RESEARCH ON ACTIVATED CARBON PREPARATION FROM MACADAMIA SHELLS USING H_3PO_4 ACTIVATING AGENT APPLIED FOR METHYLENE BLUE TREATMENT

Pham Thi Ngoc Tram[1], Nguyen Hoang[1], Nguyen Duc Dat Duc[3], Nguyen Thanh Quang[1], Dao Minh Trung[1], Nguyen Vo Chau Ngan[4], Nguyen xuan Du[2]

[1]Thu Dau Mot Unniversity, [2]Sai Gon University

[3]Ho Chi Minh City University of Food Industry; [4]Can Tho University

---

ARTICLE INFO

***Article history**: Received Oct.1.2018, Accepted Dec. 31.2018*

Contact: *trungdm@tdmu.edu.vn*

Abstract

Research on bio-activated carbon wastewater treatment material prepared from Macadamia shells using H_3PO_4 activating agent at the rate of $2H_3PO_4$: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 500^oC and 60 minutes of activated carbon shows that 1g of activated carbon will absorb 116,91mg of Methylene Blue. The result shows that the color treating efficiency of the Methylene Blue (MB) is 97.11% corresponding to the color level reduced from 349.67 Pt-Co to 10,11 Pt-Co. Research result shows that bioactive carbon prepared from Macadamia shells and activated from H_3PO_4 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 (Hirunpradikoon et al., 2011; Tzong-Horng & S-J., 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,... (Kwaghger & Ibrahim, 2013). In fact, activated carbon is made from two main sources of coal and agricultural residues including coconut shell charcoal (Kobya, 2004), rice husk (Awwad et al., 2010) and bamboo charcoal (Wang, Tsai, Lo & Tsai, 2008).

In Vietnam, in recent years, Macadamia shells 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, Marshall & Johns, 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 H_3PO_4 agent to activate. In addition, bioactive carbon is also tested for absorbability of Methylene Blue in dyeing textile wastewater.

EXPERIMENT

Material

- Research object: Methylene Blue ($C_{16}H_{18}ClN_3S.3H_2O$, 99%, China) with a concentration of 25mg/l (corresponding to 349.67 Pt-Co as defined in TCVN (Vietnamese Standard) 6185: 2005).
- Research Chemicals: $Na_2HPO_4.12H_2O$ (China, 98%), KH_2PO_4 (China, 98%), H_3PO_4 (China, 96%). HCl 1N (China).
- Research material: Macadamia shells harvested in Lam Dong province.
- Research device: Jatest.

