of many available treatment methods, physico-chemical such as adsorption make a rational process especially in treating stabilized leachate due to cost-effectiveness [9]. Adsorption technique is well known as

*Corresponding author: zawawi@uthm.edu.my

the efficient and promising approach in wastewater treatment processes [10]. It also has potential to remove or minimize different types of pollutants. Thus, to encounter landfill leachate that heavily contaminate, adsorption is a good choice especially when leachate consists of high refractory and non-biodegradable compound that has been a major challenge to biological treatment [8].

Composites adsorbent can be defined as natural or synthesized materials made from two or more materials with significantly different physical and chemical properties that remain separate and distinct at the microscopic or macroscopic scale within the material [11]. They represent an interesting and attractive alternative as adsorbents and/or catalysts due to their high reactivity and excellent selectivity towards specific pollutant compounds [12].

Recently, crustaceans waste such as the shell of shrimp and crab have been utilized into a product namely chitosan. Chitosan is derivative from N-deacetylation of chitin, a second most abundant naturally occurring biopolymer next to cellulose. Chitosan exhibit unique characteristics such as hydrophilicity, biocompatibility, biodegradability, non-toxicity, adsorption properties, film-forming ability, bio-adhesively, poly-functionality [13-14]. It also has a high percentage of nitrogen compared to synthetically substituted cellulose. The uniqueness of chitosan can be considered as an answer to researcher's interest in developing cost-effective and environmentally friendly technologies for the remediation of soil and water polluted with toxic trace elements [15].

Therefore, the combination between chitosan and mineral material such as zeolite and feldspar has enhanced the composite adsorbent capability and capacity. The adsorbent has potential to be utilized in treating a stabilize leachate that contains high concentration of ammonia and heavy metal. Thus, the objective of this study is to investigate the efficiency of CFZ biocomposite adsorbent for ferum (Fe) removal in leachate and optimize the Iron removal system parameter.

2.0 METHODOLOGY

2.1 Leachate Sampling

Leachate sample was collected from Simpang Renggam Landfill Site (SRLS). The sample was collected in a clean airtight HDPE ('high density polyethylene") container that had been rinsed with leachate beforehand to ensure an accurate result and reduce sample contamination container. Once the leachate samples arrived at the laboratory, the leachate was stored at 4ºC to minimize any further change that might occur in physiochemical and biological properties until the experiments analyses were carried out later. For dissolves metals, leachate was filtered immediately before adding HNO_3 until the pH became less than 2. All reagents and chemical were of analytical grade and laboratory test were carried out according to standard methods [16].

2.2 Preparation of Biocomposite Adsorbent

CFZ biocomposite produced by using 4g of chitosan and Feldspar: zeolite (FZ) composite. The chitosan was dissolved in 50 ml of 2 % (V/V) acetic acid solution first, before the powdered feldspar and zeolite added into the dissolved chitosan solution. The mixture was stirred for 3 hours at room temperature. Then, the mixture was dropped through a syringe into 500 ml alkaline precipitation bath. The beads produced were washed extensively with de-ionized water. The beads dried at 60^0c in an oven until their weight become constant.

2.3 Experimental

Initial values of ferum were determined before further experiment on ferum removal conducted. Then, batch adsorptions were performed in series of 250 ml conical flask with a varied value of a variable like wastewater pH, adsorbent dosage and contact time. The adsorbent was shaken in leachate and at the end of each experiment; a 0.45μm filter paper filtered the adsorbent and kept it in an airtight container for further analysis. The residue concentration of ferum in leachate tested by Atomic Absorption Spectrometer (AAS). This experimental step applied for adsorbent including CFZ biocomposite as well as zeolite, feldspar and chitosan as single adsorbent. Meanwhile, the removal efficiency and uptake capacity were evaluated using equation (1) and (2) respectively.

$$\mathrm{Re}\,moval\% = \left(\frac{C_o - C_f}{C_o}\right) \times 100 \qquad (1)$$

Where:
C_0 = Initial concentration (mg/L)
C_f = Final concentration (mg/L)

$$q = \left(C_o - C_f\right) \times \frac{V}{m} \qquad (2)$$

Where:
q = Uptake (mg/g)
C_0 = Initial concentration (mg/L)
C_f = Final concentration (mg/L)
V = Volume of solution (L)
m = Mass of bioadsorbent (g)

3.0 RESULTS AND DISCUSSION

3.1 Chemical Analysis of Leachate

Chemical analysis of leachate performs to find the characteristic of Simpang Renggam Landfill Site leachate. The characteristics of leachate shown in Table 1.

