

RESEARCH ON ACTIVATED CARBON PREPARATION FROM MACADAMIA SHELLS USING KOH ACTIVATING AGENT APPLIED FOR METHYLENE BLUE TREATMENT

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ARTICLE INFO

Article history: Received May. 8.2018, Accepted Dec. 29.2018.

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Abstract

Research on bio-activated carbon wastewater treatment material prepared from Macadamia shells using KOH activating agent at the rate of 1KOH: 1 piece of activated carbon is carried out through influencing factors of temperature and time. The research result of Methylene blue (MB) absorption ability at respectively optimal temperature and time of 200°C and 60 minutes of activated carbon shows that 1g of activated carbon will absorb 91,50mg of Methylene Blue. The result shows that the color treating efficiency of the Methylene Blue (MB) is 83,41% corresponding to the color level reduced from 349.67 Pt-Co to 58 Pt-Co. Research result shows that bioactive carbon prepared from Macadamia shells and activated from KOH can treat color in dyeing textile wastewater.

Keywords: activated carbon, Macadamia shells, Methylene Blue color absorption

INTRODUCTION

Activated carbon is known to be a highly absorbent material and is used in many fields including water treatment (Samorn Hirunpraditkoon et al., 2011; Tzong-Horng, 2009). The absorption ability of activated carbon is influenced by many factors such as structural characteristics, surface functional group (Yan-Juan et al., 2014), surface area, ash content,... (Kwagher & Ibrahim, 2013). In fact, activated carbon is made from two main sources of coal and agricultural residues including coconut shell charcoal (Koby, 2004), rice husk (Awwad et al., 2010) and bamboo charcoal (Wang et al., 2008).

In Vietnam, in recent years, Macadamia have been grown in the North West and Central Highlands. It is estimated that by 2020, the area used for planting Macadamia will reach 10,000 hectares (MARD., 2015). With annual harvesting output, about 70% - 77% tons of shells is released. Most of shell is considered as waste, only a few is used as fuel (Caturla et al., 1991; Martins et al., 2015).

However, according to the study by Toles (1998), it shows that Macadamia shells contain many attractive features to make activated carbon such as carbon content (47-49%) is higher than the carbon content in bamboo (45.53%) (Daud & Ali, 2004) and is equivalent to carbon content in coconut shells 48.63% (Daud & Ali, 2004). Moreover, the shell also contains oxygen content of 46.52%, hydrogen content of 6.10%, nitrogen content of 0.36% and relatively low ash content of only 0.22% (Toles et al., 1998), this shows that Macadamia is potential to become activated carbon thanks to the mentioned characteristics above.

In this study, bioactive carbon is made from Macadamia shells by chemical method using KOH agent to activate. In addition, bioactive carbon is also tested for absorbability of Methylene Blue in dyeing textile wastewater.

RESEARCH METHODS

Research means

- Research object: Methylene Blue ($C_{16}H_{18}ClN_3S \cdot 3H_2O$, 99%, China) with a concentration of 25mg/l (corresponding to 349.67 Pt-Co as defined in TCVN (Vietnamese Standard) 6185: 2005).
- Research Chemicals: $Na_2HPO_4 \cdot 12H_2O$ (China, 98%), KH_2PO_4 (China, 98%), KOH (China, 96%), HCl 1N (China).
- Research material: Macadamia shells harvested in Lam Dong province.
- Research device: Jatest.

