Journal of Environmental Treatment Techniques 2014, Volume 2, Issue 1, Pages: 23-27

Enviro. Treat. Tech.
ISSN: 2309-1185
Journal weblink: http://www.jett.dormaj.com

Evaluation of wastewater treatment contaminated with formaldehyde by using activated carbon prepared from cypress leaf

Amin Goli[1] , Poyan Ghanbarnejad[2], Benyamin Bayat[2], Zahra Abaspur[2], Zakiye Norozzade[2], Mohammad Frotan[3], Amirreza Talaiekhozani[4,5]

1. Jami Institute of Technology, Department of Mechanical engineering, Isfahan, Iran.
2. Jami Institute of Technology, Department of Chemical Engineering, Isfahan, Iran.
3. University of Hakim Sabzevari, Department of Geography and Urban Planning, Sabzevar, Iran.
4. Jami Institute of Technology, Department of Civil Engineering, Isfahan, Iran.
5. Institute of Environmental and Water Resources Management, Water Research Alliance, Universiti Teknologi Malaysia, UTM Skudai, 81310 Johor Bahru, Malaysia.

Received: 03/20/2014 Accepted: 04/05/2014 Published: 04/10/2014

Abstract

Rising standards of effluent discharge to the environment has led many researches to look for more effective and at the same time, economical treatment methods. Utilization of Activated carbon, as an effective and economical method, could be a helpful solution in this regard. The purpose of this study is to investigate the preparation of activated carbon from cypress tree leaves and its abilities in treatment of formaldehyde contaminated wastewater. Besides investigating the adsorption ability of the adsorbent, the effects of various parameters such as pH, retention time, Formaldehyde concentration and temperature were also investigated. Test results obtained from this study demonstrate that the most efficient condition for formaldehyde removal by the adsorbent is on the temperature of 10 C, retention time of 5 minutes and pH equal to 7 which led to a 98% formaldehyde removal. According to the survey conducted in this study it can be concluded that the utilization of activated carbon, obtained from the cypress tree leaves, is an efficient and economical solution in the removal of toxic waste such as formaldehyde.

***Keywords*:** Formaldehyde, Adsorption, Formalin, Cypress leaf

1 Introduction

Formaldehyde is an organic chemical compound which has a wide range of application in various industries such as melamine production, adhesives, and wooden items including fixtures and furniture. It is also widely used as a solvent in paint industry [1, 2]. 10 ppm of this highly toxic substance in the air can cause severe intoxication in humans and can also lead to skin diseases, lung cancer and respiratory tract and may deliver serious damage to the eyes [3,4]. After being discharged to the environment, formaldehyde can enter the groundwater resources and cause water contamination [5, 6, 7, 8, 9]. The adsorption is generally the process of material accumulation at the interface between the two phases. Various materials could be utilized in an adsorption process, such as: powder or granular activated carbon, ash, bentonite, phosphate hydrate cellulose, biomass, coal, coke and china clay. Activated carbon is the most effective adsorbent in toxic waste treatment. The high costs of the adsorbents restoration have led many researchers to look for new adsorbents in this regard, thus, many investigations have been conducted for the application and the use of low-cost adsorbents. The industrial waste standards and regulations are becoming increasingly strict; on the other hand, due to the expansion of various industries, new efficient and cheap treatment methods are ought to be used. Among all, the trees Foliages have been widely applied due to low cost, ease of access to the required raw material and the ability to remove heavy metals [10, 11, 12, 13, 14, 15]. Various researches including libojeng ET al were able to achieve 98% pollutant removal from the waste water by utilizing the activated carbon obtained from tobacco roots. Badkoby et al were also able to achieve chromium (VI) Ions removal with a yield of 37% by utilizing the activated carbon obtained from poplar wood. Darvishi et al utilized biological sludge to eliminate cadmium [16]. Samadi et al took advantage of bone char to eliminate fluorine from drinking water. They realized that the removal rate of fluorine is dependent on parameters such as retention time, bone size and pH of the solvent [17]. Rahmani et al compared the performance of three kinds of saw dust as low cost adsorbent in the removal of arsenic and cyanide. They were able to achieve 83% cyanide removal by using poplar sawdust and 96% arsenic removal by utilizing Raj tree sawdust [18]. Eramy was also able to utilize activated

Corresponding Author: Amirreza Talaiekhozani,
(a) Department of Civil Engineering Jami Institute of Technology, *E-mail*: atalaie@jami.ac.ir.
(b) Institute of Environmental and Water Resources Management, Water Research Alliance, Universiti Teknologi Malaysia, UTM Skudai, 81310 Johor Bahru, Malaysia.

carbon obtained from coconut fibers to adsorb nickel as a heavy metal. His research revealed that the adsorption rate in highly dependent on the pH rate and the highest removal rate was achieved in the pH rate of 4.

