Poll Res. 41 (2) : 735-738 (2022)

ISSN 0257–8050
DOI No.: http://doi.org/10.53550/PR.2022.v41i02.047

PROCESS OPTIMIZATION AND SYNTHESIS OF ACTIVATED CARBON FROM COCONUT SHELL USING PHOSPHORIC ACID

D. PRAVEEN KUMAR[1], D. RAMESH[1*], P. SUBRAMANIAN[1], S. KARTHIKEYAN[1], AND A. SURENDRAKUMAR[2]

[1]Department of Renewable Energy Engineering,
[2]Department of Farm Machinery and Power Engineering,
Agricultural Engineering College and Research Institute, Tamil Nadu Agricultural University, Coimbatore 641 003, T.N., India

(Received 19 February, 2022; Accepted 30 April, 2022)

ABSTRACT

Activated carbon was produced from coconut shell through chemical activation using phosphoric acid. The optimization of process parameters namely activation temperature (400-700 °C), time (30-120 min) and impregnation ratio (0.5-3:1) was carried out using L16 Taguchi design with iodine number as a response variable. Sixteen experiments were performed at the conditions given by the Taguchi design and their corresponding iodine number was estimated. The optimized conditions for phosphoric acid activation were found to 600 °C, 60 min and 2:1 impregnation ratio with the iodine number of 1230 mg g^{-1}.Activation temperature was the most influential factor on pore characteristics of activated carbon followed by impregnation ratio and time. The study inferred that phosphoric acid can be an effective activating agent for the production of porous carbons.

KEY WORDS : Activated carbon, Chemical activation, Phosphoric acid, Taguchi, Iodine number

INTRODUCTION

In recent days activated carbon has emanated as an effective porous material that can be widely employed for varied applications owing to its improved surface area and porous structure. Activated carbon can be used as an adsorbent, electrodes, catalyst, catalyst support and gas storage (Moralý *et al.,* 2018). Synthesis of activated carbon can be done either through physical or chemical activation. Physical activation is carried out in two stages namely carbonization and activation. In physical activation the precursor material will be initially carbonized and the obtained biochar undergoes partial oxidation using either steam or oxygen or carbon dioxide (CO_2) as an activating agent. Whereas in chemical activation, the precursor material will be impregnated with chemical dehydrating agents such as zinc chloride ($ZnCl_2$), potassium hydroxide (KOH), potassium carbonate (K_2CO_3), phosphoric acid (H_3PO_4), sodium hydroxide (NaOH) followed by activation at 400-900 °C based on the type of dehydrating agent used (Nedjai *et al.,* 2021 and Yagmur *et al.,* 2018). These dehydrating agents facilitates dehydration and degradation reactions leading to the development of porosity. Chemical activation shows following advantages over physical activation mainly lower activation temperature, higher carbon yield and better porous structure (Ogungbenro *et al.,* 2020). Among many other activating agents, H_3PO_4 will be less corrosive, requires lower activation temperature and provides better surface area and pore volume (Yang *et al.,* 2020). Mbarki *et al.* (2022) studied the chemical activation of corn stigmata fibres using $ZnCl_2$, KOH and $H_3PO_4$and concluded that H_3PO_4 activation resulted in higher surface area and yield of activated carbon than other two activating agents. Tuli *et al.,* (2020) synthesized activated carbon from tea waste using $ZnCl_2$, KOH and H_3PO_4as activating agent. Their result concluded that activation with H_3PO_4had higher surface area (850.58 m^2 g^{-1}). The

three important process parameters that influence the pore characteristics of the activated carbon are impregnation ratio, activation temperature and time. The optimization of these parameters plays a significant role in chemical activation process (Moralý *et al.,* 2018). Taguchi based Design of Experiment (DOE) is one of the commonly employed tool used for process optimization studies (Karmakar *et al.,* 2018).