Experimental layout

Experiment 1: Preparation of bioactive carbon from Macadamia shells

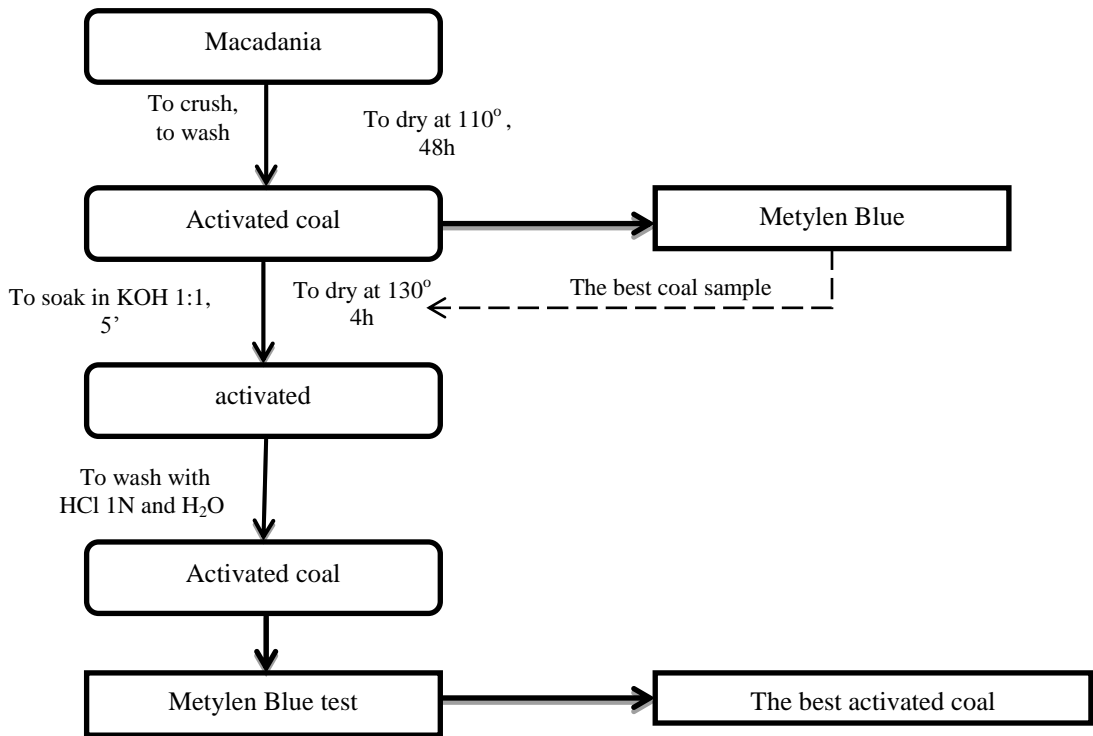


Figure 1. Experiment layout of activated carbon preparation

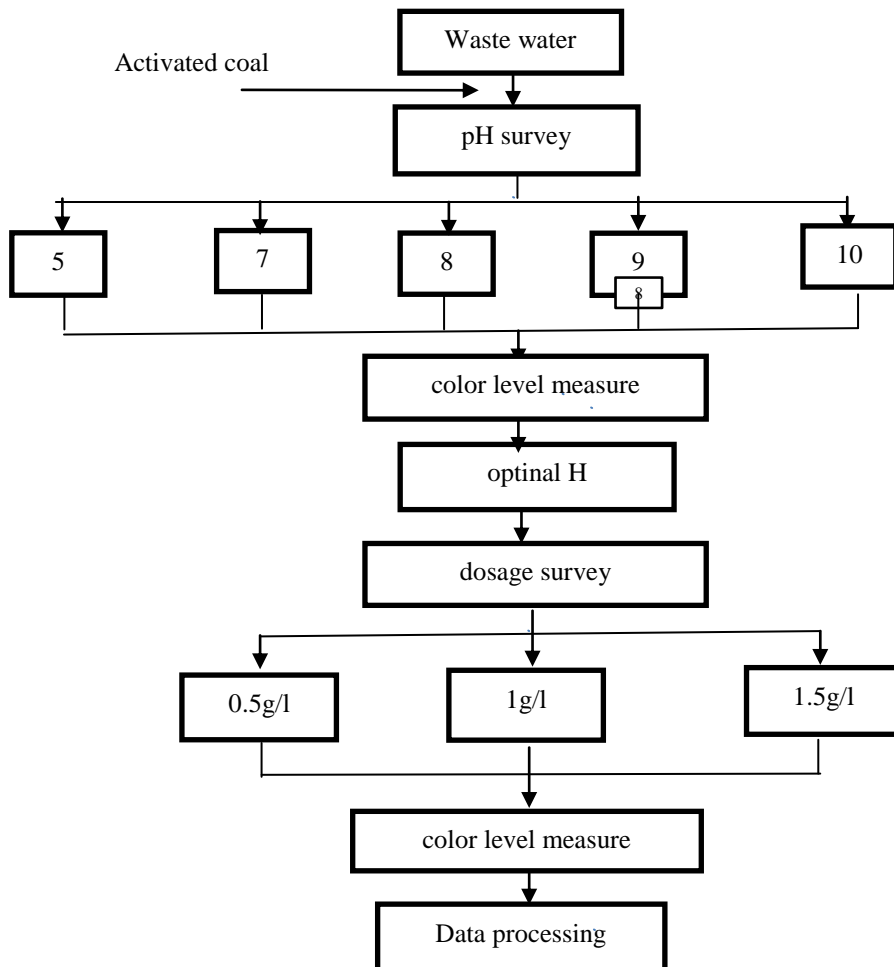
Experiment 2: Survey of MB's color treatment ability