Experimental method

Experiment 1: *Preparation of bioactive carbon from Macadamia shells*

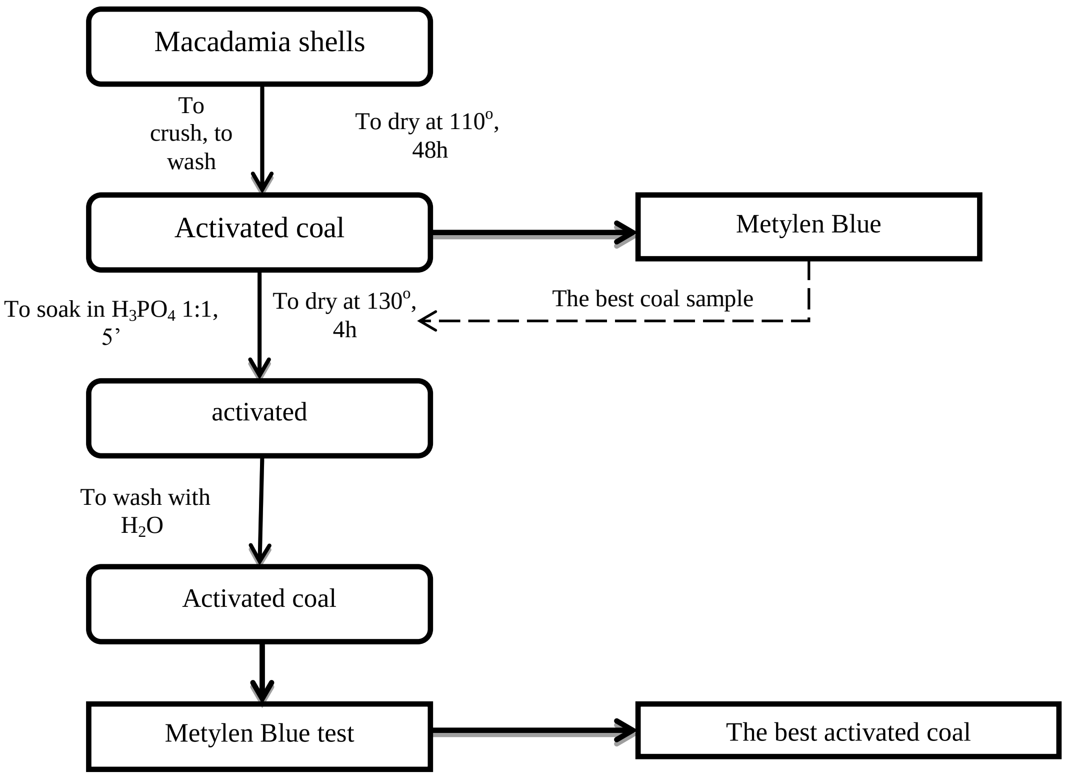

Figure 1. *Experiment layout of activated carbon preparation*

Experiment 2: *Survey of Methylene blue color treatment ability*

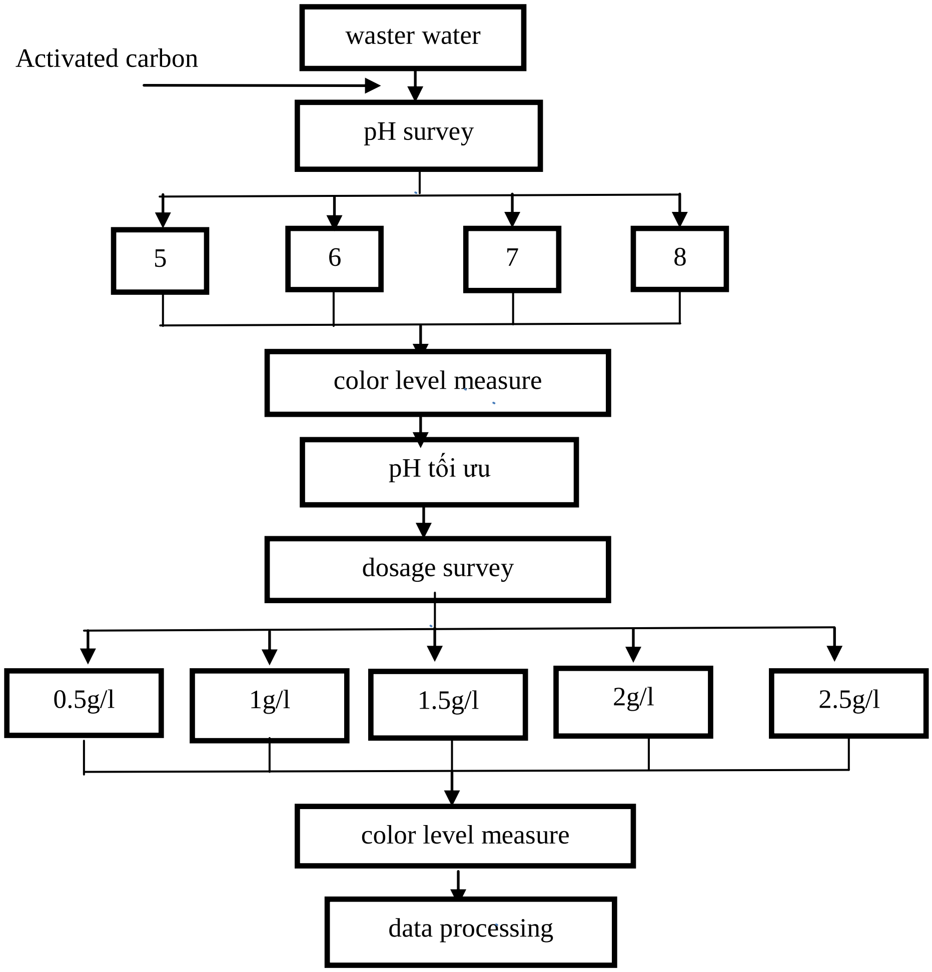

Figure 2. *Experiment layout of Methylene blue treatment with activated carbon*

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

Survey of proper temperature affecting the activation process

The results from Fig. 3.1 show that in the temperature range increasing from 350 to 550^{o}C and burning time in 90 minutes, the maximum absorption of MB reached its maximum at 500^{o}C with 101,63 mg MB/g coal.

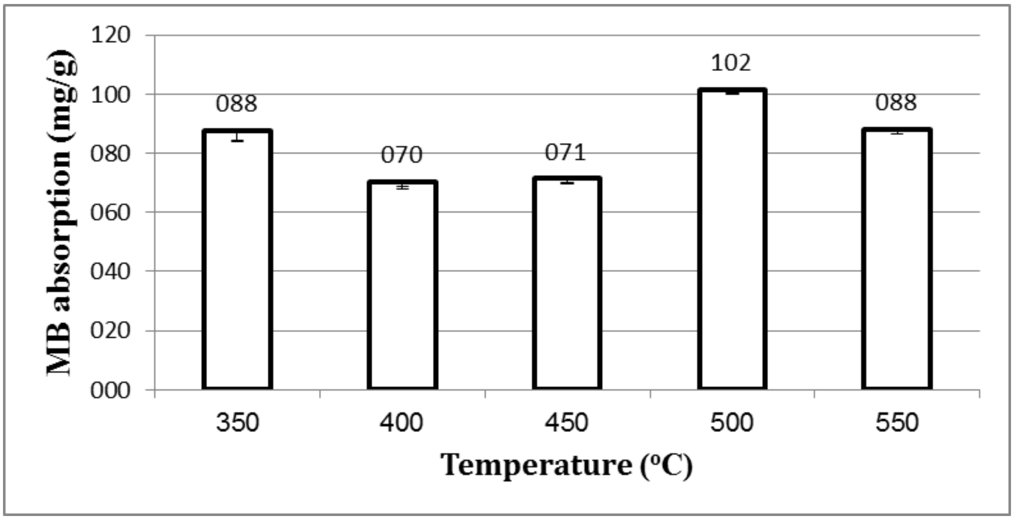

Figure 3. *Optimal temperature determination results according to Methylene Blue Absorption*

The research result shows that the research result on activated carbon H_3PO_4 agent has higher absorption level than some previous studies such as Hameed and Ahmad (2009). Garlic for MB color absorption of 82.64 mg/g or the research result using tea leaves of Uddin et al. (2009) reaching an absorption level of 85.16 mg/g, Moreover, research result of Vadivelan and Kumar (2005) on rice husk reaching 40.59 mg/g and the result of Annadurai et al. (2002) on MB absorption ability of orange peel reaching 18.6 mg/g or the research result of Janos et al. (2003) on fly ash reaching 75.52 mg/g. Accordingly, the research result on determining the optimal temperature found at 500°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