Table 1: Characteristic of Simpang Renggam Landfill Site Leachate

Parameter	Average	Standard B
pH	8.65	5.5-9
Temperature	24	-
Ammoniacal nitrogen (mg/L)	1808	-
COD(mg/L)	9811	100
BOD_5*	937	50
BOD_5/COD	0.095	0.5
Iron(mg/L)	15.82	5.0

3.2 Effect of pH

pH is one of the parameters affecting adsorption process. The effect of pH on adsorption can occur by the protonation of functional group, where protonation and deprotonation phenomenon occur in most acid-base reactions [17]. Figure 1 shows the effect of pH on ferum adsorption. This figure shows that the removal was occurred optimally during acidic phase but reducing when approaching alkaline phase. The maximum removal occurred at pH 5, where the maximum uptake capacity reached 0.2804 mg/g.

This situation happened due to the availability of hydrogen ion, whereby when the leachate pH is in acidic phase, more hydrogen ions were available to protonate amine groups ($-NH_2$) [18]. On the other hand, when leachate pH increased, the positive charged in leachate and adsorbent interface would decline, resulting the emerging of negatively charged on adsorbent surface. Therefore, a lower adsorption at higher pH may result by the ionic repulsion between the negatively charged surface of adsorbent and ferum ion [19].

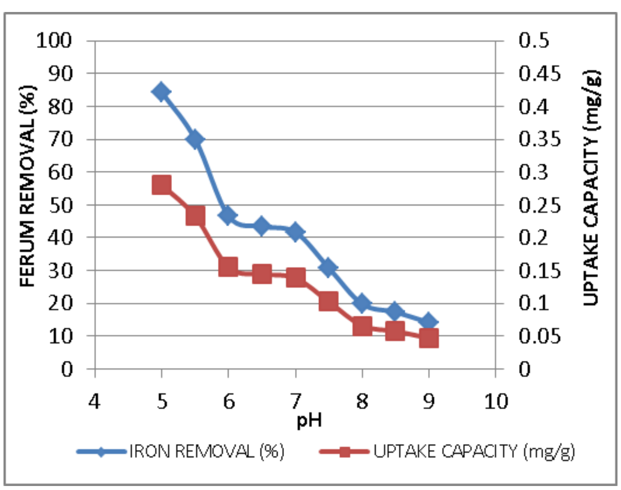

Fig. 1: The effect of pH in ferum adsorption

3.3 Effect of Biocomposite Dosage

The dosage added has a great influence on sorption process since the dose added into the solution determine the number of binding site for adsorption[20]. The effect of adsorbent dosage toward ferum adsorption shown in Figure 2. From this figure, the removal percentage of ferum is increasing when the dosage is increases. However, when the dosage reached 5g - 6g, a further dosage did not increase too much. Hence, this shows that it has reach equilibrium. This pattern happened because when the dosage is increasing, the adsorption surface area is also increasing. Hence, the availability of exchangeable site is higher [21].

Meanwhile, the uptake capacity shows a total opposite pattern, where the uptake capacity is decreasing when the dosage is increasing. This pattern occurred due to an increased pollutant adsorbed- to- adsorbent ratio and may be attributed to overlapping or aggregation of adsorption site resulting in a decrease in the total adsorbent surface area [21]. In conclusion, the maximum adsorption efficiency of ferum and its adsorption capacity is 69.89% and 0.2 mg/g respectively. Where it occurs with 6g of adsorbent dosage, which slightly higher than 5g, where removal percentage is 69.75%. Hence, the optimum dosage for ferum adsorption by CFZ biocomposite is 6g.

