

Landfill leachate treatment by Low Cost Activated Carbon Prepared from Agriculture Waste

¹Nurshazwani Bt. Azmi, ²Alexanderrayar Singarayah, ³Mohammed J.K. Bashir*, ⁴Sumathi Sethupathi

^{1,2,3,4} *Department of Environmental Engineering, Faculty of Engineering and Green Technology (FEGT), University Tunku Abdul Rahman, 31900 Kampar, Perak, Malaysia.*

¹*shazwani103@gmail.com (N.B.Azmi)*

²*alex_rxz89@yahoo.com (A.Singarayah)*

³*jkbashir@utar.edu.my (M.J.K.Bashir)*

⁴*sumathi@utar.edu.my (S.Sethupathi)*

Abstract—Adsorption via activated carbon (AC) is one of the superior treatments for stabilized landfill leachate, but expensive and limited resource of AC precursor (bituminous and lignite) limit application of this technique in landfill leachate treatment. Based on previous studies, agriculture waste performed as an excellence potential for AC precursor. Thus, present study evaluates the sugarcane bagasse derived activated carbon (SBAC) for adsorptive removal of ammonical nitrogen, COD, and color from old anaerobic landfill leachate located in Perak, Malaysia. The chemical and physical properties of adsorbent were examined by Fourier transform infrared spectroscopy (FTIR) and scanning electron microscope (SEM). The effects of AC dosage (g) on adsorption performance were investigated in a batch mode study. Equilibrium data were favorably described by Langmuir isotherm model, with a maximum monolayer adsorption capacity for NH₃-N, COD and color at 14.62 mg/g, 126.58 mg/g and 555.56 Pt/Co, respectively. The results illustrated the potential usability of SBAC for treatment of anaerobic landfill leachate.

Index Terms: Landfill, Landfill leachate, Sugarcane Bagasse, Adsorption, Activated Carbon.

I. INTRODUCTION

Landfill is a well-known municipal solid waste (MSW) disposal method as up to 95% of MSW collected worldwide is buried in the landfill [1]. Due to such advantages such as low cost, simple disposal procedure [2] and ability to deal with high amounts of waste [3], landfill still widely used option for MSW disposal. However, the major drawback of landfilling is due to the generation of hazardous landfill leachate which can cause a serious environmental and aesthetic problem.

Landfill leachate generally define as a dark color liquid with a strong odor due to the percolation of excess water with mixture of organic and inorganic pollutant deposited within the waste layers of the landfill [4]. As the consequences, landfill leachate characterized with high concentration of pollutant includes the organic matter, BOD, COD, ammoniacal-nitrogen, heavy metal, and inorganic salt. Moreover, stabilized landfill leachate contain humid and fulvic substances that simply hard to be

treated by biological treatment. Thus, physico-chemical method such as activated carbon (AC) adsorption could be a considerable option for stabilized landfill leachate treatment. Due to its unique properties such as larger surface area, high micropore volume, rapid adsorption, low acid-base reactivity, and favor pore size distribution [5], AC becomes as one of the best filtration media in the world. However, high manufacturing cost and expensive carbonaceous material [6] limit AC adsorption method applications in stabilized landfill leachate treatment. Currently, there is a great interest in finding low-cost and effective alternative to the existing commercial activated carbon [7]. The cost production of AC from cellulose waste material is very low compared to the cost of commercial AC. Furthermore, low-cost activated carbon may contribute to environmental sustainability and offer benefits for future commercial applications [8].

Thus, the present work focusing on production of Sugarcane bagasse derive activated carbon (SBAC) for adsorptive removal of ammoniacal-nitrogen, COD and color from stabilized landfill leachate.

The structural, functional and surface chemistry of the prepared AC was evaluated. The adsorptive removals of pollutants were studied by adsorption equilibrium and isotherm model.

II Material and Methods

Landfill leachate sampling

Landfill leachate was collected from an anaerobic landfill known as Sahom landfill located at Kampar, Perak, Malaysia. The landfill is equipped with leachate collection system, However, there is no leachate treatment system prior to discharge. Leachate samples were collected from leachate collection pond and characterized according to standard Method of Water and Wastewater [9].