Commercially activated carbon was produced from fossil resources namely coal, petroleum pitch, lignite and coke (Charola *et al.,* 2018). To decrease the dependency on fossil sources, the synthesis of activated carbon from renewable sources had become more prominent. In this regard, agricultural residues are the more feasible and effective precursor for activated carbon production due to its availability and low cost. One such abundant renewable resource is the coconut shell. In light of the above, the present study aims to optimize the process parameters such as impregnation ratio, activation temperature and time for H_3PO_4 activation with coconut shell as a precursor material using Taguchi design of optimization.

MATERIALS AND METHODS

Sample preparation

Coconut shell was collected from the Coimbatore regions of Tamil Nadu. The collected coconut shell was washed with distilled water to remove any impurities and sun dried. It was then size reduced to 1-2 mm for better access of chemicals during impregnation and better heat transfer while activation. The size reduced feedstock was stored in ait tight package and further utilized for chemical activation.

Taguchi experimental design

In this study, L16 Taguchi experimental design was used for the process optimization. Taguchi method employs orthogonal array which provide detailed information on the effect of control factors on a response variable. To identify the best level of each factor, Taguchi utilizes response table. The response table contains Signal to Noise (S/N) ratio, delta and rank. Among the three algorithms of S/N ratio (nominal the better, smaller the better and larger the better), larger the better algorithm was selected, since our aim was to synthesize activated with better pore characteristics. Delta gives the difference between highest and lowest average response. Based on the delta values, ranks had been assigned to predict the most influential factor on response variable.

The control factors taken in this study are impregnation ratio, activation temperature and activation time. The corresponding levels were 0.5, 1, 2 and 3 :1 for impregnation ratio, 400, 500, 600 and 700 °C activation temperature and 30, 60, 90 and 120 min activation time. Iodine number (mg g^{-1}) was taken as a response variable and it was estimated using standard ASTM procedure(ASTM, 2006).

Activated carbon production

The dried and size reduced coconut leaflet biomass was treated by impregnating with phosphoric acid. The impregnation was performed by mixing the coconut shell with H_3POat varied impregnation ratios (0.5:1- 3:1).The impregnation ratio is the ratio of weight of the chemical to the weight of biomass taken. To enhance the uniform and proper access of chemicals into the material, the mixture was agitated using magnetic stirrer at room temperature and kept for overnight. The impregnated mixture was dried at 105 ± 5°C for 24 h in hot air oven. The dried mixture was further activated at varied activation temperatures (400- 700 °C) and time (30 - 90 min) at a heating rate of 10 °C min^{-1} in the N_2 environment with a gas flow of 150 mL min^{-1}. The activated carbon sample was washed several times with hot distilled water followed by cold distilled until pH became neutral so as to remove any left-over chemicals from the activation process. Finally, the product was dried at 105 ± 5°C for 24 h.The produced activated carbon was analysed for its iodine number.

RESULTS AND DISCUSSION

Sixteen experiments were carried out with varying process conditions as given by the Taguchi method. The corresponding iodine number of activated carbon was calculated and used as a response variable to study the influence of process conditions on pore characteristics. The reaction of phosphoric acid with biomass begins once the chemical and the biomass come into contact with each other. Phosphoric acid diffuses into the raw material, causing swelling of the biomass followed by depolymerization reactions (Gao *et al.,* 2020). Initially, the dissolution of glycosidic linkages of hemicellulose and aryl ether bonds of lignin occurs

due to their amorphous structure. Since, the crystalline structure of cellulose is resistant to acid hydrolysis, fractionation of cellulose occurs later. Due to these dehydration and depolymerization reactions, volatiles will be evolved leading to the formation of a porous structure (Kumar and Jena, 2016). Furthermore, phosphoric acid during thermal treatment produces phosphate esters, which after washing leaves an accessible porous structure (Yagmur *et al.*, 2018).