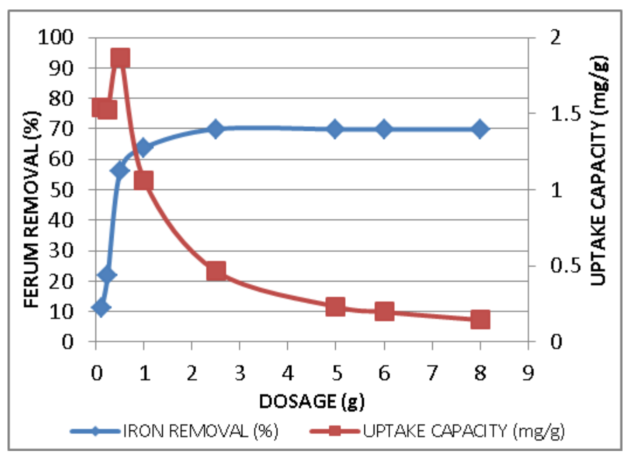

Fig. 2: The effect of adsorbent dosage in ferum adsorption

3.4 Effect of Contact Time

Figure 3 shows the effect of contact time in ferum adsorption. From the figure, the optimum contact time occured at 180 minutes with 89.03% removal efficiency and 2.22 mg/g uptake capacity. The pattern of removal showed that with the increasing of time, the removal of ferum also increasing until it reached saturated level where the reading became constant, which indicate equilibrium. This observation could be explained, as at the very beginning of the adsorption process, a large number of vacant surface sites were available for adsorption, which may be the reason for the rate of adsorption boosted in the initial stages. Then, after a lapse of some time, the remaining vacant surface sites were difficult to be occupied and beyond indicated contact time where it reached saturated level, the percentage of removal almost constant indicating the attainment of equilibrium conditions [22,28].

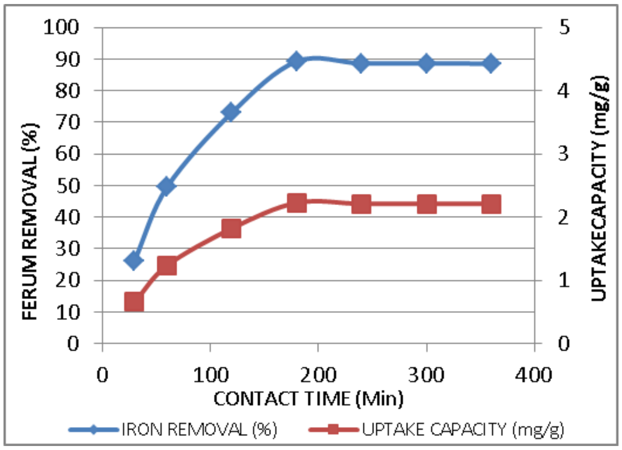

Fig. 3: The effect of contact time in ferum adsorption

3.5 Removal under optimum condition

The parameter affecting adsorptions were studied to find the optimum condition for the adsorbent to perform optimally. Optimum condition determined is pH 5, 6 gram of biocomposite dosage and 180 minutes contact time. The removal obtained for ferum is 90% with 0.0127 mg/g uptake capacity.

3.6 Comparison between Media in Biocomposite Adsorbent

Figure 4 shows the result of comparison between CFZ biocomposite adsorbent with its single media. Chitosan has shown the best removal compared to others and slightly higher than CFZ biocomposite.

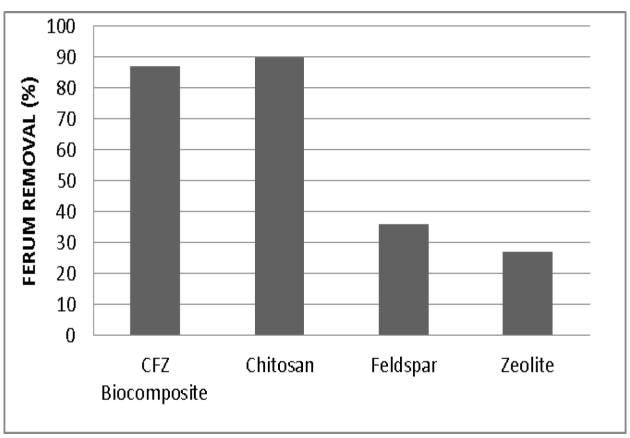

Figure 4 Comparison on ferum removal between media

Hance, it shows that the presence of chitosan is mostly the reason for CFZ biocomposite in enhancing the removal of iron. Thus, it shows that the combination is not only enhancing the adsorptive capacity, but also widen the variety of pollutant adsorbed due to each material distinctive properties.