Preparation of Sugarcane bagasse derived activated carbon (SBAC)

Sugarcane bagasse (SB) was used as AC precursor in this study. SB was obtained from the neighboring shop. The SB was cut into small pieces, boiled and washed exhaustively in order to eliminate the impurities from the surface followed by drying at 105°C overnight in order to remove the unwanted moisture content. Dried bagasse was ground using grinding machine (ZM2000, Germany) with 0.1 mm blade and sieved to retain the particle sized ranging from 1.4 mm to 0.5 mm. The prepared bagasse was used for the char production with performed by the carbonization unit. The prepared bagasse was placed into a muffle furnace and carbonized at 700°C. The char produced was mixed with potassium hydroxide (KOH) solution with (Char: KOH) impregnation ratio at 1: 2.73 (wt%) and the wet bagasse was dried at 105 °C for three days before activated in muffle furnace for 3 hours at 600°C with a ramping rate at 10°C/min [10]. The resultant activated carbon was washed with 0.1M HCl and rinsed repeatedly with deionized water until the pH of the filtrate reach 6.5-7 for removing organic matters residue and alkalis. Finally, the prepared SBAC was dried at 105°C for 24 hours prior to leachate treatment process.

Characterization of SBAC

Textural morphology of SBAC and chemical characterization of surface functional group was carried out by Scanning Electron Microscope (FESEM-JEOL 6701-F) and Fourier Transform Infrared spectrometer (Perkin-Elmer Spectrum RXI).

Chemical characterization of surface functional groups was detected using the pressed potassium bromide (KBr) pellets which containing 5% of carbon sample. The FTIR spectra were recorded between 4000-400 cm⁻¹.

Batch Study

This study was concentrated on the identification of the optimum operational conditions. The experiments were conducted in a series of 250 ml Erlenmeyer flask containing mixture of 100 ml raw leachate and SBAC with agitation speed of 200 rpm and contact time of 180 min. After each run, the media were filtered and the filtrates were kept for adsorptive uptake analysis of color, COD and NH₃-N. The color concentration was measured at 455 nm wavelength using Hach color method 8025, whereas COD concentration was measured using HR+ COD vials by DR 6000 spectrophotometer, while NH₃-N was measured with spectrophotometer DR6000 at wavelength 640nm. All tests were conducted in accordance with the standard methods for examination of water and wastewater [9].

Equilibrium study

The performance of the experiment was studied using adsorption isotherm which describes the relationship between the concentrations of adsorbate accumulated on the adsorbent and the concentration of the dissolved adsorbate at equilibrium [11]. Amounts of adsorbate accumulate on the adsorbent were measured by the difference between the initial concentration of adsorbate with the concentration of adsorbate at equilibrium within the dissolve solution where it is expressed as following equation:

$$q_e = \frac{(C_o - C_e)V}{m} \quad (1)$$

where q_e is amount of adsorption at equilibrium, C_o is initial concentration for the adsorbate while C_e is the amount of adsorbate at equilibrium. V (L) is the volume of the solution and m (g) is the mass of the dry sorbent used.

The pollutant removal percentage is calculated by the following equation:

$$Removal (\%) = \frac{(C_o - C_e)}{C_o} \times 100 \quad (2)$$

where C_o and C_e are the initial and equilibrium stage liquid-phase concentration of adsorbate, in term of color, COD and NH_3-N .

III. RESULT AND DISCUSSION

Leachate Characteristic

Table 1 shows leachate characteristic of Sahom landfill located in Kampar, Perak, Malaysia. The leachate has high concentration of color, COD, and NH_3-N , with lower value of BOD_5 : COD ratio (< 0.1). Thus Sahom landfill can be categorized as stabilized landfill leachate [12]. As stabilized landfill leachate contains refractory organic compound [13], effectiveness of biological process decreases and physico-chemical processes in particular AC adsorption may become one of the appropriate options.