Figure 2. Experiment layout of Methylene blue treatment with activated carbon

Evaluation methods

- Determination of pH is measured directly using pH Meter Toledo (2017).
- Determination of color in accordance with TCVN 6185: 2005.
- Determination of the surface observation by Scanning Electron Microscope (SEM).
- Determination of functional groups in molecules by FT-IR method (Fourier Transformation Infrared Spectrometer).
- Determination of Methylene Blue Absorption Index under standard of GB/T 12496.10 - 1999.

RESULTS AND DISCUSSION**Results of preparation of activated carbon from Macadamia shell**

The results from Fig. 3.1 show that in the temperature range increasing from 150 to 450°C and burning time in 60 minutes, the maximum absorption of MB reached its maximum at 200°C and burning time in 60 minutes with 91,18 mg MB/g coal.

Survey of proper temperature affecting the activation process

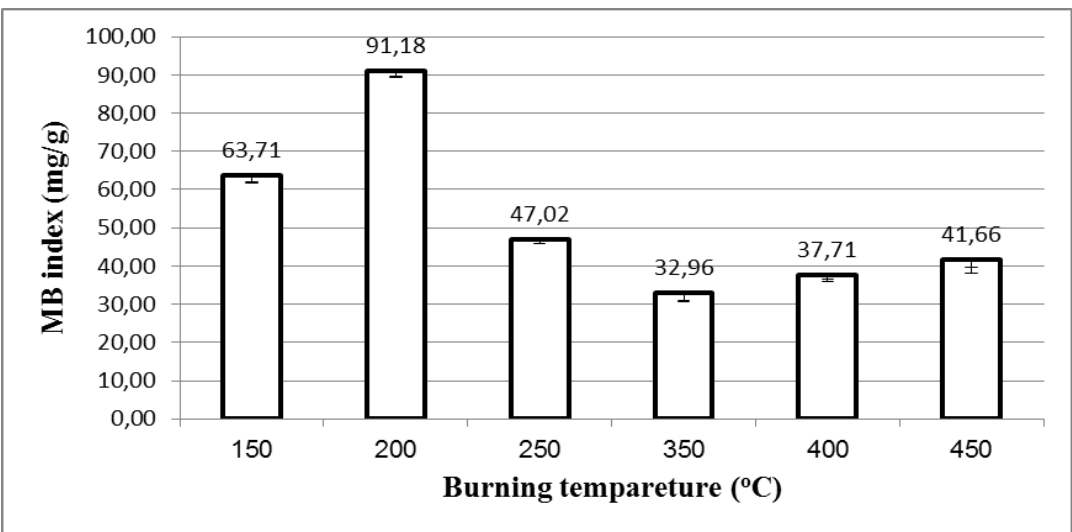


Figure 3. Optimal temperature determination results according to Methylene Blue Absorption

The results of this study are consistent with previous studies such as Hameed and Ahmad (2009). The study used garlic shell to absorb MB color effectively with 82.64 mg/g or the research result using tea leaves of Uddin et al., (2009) had similar absorption rates of 85.16 mg/g.