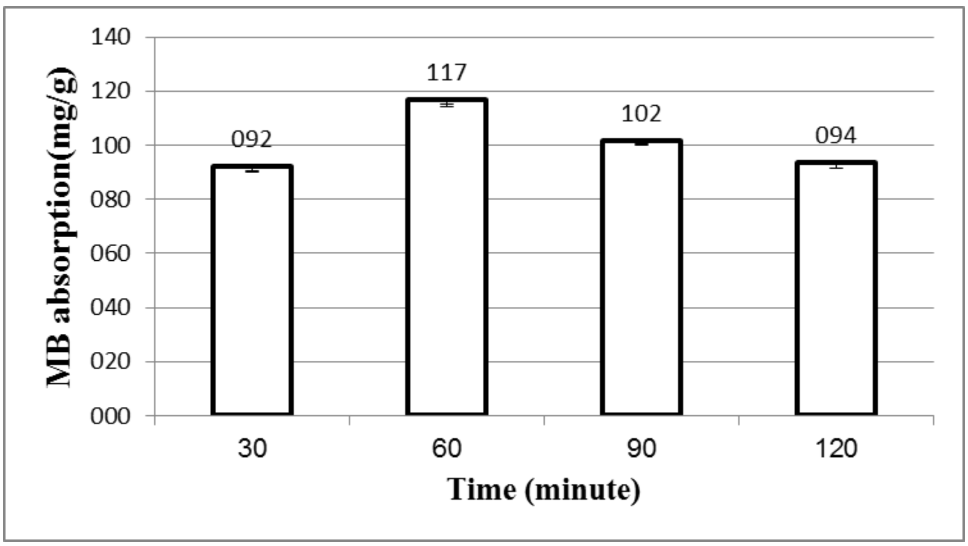

Figure 4. *The result showing the optimal burning time according to MB absorption*

The research results from Fig. 3.2 surveying the burning time in the range of 30, 60, 90, 120 minutes at optimal temperature of 500°C, showing a gradual increase in absorption from 92,23 mg/g (at 30 minutes) to 116,91 mg/g (at 60 minutes); and continued to decrease to 100.69 mg/g at

90 minutes and 93.68 mg/g at 120 minutes. So the results show that the temperature at 300°C and the heating time in 60 minutes is the best result.

Compared with previous studies such as the research result by 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. (2006) 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 H_3PO_4 has absorbability of MB high than above researches.

Therefore, the activated coal prepared from the Macadamia shell studied in this paper is capable of removing MB color in textile wastewater.

Survey result of activated carbon treatment ability on Methylene Blue color

Survey of pH suitable for treatment process

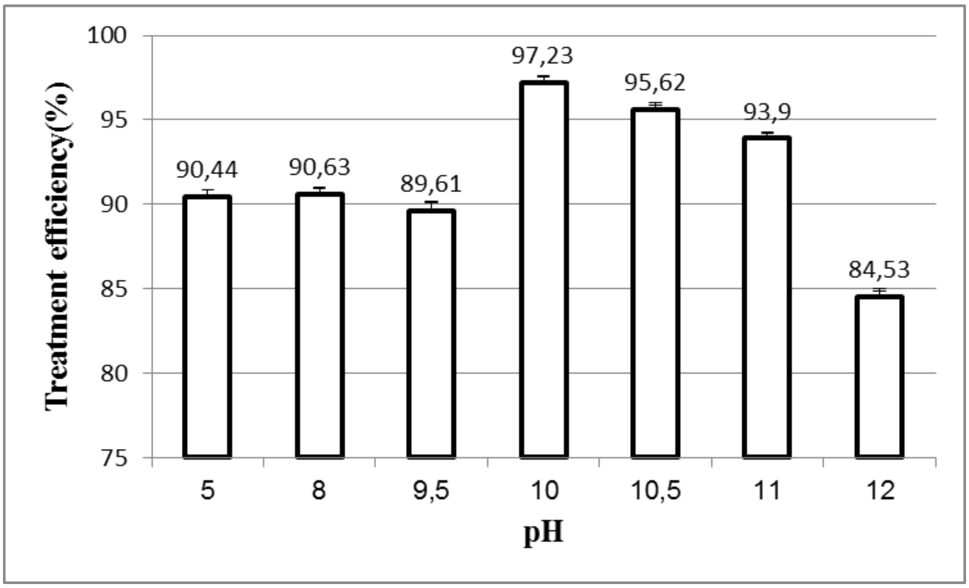

Figure 5. *Results determine the effect of pH on MB color treatment efficiency*

The research results of MB's color treatment ability of the material as shown in Figure 3.3 show that with pH range from 5 to 12, the treatment efficiency reaches its lowest (84,53%) at pH = 12 and reaches its highest (97.23%) at pH = 10. Thereby, at pH =10, this is pH range that reaches the 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's color treatment ability at pH = 10 with the following dosage range.