Table 1: Sahom Landfill Characteristics

Parameters	Unit	Average Value
Temperature	°C	26.9-27.1
pH	-	8.60-8.75
Conductivity	ms	10.93-11.02
Resistivity	Ω	90.04-90.08
Turbidity	ntu	105.6-126.0
Color	Pt/Co	3300-3500
COD	mg/L	1490-1570
NH_3-N	mg/L	1860-1950
BOD_5	mg/L	106-120
BOD_5/COD	-	0.071-0.076
Total Suspended Solids	mg/L	203-227

SBAC Characterization

FTIR Analysis

The FT-IR analyses of SBAC before and after treatment were illustrated in Figure 1. The spectra of adsorbents were plotted to determine the vibration frequency changes in the functional group of the adsorbent. The absorption peaks around 3426 cm^{-1} indicates the free and intermolecular bonded hydroxyl groups [14]. The peak around 1562 cm^{-1} may be attributes to aromatic group of lignin compound. The peak observed at 1084 cm^{-1} can be assigned to C-O band, due to OCH_3 group also confirm the presence of lignin structure of SBAC [15]. Meanwhile, peak at 1384 cm^{-1} may involve C-H deformation. After adsorption treatment, it was found that oxygen containing $-OH$ groups are affected after uptake process. This is judged from shifts of its position to the lower frequency ($3426-3423 \text{ cm}^{-1}$). Other remarkable shift included the C-O band from 1084 to 1090 cm^{-1} . The results indicated that the participation of these groups via oxygen for pollutant binding in leachate to SBAC in agreement with Person principal for hard-soft acids and bases [16].

SEM Analysis

Scanning electron microscope (SEM) analysis of SBAC was presented in Figure 2. Base on the figure, it showed the development of micropore structure on the SBAC. As the non-carbon elements such as hydrogen, oxygen and nitrogen released in the form of tars and gases during pyrolysis process, a rigid carbon skeleton with a rudimentary pore structure known as char formed from the aromatic compound [17]. Pretreatment of the char with dehydrating agent (KOH) inhibit formation of tar and other undesired product [17]. Consequently, CO_2 creates activated carbon with larger micropore volume and narrower micropore size distribution [17] that bring to higher adsorption capacity of the pollutant.

Experimental performance

Adsorbent dosage

The effect of AC dosage on percentage removal of color, COD and NH_3-N was illustrated on Figure 3. The shaking speed (200 rpm) and contact time (180 minutes) were remained constant with varied AC dosages (0-9g) throughout the experiment. Based on Figure 3, it is apparent that adsorptive removal

of color, COD and $\text{NH}_3\text{-N}$ increased considerably by increasing adsorbent dosage from 0g/100ml to 2g/100ml. However, further increase in adsorbent dosage up to 9g/100ml showed steadily increased of pollutant uptake. According to Moodley et al. (2011), further increase of adsorbent dosage lead to overlapping of surface site due to the overcrowding of adsorbent particles [18]. As the result, it will bring to the decrease of accessible surface area of adsorbent, thus lowering the pollutant removal per

unit adsorbent. Besides, adsorbent dosage presents a profound effect on the adsorption process due to the reason that it predicts the cost of adsorbent per unit of pollutant to be treated [19]. Thus, the optimum SBAC dosage for pollutant removal is 7g/100ml with percentage removal of 94.7 % color, 83.6% COD and 46.6% $\text{NH}_3\text{-N}$.