In addition, the research results of MB adsorption capacity of Macadamia activated coal are better than those of research result of Vadivelan and Kumar(2005) on rice husk at 40.59 mg/g; The results of Annadurai et al., (2002) on MB absorption ability of orange peel reaches 18.6 mg/g or the results of Janos et al., (2003) on fly ash reaches 75.52mg/g.

Accordingly, the research result on determining the optimal temperature found at 200°C is the premise for further survey of the best coal-fired heating time for activation process of activated carbon.

Survey of reaction time affecting the activation process

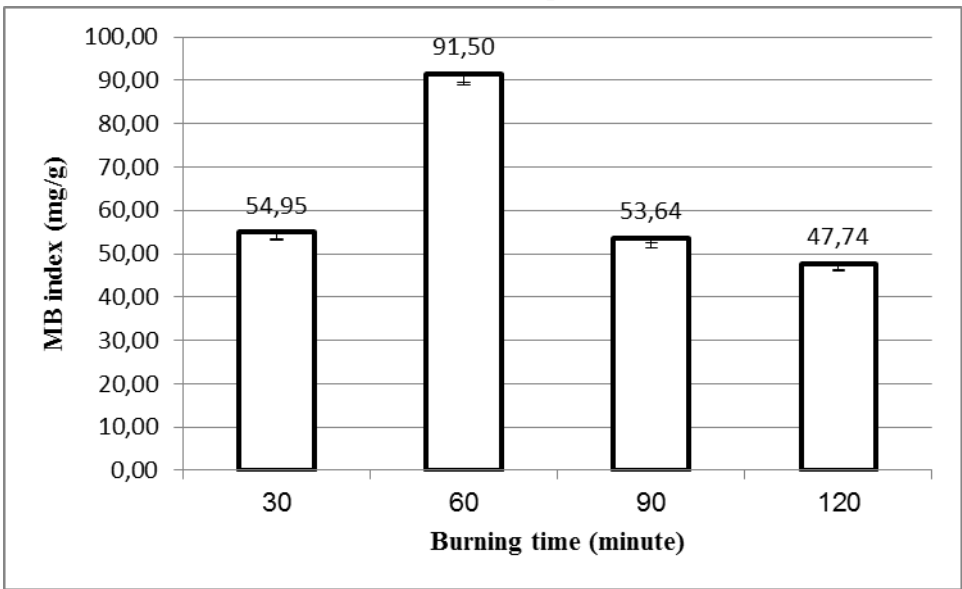


Figure 4. The result showing the optimal burning time according to MB absorption

The research results from Fig. 3.2 surveying the burning time in the range of 30, 60, 90, 120 minutes at optimal temperature of 200°C, showing a gradual decrease in absorption from 54,95 mg/g (at 30 minutes) to 91,5 mg/g (at 60 minutes) and then decreases to 47,74mg/g (at 120 minutes). So the results show that the temperature at 200°C and the burning time in 60 minutes is the best result.

Compared with previous studies such as the research result by San Miguel et al., (2002), using activated carbon prepared from rubber waste to eliminate MB from the aqueous solution and the absorption ability of this activated carbon reported to be 49 mg/g; Research results from Kavitha and Namasivayam (2007) show the successful research on MB absorbability of quartz and the absorption level reaching 5.87 mg/g or based on the results of Han et al., (2006) Reports on cereal grains have a maximum absorption of 26.3 mg/g and in 2007 Han et al., (2007) successfully surveyed the absorbability of phoenix leaves with absorption level up to 89.7 mg/g; According to a study by Dogan et al., (2008) Hazelnut shell's color elimination ability reaches 38.22 mg/g. It shows that the research results on activated carbon with KOH has absorbability of MB high than the above researches.

The research result of activated carbon activated from Macadamia shells shows the absorbability of MB at 91,5mg /g at 200°C and in 60 minutes.