Survey of the appropriate dosage of activated carbon for treatment process

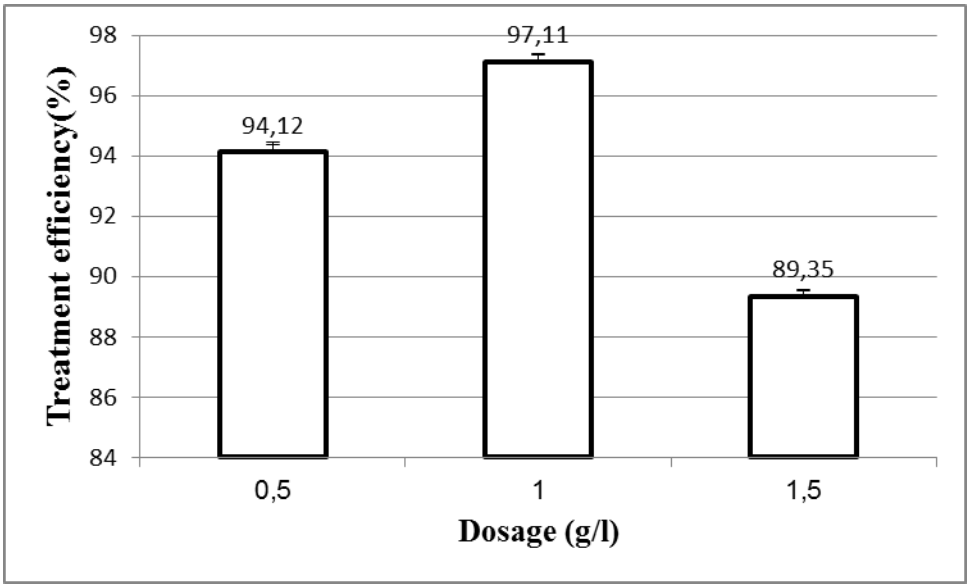

Figure 6. *Survey diagram of dosage impact on MB's color treatment efficiency*

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 97.11%, 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 is only 35.8% when being used with H_2SO_4 activating agent and it only reaches 22.8% when being used with Formaldehyde agent. This proves that the treatment ability of activated carbon studied in this article is better than some previous studies.

This shows that activated carbon in the research prepared from Macadamia shells is capable of MB's color treating 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 H_3PO_4 as activator at 300°C for 90 minutes. Adsorption of MB up to 116.91 mg / g can be demonstrated by activated carbon capable of treating dye. The results indicate that two factors affect performance at pH=10 and the mass of suitable coal 1g/l can be treated with efficiency up to 97.11%.

REFERENCES

A. C. Martins, O. Pezoti, A.L. Cazetta, K.C. Bedin, D.A.S Yamazaki, G.F.G. Bandoch, et al., (2015). Removal of tetracycline by H_3PO_4-activated carbon produced from macadamia nut shells: kinetic and equilibrium studies. *Chemical Engineering Journal, 260*, 291–299.

A. Kwaghger and J.S. Ibrahim (2013). Optimization of conditions for the preparation of activated carbon from mango nuts using HCl. *American Journal of Engineering Research, 2*, 74–85.

B. H. Hameed and A.A. Ahmad (2009). Batch adsorption of methylene blue from aqueous solution by garlic peel, an agricultural waste biomass. *Journal of hazardous materials, 164*, 870–875.

B. Y. Gao, Q.Y. Yue, Y. Wang, and W. Z. Zhou (2005). Color removal from dye containing wastewater by magnesium chloride. *Journal of Environmental Management, 82*, 167–172.

C. A. Toles, W.E. Marshall, and M.M. Johns (1998). Phosphoric acid activation of nutshells for metals and organic remediation: process optimization. *Journal of Chemical Technology and Biotechnology, 72*, 255–263.