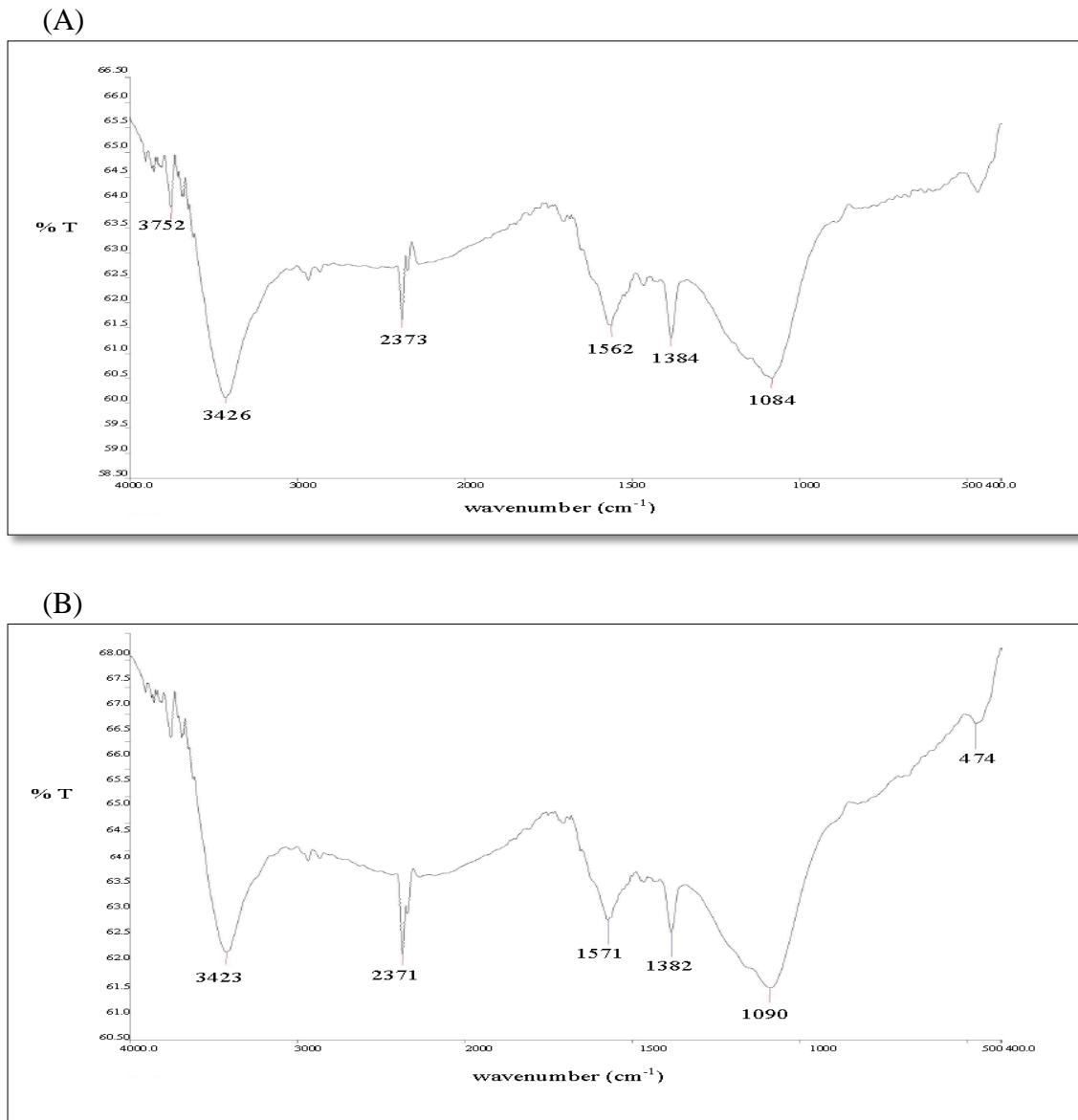


Figure 1: FTIR analysis of (A) SBAC before treatment and (B) SCAC after treatment.