The main aim of this study is to evaluate the formaldehyde removal efficiency by utilizing an activated carbon obtained from cypress tree leaves. After the production of formaldehyde contaminated waste in laboratory conditions, the capabilities of this adsorbent was scrutinized. Various affecting parameters such as retention time, pH, Temperature and initial concentration were also evaluated.

2 Methods and materials

2.1 Experiment methods

In this study, the formaldehyde concentration was determined using the Chemical Oxygen Demand (COD) method. COD was also done using the Closed Distillation method. In addition, the pH rate was measured using a HI-8424 Digital pH meter made by Henna instrument factory. Temperature was also measured using a mercury thermometer[19, 20].

2.2 Production of Activated Carbon from cypress tree leaf

Prior to the experiment, a certain amount of cypress tree leaf was prepared and it was added to an oven to dry up after being washed for two hours. The dried leaves were then added to a pod with high thermal resistance. The container was filled completely with dried leaves and the led was kept shut tightly to avoid air ingression so there would be a limited oxygen condition within the container. The prepared sample was then kept in a furnace for 3 hours under the temperature of 700 C. During this three hours period, the leaf's organic material would produce water vapor, Carbon Dioxide and ash due to high temperature. Although due to the lack of oxygen, the leaves were only partially converted into the mentioned materials. This leads to a highly porous structure and wide surface on the leaf and led to a wide surface. After 3 hours, the leaves were transferred into a desiccator and were cooled up-to the ambient temperature. As mentioned before, large parts of the leaves were converted into water vapor, Carbon dioxide and ash. However, the generated ash would remain in the depths of the leaves structure which reduces the porosity of the produced activated carbon. Thus, to remove parts of the in-depth ash, the leaves were added to an autoclave and were pressurized for 15 minutes by hot steam. This has led to a special surface for activated carbon.

2.3 Evaluation pH effects on formaldehyde adsorption onto cypress tree leaf

In this part of the experiment the effects of pH shifts in formaldehyde adsorption process were evaluated. For this purpose the pH rates of 2, 7 and 14 were scrutinized. In this experiment 3 separated 250 ml Erlenmeyer flasks containing 100 ml distilled water were prepared and 0.1 ml formaldehyde and 0.5 g of activated carbon were administered to it. Administration of this amount of formaldehyde into 100 ml of water generated a COD rate of 3/19 mg/l. with adding a proper amount of H_2So_4 and Na(OH) the pH rates of each Erlenmeyer flask were adjusted onto the desired pH rates (2, 7 and 14) 3 more Erlenmeyer flasks with the similar conditions were prepared as holotype samples although without the activated carbon. All samples were kept under the fixed temperature of 31 C for two hours inside an Incubator. At the end of the mentioned period, the CODs of each sample were measured.

2.4 Evaluation of temperature effects on formaldehyde removal by activated carbon

In this survey, the temperatures of 10, 15, 30, 40, 60, 80 and 100 were respectively evaluated to investigate the effects of temperature on formaldehyde adsorption by activated carbon. 7 separated 250 ml Erlenmeyer flasks containing 100 ml distilled water were prepared and 0.1 ml formaldehyde and 0.5 g of activated carbon were administered to it. In all samples the pH rate was adjusted to 7. A holotype sample was prepared for each sample with no activated carbon. The samples were kept under the mentioned temperatures inside an Incubator for 2 hours. After the mentioned period, the remained COD of the samples were measured.

2.5 Evaluation of Retention time in formaldehyde adsorption by activated carbon

One of such effecting factors in formaldehyde adsorption by activated carbon is considered to be retention time. The retention times of 5, 10, 15, 20, 30 and 60 were respectively evaluated in this survey. 6 separated 250 ml Erlenmeyer flasks containing 100 ml distilled water were prepared and 0.1 ml formaldehyde and 0.5 g of activated carbon were administered to it. In all samples the pH rate was adjusted to 7. A holotype sample was prepared for each sample with no activated carbon. Each sample was kept under the temperature of 31 C for the mentioned retention times. At the end, the COD rates were carefully measured.

2.6 Evaluating the effects of formaldehyde concentration in adsorption by activated carbon

In this part of the experiment various concentration of the pollutant were prepared to evaluate its effects on the adsorption efficiency. 4 separate 250 ml Erlenmeyer flasks containing 100 ml distilled water were prepared and were named as "Flask 1 to 4". Respectively 0/02, 0/04, 0/08 and 0/1 ml of 37% formaldehyde solution were added to each flask. 0/5 g of activated carbon was also administered to each sample. In all samples the pH rate was adjusted to 7. The samples were kept under the temperature of 31 C inside an Incubator for 2 hours. After the mentioned period, the remained COD of the samples were measured.

3 Results

3.1 Evaluation of pH effects on formaldehyde adsorption by Cypress tree leaf

Numerous researches regarding the pH effects on the adsorption efficiencies on several adsorbents and pollutants identify pH as one of the affecting parameters. it is of high importance to carefully observe this parameter in this study. The test result of this section is illustrated in Figure.1. As it is indicated, the study was carried in 3 different pH rates (2, 7, and 14) which relatively represent the Acidic, Neutral and Alkali conditions. These results indicate that Acidic and Alkali pH rates can decrease the adsorption efficiency up to 20%. Thus, the neutral pH was identified as the most proper condition. Under this condition, we were able to achieve approximately 99% formaldehyde removal.