D. Kavitha and C. Namasivayam (2007). Experimental and kinetic studies on methylene blue adsorption by coir pith carbon. *Bioresource Technology, 98*, 14–21.

F. Caturla, M. Molina-Sabio, and F. Rodriguez-Reinoso (1991). Preparation of activated carbon bu chemical activation with ZnCl2. *Great Britain, 29*, 999–1007.

G. Annadurai, R.S. Juang, and D.J. Lee (2002). Use of cellulose-based wastes for adsorption of dyes from aqueous solutions. *Journal of hazardous materials, 92*, 263–274.

G. San Miguel, G.D. Fowler, and C.J. Sollars (2002). Adsorption of organic compounds from solution by activated carbons produced from waste tyre rubber. *Separation science and technology, 37*, 663–676.

M. Doğan, H. Abak, and M. Alkan (2008). Biosorption of methylene blue from aqueous solutions by hazelnut shells: equilibrium, parameters and isotherms. *Water, air, and soil pollution, 19*2, 141–153.

M. Ghaedi, S.H. Heidarpour, S.N. Kokhdan, R. Sahraie, A. Daneshfar, and B. 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.

M. Kobya (2004). Removal of Cr (VI) from aqueous solutions by adsorption onto hazelnut shell activated carbon: kinetic and equilibrium studies. *Bioresource technology, 91*, 317–321.

M. T. Uddin, Md. A. Islam, S. Mahmud, and Md. Rukanuzzaman (2009). Adsorptive removal of methylene blue by tea waste. *Journal of Hazardous Materials,164*, 53–60.

Ministry of Agriculture and Rural Development (MARD) (2015). Macaques - status quo and development orientation, ed.

N. S. Awwad, H.M.H. Gad, M.I. Ahmad, and H.F. Aly (2010). Sorption of lanthanum and erbium from aqueous solution by activated carbon prepared from rice husk. *Colloids and Surfaces B: Biointerfaces, 81*, 593–599.

P. Janos, H. Buchtova, and M. Rýznarová (2003). Sorption of dyes from aqueous solutions onto fly ash. *Water research, 37*, 4938–4944.

R. Han, W. Zou, W. Yu, S. Cheng, Y. Wang, and J. Shi (2007). Biosorption of methylene blue from aqueous solution by fallen phoenix tree's leaves. *Journal of Hazardous Materials*, *141*, 156–162.

R. Han, Y. Wang, P. Han, J. Shi, J. Yang, and Y. Lu (2006). Removal of methylene blue from aqueous solution by chaff in batch mode. *Journal of Hazardous Materials, 137*, 550–557.

S. Y. Wang, M.H. Tsai, S.F. Lo, and M.J. Tsai (2008). Effects of manufacturing conditions on the adsorption capacity of heavy metal ions by Makino bamboo charcoal. *Bioresource Technology, 99*, 7027-7033.

Samorn Hirunpraditkoon., T. Nathaporn, R. Anotai, and N. Kamchai (2011). Adsorption capacities of activated carbons prepared from Bamboo by KOH Activation. *International Journal of Chemical*, *5*, 447–481.

Tzong-Horng L. and S.-J. W. W (2009). Characteristics of microporous/mesoporous carbons prepared from rice husk under base- and acid-treated conditions. *Journal of Hazardous Materials*, *171*, 693–703.

V. K. Garg, M. Amita, R. Kumar, and R. Gupta (Dec. 2004). Basic dye (methylene blue) removal from simulated wastewater by adsorption. *Arabian Journal of Chemistry*, *63*(3), 243–250.

V. Vadivelan and 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,* 90–100.

W. M. A. W. Daud and W.S.W. Ali (2004). Comparison on pore development of activated carbon produced from palm shell and coconut shell. *Bioresource Technology*, *93*, 63–69.

Yan-Juan Z., X. Zhen-Jiao, D. Zheng-Kang, L. Meng, and W. Yin (2014). Effects of steam activation on the pore structure and surface chemistry of activated carbon derive from bamboo waste. *Applied Surface Science, 315*, 279–286.