Survey result of activated carbon treatment ability on Methylene Blue color

Survey of pH suitable for treatment process

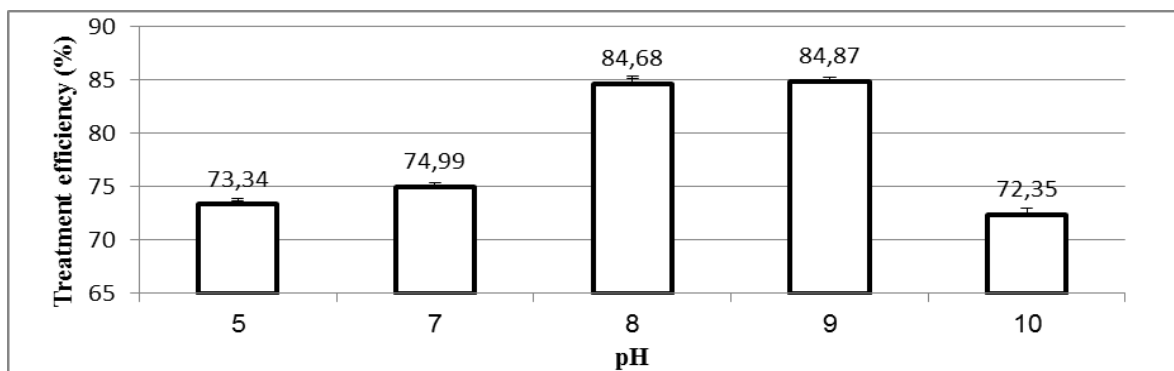


Figure 5. Results determine the effect of pH on MB's color treatment efficiency

The research results of MB's color treatment ability of the material as shown in Figure 3.3 show that with a pH ranging from 5, 7, 8, 9, 10, the processing efficiency was quite high at 73.34% The 74.99%; 84.68%; 84.87% and 72.35% respectively. Thereby, at pH=9 value, this is the pH range of highest treatment efficiency.

According to the research result report of Ghaedi et al., (2012) and the research result of Gao et al., (2005) explained the effect of pH on MB absorbability on activated carbon material when pH in the solution is adjusted to low level, the coal's color treatment ability is based on the protonation of the coal's functional groups and through electrostatic repulsion, MB is readily eliminated from the solution. When the solution reaches high pH, the coal surface will be negative charged, so they rely on electrostatic attraction and hydrogen bonding to eliminate color.

The research result is more likely to be treated better than previous studies, such as the research result of activated carbon made from sawdust of Garg et al., (2004) which is shown that at pH = 8, the color elimination efficiency of sawdust is only 74% and the research result of Han et al.,

(2006) on the absorbability of the cereal shells for solution containing MB, at the same range of pH, the treatment efficiency of the cereal shell is 84%.

Activated carbon studied in preparation from Macadamia shell has the best MB treatment ability at pH = 9 with the following dosage range.