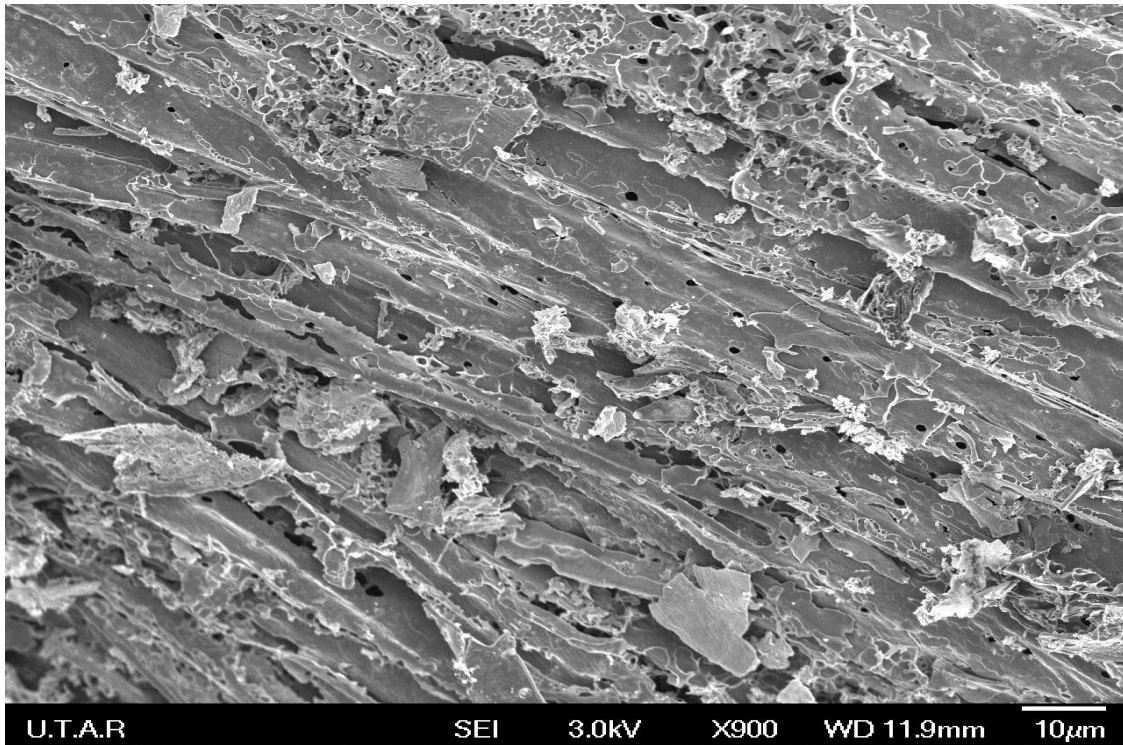


Figure 2: SEM micrograph of sugarcane bagasse derive activated carbon at 900X magnification

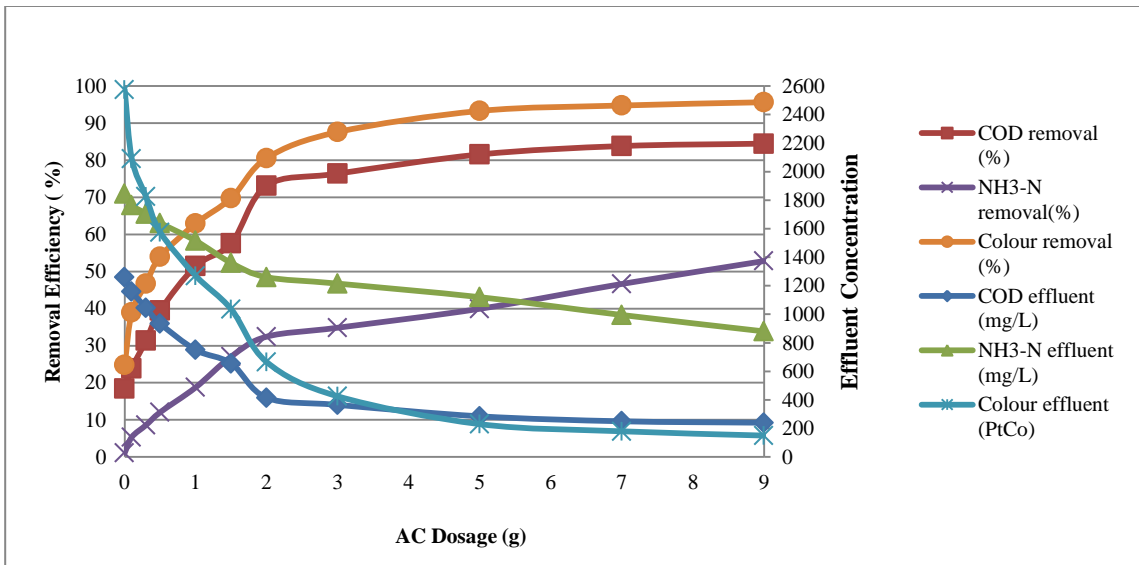


Figure 3: Removal efficiency in terms of COD, NH3-N and colour vs activated carbon dosage

Equilibrium Study

Adsorption properties of SBAC was studied using Langmuir and Freundlich Isotherms, which are the most common models for describing the adsorption characteristic of adsorbents used in water and wastewater treatment [20]. Langmuir isotherm describes the monolayer adsorption of adsorbate on specific homogenous site of adsorbent meanwhile, Freundlich isotherm theory assumes multilayer coverage of adsorbate over a heterogenous adsorbent surface. Equation of both isotherm were expressed as below:

Langmuir isotherm:

$$\frac{1}{q_e} = \frac{1}{QbC_e} + \frac{1}{Q} \quad (3)$$

Freundlich isotherm:

$$\log q_e = \log K + \frac{1}{n} \log C_e \quad (4)$$

where C_e is the equilibrium liquid-phase concentration and q_e is the equilibrium uptake capacity (mg/g), while Q (mg/g), b (L/mg), $1/n$ and K are the constant.

Based on Table 2, the adsorption of colour, COD and $\text{NH}_3\text{-N}$ was rationally explained by Langmuir and Freundlich isotherm. The R^2 value of Langmuir isotherm model for colour, COD and $\text{NH}_3\text{-N}$ were 0.9818, 0.9653 and 0.9728 while for Freundlich isotherm model were at 0.9524, 0.9085 and 0.923. As Langmuir model yielded relatively high R^2 value and close to unity, thus the adsorption of pollutant on SBAC took places as monolayer adsorption, with the maximum adsorption capacity of 555.56 Pt/Co, 126.58 mg/L and 14.62 mg/L for color, COD and $\text{NH}_3\text{-N}$ removal.

Table 2: Isotherm Equation Parameters for colour, COD and $\text{NH}_3\text{-N}$ Adsorption onto Activated Carbon

Parameter	Langmuir isotherm coefficient			Freundlich isotherm coefficient		
	Q mg/g	b (L/mg)	R^2	K mg/g (L/mg) ^{1/n}	1/n	R^2
Colour	555.556	0.0005224	0.9818	0.678734617	0.8199	0.9524
COD	126.582	0.0005521	0.9653	0.002761849	1.5929	0.9085
$\text{NH}_3\text{-N}$	14.6199	0.0004753	0.9728	0.000000037	2.847	0.923

(IV) RECOMMENDATION

The present study determines the optimum treatment for stabilized landfill leachate in terms of adsorbent dosage as well as the best fitted isothermal models for color, COD and $\text{NH}_3\text{-N}$ removal.

There are much more research gaps which are yet to be explored in terms of landfill leachate treatment. Following are suggestions on research area for future studies.

- Comparison on treatment efficiency using various types of experimental conditions such as the shaking speed, contact time and pH of the adsorbate.
- Removal of different types of pollutant such as heavy metal (manganese, zinc, chromium, lead, copper and cadmium), organic and inorganic compound.
- Activated carbon derived from other source of agricultural waste such as from fruit peel, fruit seed and cellulosic waste material.

(V) CONCLUSION

The potential of Sugarcane Bagasse derived activated carbon (SBAC) for adsorptive removal of colour, COD and NH₃-N collected from a stabilized landfill leachate was examined. Based on the batch adsorption study, the optimum percentage removals of color (94.71), COD (83.61) and NH₃-N (46.65) were obtained at optimum adsorbent dosage of 7g/100ml of leachate. The adsorptive removal was well-fitted with Langmuir isotherm model with the maximum adsorption capacity of color, COD and NH₃-N was at 555.56 Pt/Co, 126.58 mg/L and 14.62 mg/L, respectively