4.0 CONCLUSION

Recently, many researchers have been focusing on cost effectiveness and environmentally friendly approach. This approach is important to gain sustainability for benefit of the future generation. The biocomposite adsorbent used to treat leachate because each material in this composite adsorbent has its special characteristic to make this adsorbent conventional to treat many types of pollutant including ferum. The result shows that optimum condition occurred at pH 5 with 6 gram of biocomposite dosage and 180 minutes contact time. The removal obtained for ferum is 90% with 0.0127 mg/g uptake capacity. Hence, it indicates that chitosan has potential to remove toxic or heavy metal pollutant and can act as a binder for the composite.

Acknowledgement

The authors acknowledge the research grant provided by the Office for Research, Innovation, commercialization and Consultancy Management (ORICC) of the Universiti Tun Hussein Onn Malaysia.

References

[1] Daud, Z., Hatta, M.Z.M., Kassim, A.S.M., Aripin, A.M., Awang, H., Analysis of Napier grass (Pennisetum purpureum) as a potential alternative fibre in paper industry. *Materials Research Innovations*, 18, (2014), pp. S6-18-S6-20.

[2] Fauziah, S. H., and Agamuthu, P. 2007. SWPlan Application for Malaysian Municipal Solid Waste Management. *Malaysian Journal of Science*. 26(1): 17-22.

[3] Wurochekke, A. A., Mohamed, R. M. S. R., Al-Gheethi, A. A. S., Noman, E. A., & Kassim, A. H. M. Phycoremediation: A Green Technology for Nutrient Removal from Greywater. In *Management of Greywater in Developing Countries*, (2019), pp. 149-162. Springer, Cham.

[4] Daud, Z., Awang, H., Mohd Kassim, A.S., Mohd Hatta, M.Z., Mohd Aripin, A., Cocoa Pod Husk and Corn Stalk: Alternative Paper Fibres Study on Chemical Characterization and Morphological

Structures. *Advanced Materials Research*, Volume 911, (2014), pp. 331-335.

[5] Mahmoud, A. S., Sanni-Anibire, M. O., Hassanain, M. A., & Ahmed, W. Key performance indicators for the evaluation of academic and research laboratory facilities. *International Journal of Building Pathology and Adaptation*, (2018).

[6] Daud, Z., Awang, H., Kassim, A.S.M., Hatta, M.Z.M. & Aripin, A.M., Comparison of pineapple leaf and cassava peel by chemical properties and morphology characterization. *Advanced Materials Research*. Volume 974, (2014), pp. 384-388.

[7] Deng, Y., and Englehardt, J. D. 2007. Electrochemical oxidation for landfill leachate treatment. *Waste Management.* 27(3): 380–388.

[8] Halim, A. A., Aziz, A. A., Johari, M. A. M., and Ariffin, K. S. 2010. Comparison study of ammonia and COD adsorption on zeolite, activated carbon and composite materials in landfill leachate treatment. *Desalination*. 262: 31-35.

[9] Habeeb, S.A., Latiff, A.A.A., Daud, Z., Ahmad, Z. The start-up of hybrid, anaerobic up-flow sludge blanket (HUASB) under a range of mesophiclic and thermophilic temperatures. *EnvironmentAsia*, Volume 4 (2), (2011), pp. 63-68.

[10] Daud, Z., Bakar, M.H.A., Rosli, M.A., Ridzuan, M.B., Aliyu, R., Application of Response Surface Methodology (RSM) to Optimize COD and ammoniacal nitrogen removal from leachate using moringa and zeolite mixtures. *International Journal of Integrated Engineering*, Volume 10 (1), (2018), pp. 142-149.

[11] Soltani, R. D. C., Safari, M., Rezaee, R., Teymouri, P., Hashemi, S. E., Ghanbari, R., & Zandsalimi, Y. Preparation of Chitosan/Bone Char Nanocomposite for Adsorption of Hexavalent Chromium in Aquatic Environments. *Arabian Journal for Science and Engineering*, (2017). Pp.1-10.

[12] Daud, Z., Abubakar, M.H., Abdul Latiff, A.A., Awang, H., Ahmad, Z., Ridzuan, M.B., Cod and ammonia removal from landfill leachate using mixed granular adsorbent media. *Jurnal Teknologi*, 80 (4), (2018), pp. 81-86.