Survey of the appropriate dosage of activated carbon for treatment process

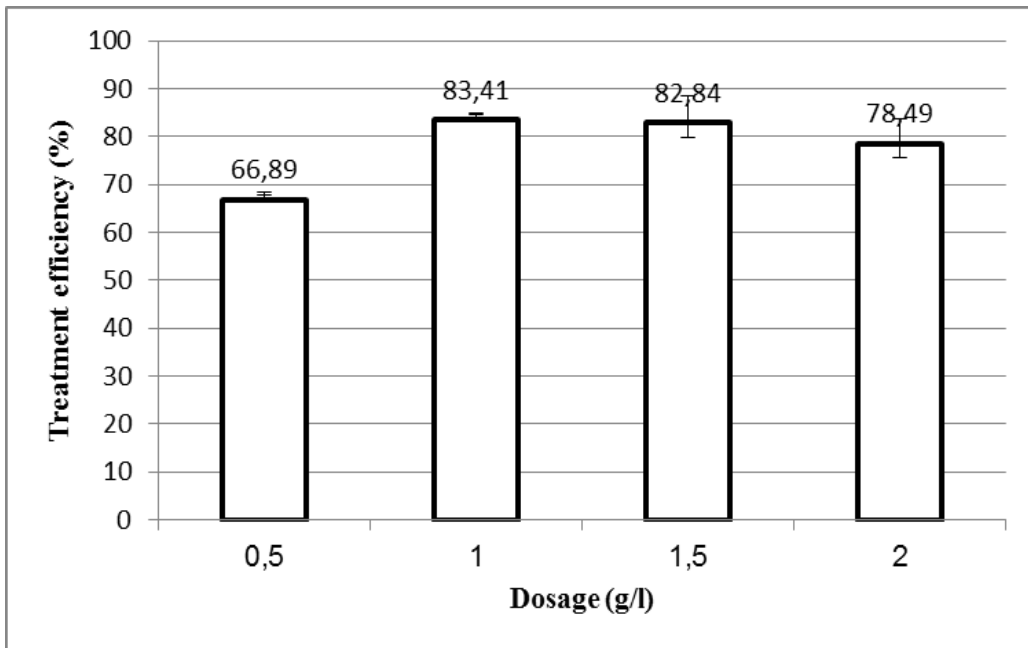


Figure 6. Survey diagram of dosage impact on MB color treatment efficiency

The survey results of the dosage in Figure 3.4 show that the dosage of 1g/l is the most suitable dosage for color treatment with 84,37%, higher than other dosages. According to the research by Ghaedi et al., (2012), it is shown that the pore size and activated carbon dosage are two factors that significantly influence MB absorbability. By increasing the absorbing surface area, the absorbability will increase significantly.

Compared to previous studies such as Garg et al., (2004), it is shown that after 30 minutes of treatment, MB treatment efficiency of activated carbon from sawdust was only 35.8% when used with H_2SO_4 and only 22.8% when used with agent Formaldehyde. This proves that the activated carbon studied in this article is better able to handle than some previous studies.

This shows that activated carbon in the research prepared from Macadamia shells is capable of treating MB in dyeing textile wastewater.

CONCLUSION

Research results show that bioactive carbon material in the research which is successfully prepared from agricultural waste, Macadamia shells by chemical methods using KOH as activator at 200°C for 60 minutes. The absorbance of MB up to 91.5 mg/g can be demonstrated by activated carbon capable of dyeing. The results showed that the two factors influenced the efficiency at pH=9 and the coal appropriate dosage of 1g/L reaches 83.41%.