Effect of activation temperature on iodine number

The iodine number of the activated carbon increased from 797.25 to 114.75 mg g^{-1} with increase in activation temperature from 400 to 600 °C (Fig. 1). Increase in activation temperature favoured the depolymerization and partial gasification reaction that aid in the release of volatiles thus enhancing the porous structure (Nahil and Williams, 2012). With further increase in temperature from 600 to 700 °C, the iodine number gets reduced to 965.5 mg g^{-1}. This might be attributed to the carbon shrinkage and conversion of micropores to macropores at higher temperature (Haghbin and Niknam Shahrak, 2021).

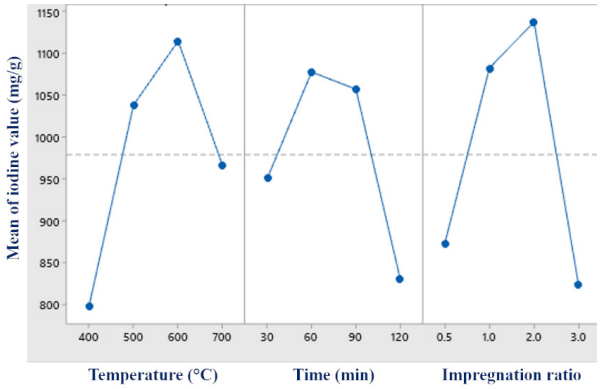

Fig. 1. Effect of temperature, time and impregnation ratio on iodine number

Effect of activation time on iodine number

Increase in activation time from 30 to 60 min resulted in increased iodine number from 950.5 to 1077.75 mg g^{-1} (Fig. 1.). However, with increase in activation time from 60 to 90 min, the iodine number was reduced to 1057.25 mg $g^{-1}$and further reduction in iodine number (830.25 mg g^{-1}) was observed at 120 min. The reduction in iodine number might be due to the destruction of porous structure at higher residence time (Haghbin and Niknam Shahrak, 2021).

Effect of impregnation ratio on iodine number

As the impregnation ratio was increased from 0.5 to 2, the iodine number gets increased from 872.75 to 1137.75 mg g^{-1}. The increase in iodine number is attributed to the increase in contact area between coconut shell and acid that enhances more diffusion of acid into the coconut shell matrix during impregnation. This results in increased porosity development. When the impregnation ratio was further increased to 3, the iodine number gets reduced to 823 mg g^{-1}. At higher impregnation ratios, the microporous structure gets collapsed and converted to macropores resulting in decreased iodine number. In addition, increased amount of acid forms an insulating layer around the biomass matrix thus preventing thepenetration of acid into the matrix and the blockage of pores due to the prevalence of more phosphate esters in the matrix even after washing (Haghbin and Niknam Shahrak, 2021; Nahil and Williams, 2012).

Response table for S/N ratio

The optimum activation levels for H_3PO_4 activation are 600 °C, 60 min and 2:1 impregnation ratio (Table 1.). The iodine number estimated at this optimized condition was 1230 mg g^{-1}. The delta was calculated as the difference between the highest and lowest mean values. Ranks were assigned based on the delta values. Higher delta value was observed for activation temperature and hence rank 1 was assigned to activation temperature. Therefor the most influential factor on iodine number was the activation temperature followed by impregnation ratio and time.

Table 1. Response table for S/N ratio

Level	Temperature (°C)	Time (min)	Impregnation ratio
1	57.58	59.28	58.60
2	60.31	60.64	60.67
3	60.92	60.45	61.10
4	59.49	57.94	57.93
Delta	3.34	2.70	3.17
Rank	1	3	2

CONCLUSION

The feasibility of coconut shell for activated carbon production was studied using phosphoric acid as an activating agent. The optimization experiments were designed utilizing Taguchi L16 orthogonal

array. The control factors such as activation temperature, time and impregnation ratio were varied as 400-700 °C, 30-120 min and 0.5-3:1, respectively. Maximum iodine value of 1230 mg g^{-1} was achieved at 600 °C, 60 min and 2:1. Temperature had more significant effect on pore characteristics of activated carbon than impregnation ratio and time. The produced activated carbon can be used as adsorbent, catalyst, catalyst support and electrodes.