[13] Antonyová, A., Abdullah, A. H., Nagapan, S., & Daud, Z.. Certain Building Materials with Respect to Their Thermal Properties as Well as to Their Impact to Environment. *International Journal of Integrated Engineering*, volume *10*(4), (2018).

[14] Nitayaphat, W. Utilization of Chitosan/Bamboo Charcoal Composite as Reactive Dye Adsorbent. *Chiang Mai Journal Sciences.* Volume 41(1), (2014) : 174-183.

[15] Daud, Z., Hatta, M.Z.M., Ridzuan, M.B., Awang, H., Adnan, S.,Studies on physical and mechanical properties by soda-AQ pulping of Napier grass. *Defect and Diffusion Forum*, 382 DDF, (2018), pp. 318-321.

[16] APHA .2005. Standard Methods for the Examination of Water and Wastewater. 21 ed. Washington D.C: American Public Health Association.

[17] Witek-Krowiak, A., Szafran, R. G., and Modelski, S. Biosorption of heavy metals from aqueous solutions onto peanut shell as a low-cost biosorbent. *Desalination*. Volume 265(1-3) (2011), pp. 126- 134.

[12] Wan Ngah, W. S., Teong, L. C., Toh, R. H., and Hanafiah, M. A. K. M. Utilization of chitosan–zeolite composite in the removal of Cu(II) from aqueous solution: Adsorption, desorption and fixed bed column studies. *Chemical Engineering Journal*, Volume 209(2012), pp. 46–53.

[19] Daud, Z., Aziz, H.A., Adlan, M.N., Hung, Y.-T., Application of combined filtration and coagulation for semi-aerobic leachate treatment. *International Journal of Environment and Waste Management*, Volume 4 (3-4), (2009), pp. 457-469.

[20] Zafar, M. N., Nadeem, R., and Hanif, M. A. 2007. Biosorption of nickel from protonated rice bran. *Journal of hazardous materials.* 143 (1-2): 478-485.

[21] Nadeem, R., Ansari, T. M., and Khalid, A. M. 2008. Fourier Transform Infrared Spectroscopic characterization and optimization of Pb(II) biosorption by fis(Labeo rohita) scales. *Journal Of Hazardous Materials*.156(1-3): 64-73.

[22] Srivastava, V. C., Mall, I. D., and Mishra, I. M. 2006. Equilibrium modelling of single and binary adsorption of cadmium and nickel onto bagasse fly ash. *Chemical Engineering Journal.* 117(1): 79-91.

[23] Ratha, N., Ismail, T. N. H. T., Wijeyesekera, D. C., Bakar, I., Siang, A. J. L. M., Talib, M. A., & Zainorabidin, A. Developmental research of sustainable technologies to minimise problematic road embankment settlements. In *Journal of Physics: Conference Series*, Vol. 1049, No. 1, ((2018), p. 012046). IOP Publishing

[24] Yunus, R., Abdullah, A.H., Yasin, M.N., Masrom, M.A.N., Hanipah, M.H. Examining performance of Industrialized Building System (IBS) implementation based on contractor satisfaction assessment. *ARPN Journal of Engineering and Applied Sciences*, Volume 11 (6), (2016), pp. 3776-3782.

[25] Bakar, S.K.A., Abdullah, A.H. Simulation of thermal performance in an office building. BEIAC 2012 - 2012 IEEE Business, *Engineering and Industrial Applications Colloquium*, art. no. 6226074, (2012), pp. 318-323.

[26] Balakrishnan, B., Awal, A.S.M.A., Abdullah, A.B., Hossain, M.Z. Flow properties and strength behaviour of masonry mortar incorporating high volume fly ash. *International Journal of GEOMATE*, Volume 12 (31), (2017), pp. 121-126.

[27] Al-Gheethi, A.A., Mohamed, R.M.S.R., Efaq, A.N., Norli, I., Halid, A.A., Amir, H.K., Kadir, M.O.A. Bioaugmentation process of secondary effluents for reduction of pathogens, heavy metals and antibiotics.

Journal of Water and Health, Volume 14 (5), (2016), pp. 780-795.

[28] Sohu, S., Abd Halid, A., Nagapan, S., Fattah, A., Latif, I., Ullah, K. Causative factors of cost overrun in highway projects of Sindh province of Pakistan. IOP Conference Series: Materials Science and Engineering, Volume 271 (1), (2017), art. no. 012036.