REFERENCES

- Annadurai, G., R.S. JuangD.J. Lee (2002). Use of cellulose-based wastes for adsorption of dyes from aqueous solutions. *Journal of hazardous materials*, 92(3), 263-274.
- Awwad, N. S., H.M.H. Gad, M.I. AhmadH.F. Aly (2010). Sorption of lanthanum and erbium from aqueous solution by activated carbon prepared from rice husk. *Colloids and Surfaces B: Biointerfaces*, 81(2), 593-599.
- Ministry of Agriculture and Rural Development (MARD) (2015). Macaques - Status and Development Directions, ed.
- Caturla, F., M. Molina-SabioF. Rodriguez-Reinoso (1991). Preparation of activated carbon bu chemical activation with ZnCl₂. *Great Britain*, 29(7), 999 - 1007.
- Daud, W. M. A. W.W.S.W. Ali (2004). Comparison on pore development of activated carbon produced from palm shell and coconut shell. *Bioresource Technology*, 93(1), 63-69.
- Doğan, M., H. AbakM. Alkan (2008). Biosorption of methylene blue from aqueous solutions by hazelnut shells: equilibrium, parameters and isotherms. *Water, air, and soil pollution*, 192(1-4), 141–153.
- Gao, B. Y., Q.Y. Yue, Y. WangZhou W. Z., (2005). Color removal from dye containing wastewater by magnesium chloride. *Journal of Environmental Management*, 82, 167–172.
- Garg, V. K., M. Amita, R. KumarR. Gupta (2004). Basic dye (methylene blue) removal from simulated wastewater by adsorption using Indian Rosewood sawdust: a timber industry waste. *Dyes and pigments*, 63(3), 243–250.
- Ghaedi, M., S.H. Heidarpour, S.N. Kokhdan, R. Sahraie, A. DaneshfarB. Brazesh (2012). Comparison of silver and palladium nanoparticles loaded on activated carbon for efficient removal of Methylene blue: Kinetic and isotherm study of removal process. *Powder Technology*, 228, 18–25.
- Hameed, B. H.A.A. Ahmad (2009). Batch adsorption of methylene blue from aqueous solution by garlic peel, an agricultural waste biomass. *Journal of hazardous materials*, 164(2), 870–875.
- Han, R., W. Zou, W. Yu, S. Cheng, Y. WangJ. Shi (2007). Biosorption of methylene blue from aqueous solution by fallen phoenix tree's leaves. *Journal of Hazardous Materials*, 141(1), 156–162.
- Han, R., Y. Wang, P. Han, J. Shi, J. YangY. Lu (2006). Removal of methylene blue from aqueous solution by chaff in batch mode. *Journal of Hazardous Materials*, 137(1), 550–557.
- Janos, P., H. BuchtovaM. Rýznarová, 2003. Sorption of dyes from aqueous solutions onto fly ash. *Water research*, 37(20), 4938–4944.
- Kavitha, D.C. Namasivayam (2007). Experimental and kinetic studies on methylene blue adsorption by coir pith carbon. *Bioresource Technology*, 98(1), 14–21.
- Koby, M., (2004). Removal of Cr (VI) from aqueous solutions by adsorption onto hazelnut shell activated carbon: kinetic and equilibrium studies. *Bioresource technology*, 91(3), 317–321.
- Kwaghger, A.J.S. Ibrahim (2013). Optimization of conditions for the preparation of activated carbon from mango nuts using HCl. *American Journal of Engineering Research*, 2(7), 74–85.
- Martins, A. C., O. Pezoti, A.L. Cazetta, K.C. Bedin, D.A.S Yamazaki, G.F.G. Bandoch, T. Asefa, J.V. VisentainerV.C. Almeida (2015). Removal of tetracycline by KOH-activated carbon

- produced from macadamia nut shells: kinetic and equilibrium studies. *Chemical Engineering Journal*, 260, 291–299.
- Samorn Hirunpraditkoon., T. Nathaporn, R. AnotaiN. Kamchai (2011). Adsorption capacities of activated carbons prepared from Bamboo by KOH Activation. *International Journal of Chemical*, 5(6), 447–481.
- San Miguel, G., G.D. FowlerC.J. Sollars (2002). Adsorption of organic compounds from solution by activated carbons produced from waste tyre rubber. *Separation science and technology*, 37(3), 663–676.
- Toles, C. A., W.E. MarshallM.M. Johns (1998). Phosphoric acid activation of nutshells for metals and organic remediation: process optimization. *Journal of Chemical Technology and Biotechnology*, 72(3), 255–263.
- Tzong-Horng L.W S.-J. W., (2009). Characteristics of microporous/mesoporous carbons prepared from rice husk under base- and acid-treated conditions. *Journal of Hazardous Materials*, 171, 693–703.
- Uddin, M. T., Md. A. Islam, S. MahmudMd. Rukanuzzaman (2009). Adsorptive removal of methylene blue by tea waste. *Journal of Hazardous Materials*, 164(1), 53-60.
- Vadivelan, V.K.V. Kumar (2005). Equilibrium, kinetics, mechanism, and process design for the sorption of methylene blue onto rice husk. *Journal of colloid and interface science*, 286(1), 90–100.
- Wang, S. Y., M.H. Tsai, S.F. LoM.J. Tsai (2008). Effects of manufacturing conditions on the adsorption capacity of heavy metal ions by Makino bamboo charcoal. *Bioresource Technology*, 99(15), 7027–7033.
- Yan-Juan Z., X. Zhen-Jiao, D. Zheng-Kang, L. MengW. Yin (2014). Effects of steam activation on the pore structure and surface chemistry of activated carbon derive from bamboo waste. *Applied Surface Science*.