(IV) REFERENCES

- [1] T.A. Kurniawan, W.H. Lo, G.Y.S. Chan, “Degradation of recalcitrant compounds from stabilized landfill leachate using a combination of ozone-GAC adsorption treatment,” *Journal of Hazardous Material*, vol.137, no.1, pp. 443-455, Sept. 2006.
- [2] M.J.K. Bashir, H.A.Aziz, M.S. Yusoff, M.N. Adlan, “ Application of response surface methodology(RSM) for optimization of ammoniacal nitrogen removal from semi-aerobic landfill leachate using ion exchange resin,” *Desalination*, vol. 254, no. 1-3, pp.154-161, May. 2010.
- [3] U.N. Ngoc, and H.Schintzer, “Sustainable solutions for solid waste management in east Asian countries,” *Waste management*, vol.2, no.9, pp. 1982-1995, 2009.
- [4] Yao, P, “Perspectives on technology for landfill leachate treatment,” *Arabian Journal of Chemistry*, doi: 10.1016/j.arabjc.2013.09.031, Sept,2013.
- [5] H.S. Li, S.Q. Zhou, Y.B. Sun, P. Feng, J.D. Li, “Advanced treatment of landfill leachate by a new combination process in a full-scale plant,” *Journal of Hazardous Material*, vol. 172, no.1, pp. 408-415, Dec. 2009.
- [6] D. Mohan, C.U.J. Pitman, “Activated carbon and low cost adsorbent remediation of tri- and hexalent chromium from water,” *Journal of Hazardous Material*, vol.137, no.2 .pp. 762-811, Sept.2006.
- [7] Z.A.AlOthman, M.A. Habila, R.Ali, M.S. Eldin Hassouna, “ Valorization of two waste streams into activated carbon and studying its adsorption kinetics equilibrium isotherms and thermodynamics for methylene blue removal,”*Arabian Journal of Chemistry*, vol.2, pp. 2-12, 2013.
- [8] M.A. Aseel, N.A. Abbas, F.A. Ayad, “ Kinetics and equilibrium study for adsorption of textiles dyes on coconut shell activation. *Arabian Chemistry Journal*, DOI:10.1016/j.arabjc.2014.01.02
- [9] APHA, “Standard Methods for the Examination of Water and Wastewater, 18th ed., American Public Health Association, Washington United States, 2005.
- [10] K.Y. Foo, B.H. Hameed, “Microwave-assisted preparation and adsorption performance of activated carbon from biodiesel industry solid residue: Influence of operational parameters,” *Bioresource Technology*, vol.103, no.1, pp.398–404, January, 2012.
- [11] R. Droste, “ Theory and practice of water and wastewater treatment,” John Wiley and Sons, Inc., USA, 1997.
- [12] H. Alvarez-Vazquez, B. Jefferson, and S. J. Judd, “Membrane bioreactors vs conventional biological treatment of landfill leachate: a brief review,” *Journal of Chemical Technology and Biotechnology*, vol. 79, no. 10, pp. 1043–1049, 2004.
- [13] HUO Shoulian, XI Beidou, YU Haichan, HE Liansheng, FAN Shilei, and Liu Hongliang, “Characteristic of dissolved organic matter (DOM) in leachate with different landfill ages,” *Journal of Environmental Sciences*, vol.20, pp. 492-498, Sept. 2007.
- [14] X.Colom, F. Carillo, F.Nagues, P. Garriga, “Structural analysis of photodegraded wood by means of FTIR spectroscopy,” *Polymer Degradation and Stability*, vol.80, pp.543-549, 2003.
- [15] U.Garg, M.P. Kaur, G.K. Jawa, D. Sud, V.K.

- Garg, “ Removal of cadmium (II) from aqueous solutions by adsorption on agriculture waste biomass,” *Journal of Hazardous Material*, vol. 154, pp. 1149-1157, 2008.
- [16] R.G. Pearson, “Hard and soft acids and bases,” *Journal of the American Chemical Society*, vol. 85, pp. 3533-3539, 2010.
- [17] A.R. Mohamed, M. Mohammadi, G.N. Darzi, “ Preparation of carbon molecular sieve from lignocellulosic biomass: A review,” *Renewable and Sustainable Energy Reviews*, vol. 14, pp.1591-1599, 2010.
- [18] K. Moodley, R.Singh, E.T.Musapatika, M.S. Onyango, A. Ochieng, “ Removal of nickel from wastewater using an agricultural adsorbent,” *Water SA*, vol. 37, pp. 41-46, 2011.
- [19] S. Kushawa, B. Sreedhar, P.P. Sudhakar, “A spectroscopic study for understanding the speciation of Cr on palm shell based adsorbents and their application for the remediation of chrome plating effluents,” *Bioresourouce Technology*, vol.116, pp.15–23, 2010.
- [20] T.L. Eberhardt, S.H. Min, “Biosorbents prepared from wood particles treated with anionic polymer and iron salt: effect of particle size on phosphate adsorption,” *Bioresource Technology*, vol.99, pp. 626-630, 2008.