ACKNOWLEDGMENT

The authors would like to thank the Coconut Development Board (CDB), Kerala, India, and the Department of Renewable Energy Engineering, Agricultural Engineering College and Research Institute, Tamil Nadu Agricultural University, Coimbatore, for their support in providing necessary funding and facilities to carry out this work.

REFERENCES

ASTM. 2006. Standard Test Method for Determination of Iodine Number of Activated Carbon 1. *ASTM Int*. 94: 1-5.

Charola, S., Yadav, R., Das, P. and Maiti, S. 2018. Fixed-bed adsorption of Reactive Orange 84 dye onto activated carbon prepared from empty cotton flower agro-waste. *Sustain. Environ. Res*. 28 : 298-308.

Gao, Y., Yue, Q., Gao, B. and Li, A. 2020. Insight into activated carbon from different kinds of chemical activating agents: A review. *Sci. Total Environ.* 746: 141094.

Haghbin, M.R. and Niknam Shahrak, M. 2021. Process conditions optimization for the fabrication of highly porous activated carbon from date palm bark wastes for removing pollutants from water. *Powder Technol.* 377 : 890-899.

Karmakar, B., Dhawane, S.H. and Halder, G. 2018. Optimization of biodiesel production from castor oil by Taguchi design. *J. Environ. Chem. Eng*. 6 : 2684-2695.

Kumar, A. and Jena, H.M. 2016. Preparation and characterization of high surface area activated carbon from Fox nut (Euryale ferox) shell by chemical activation with H3PO4. *Results Phys.* 6 : 651-658.

Mbarki, F., Selmi, T., Kesraoui, A. and Seffen, M. 2022. Low-cost activated carbon preparation from Corn stigmata fibers chemically activated using H3PO4, ZnCl2 and KOH: Study of methylene blue adsorption, stochastic isotherm and fractal kinetic. *Ind. Crops Prod.* 178 : 114546.

Moralý, U., Demiral, H. and ªensöz, S. 2018. Optimization of activated carbon production from sunflower seed extracted meal: Taguchi design of experiment approach and analysis of variance. *J. Clean. Prod*. 189 : 602-611.

Nahil, M.A. and Williams, P.T. 2012. Pore characteristics of activated carbons from the phosphoric acid chemical activation of cotton stalks. *Biomass and Bioenergy*. 37 : 142-149.

Nedjai, R., Kabbashi, N.A., Alam, M.Z. and Al-Khatib, M.F.R. 2021. Production and Characterization of Activated Carbon from Baobab Fruit Shells by Chemical Activation Using ZnCl2, H3PO4 and KOH. *J. Phys. Conf. Ser.* 2129.

Ogungbenro, A.E., Quang, D. V., Al-Ali, K.A., Vega, L.F. and Abu-Zahra, M.R.M. 2020. Synthesis and characterization of activated carbon from biomass date seeds for carbon dioxide adsorption. *J. Environ. Chem. Eng.* 8 : 104257.

Tuli, F.J., Hossain, A., Kibria, A.K.M.F., Tareq, A.R.M., Mamun, S.M.M.A. and Ullah, M.A. 2020. Environmental Nanotechnology , Monitoring & Management Removal of methylene blue from water by low-cost activated carbon prepared from tea waste/: A study of adsorption isotherm and kinetics. *Environ. Nanotechnology, Monit. Manag.* 14 : 100354.

Yagmur, E., Inal, I.I.G., Gokce, Y., Ulusoy Ghobadi, T.G., Aktar, T. and Aktas, Z. 2018. Examination of gas and solid products during the preparation of activated carbon using phosphoric acid. *J. Environ. Manage*. 228 : 328-335.

Yang, Q., Wu, P., Liu, J., Rehman, S., Ahmed, Z., Ruan, B. and Zhu, N. 2020. Batch interaction of emerging tetracycline contaminant with novel phosphoric acid activated corn straw porous carbon: Adsorption rate and nature of mechanism. *Environ. Res.* 181: 108899.