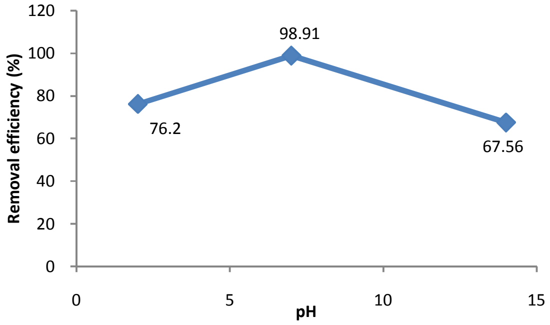

Fig.1: Formaldehyde removal efficiency changes in adsorption by activated carbon in different pHs

3.2 Evaluation of Temperature shifts in formaldehyde adsorption efficiency

Of other affecting parameters in the adsorption efficiency the temperature could be named. As it could be deduced from the test results of this section (Demonstrated in Figure.2) the temperature and adsorption efficiency have converse relation. In other words, by increasing the temperature, the adsorption efficiency will be decreased. Numerous studies illustrate that there is generally two types of adsorption involved in the process. They are either of chemical type or of physical type. In the case of chemical adsorption, by increasing the temperature, the adsorption efficiency will be increased. And in the case of the physical adsorption, the relation is converse. So based on the obtained results presented in Figure.2, the formaldehyde adsorption on cypress tree leaf is considered to be of physical type. Based on the conducted linear practice in between the Adsorption efficiency and temperature the following equation with the constant coefficient of $R^2 = 0.924$ was obtained.

$$y = -0.328x + 0.958$$

This equation could be used to approximately predict the adsorption efficiency in different temperatures.

3.3 Evaluation of retention time effects in formaldehyde removal efficiency

The maximum adsorption rate of the pollutant requires enough contact interval between the adsorbent and the pollutant. This contact interval would be variable in different conditions and adsorbents, thus, it is of great importance to evaluate the effects of retention time in the removal efficiency. In this study, the retention intervals of 5 to 60 minutes were evaluated. The test results indicate that gradually, the removal efficiency will reach a fixed number.

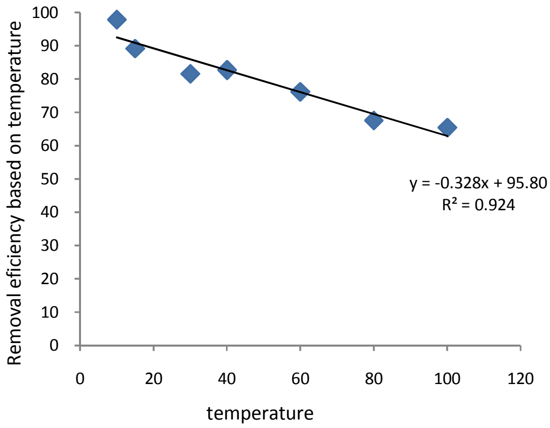

Fig.2: Formaldehyde removal efficiency by activated carbon on different temperatures

As it is illustrated in Figure.3, the removal efficiency slope is really high between the intervals of 5 to 20 minutes, although this slope will be heading toward 0 in retention intervals of 20 to 60 minutes and stands stable on the approximate value of 90%. This is due to the saturation of the adsorption sites within the adsorbent. In such condition, the adsorbent material no longer has the required removal capacity thus stands stable on higher intervals. This case is also observed in other types of adsorbent.

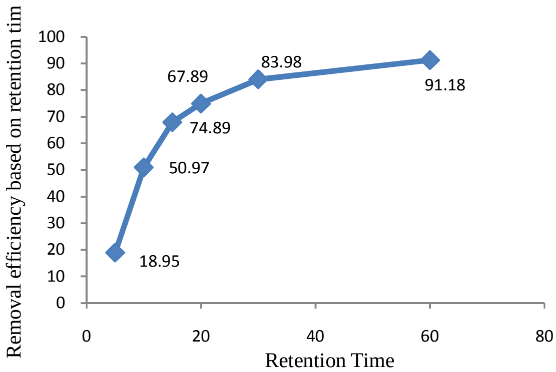

Fig.3: removal efficiency shifts in different retention times

3.4. Evaluation of formaldehyde concentration effect on the adsorption efficiency

The intensification of formaldehyde concentration in the environment can lead to higher removal efficiency but stands stable on higher concentrations. It is predicted that with further intensification of the concentration, the adsorbent will no longer have the removal capacity thus the efficiency will continuously rebate.

3.5 Modeling of the formaldehyde adsorption

The formaldehyde adsorption process modeling is crucial for the applicability of the attained results. This modeling was conducted using the Langmuir equation. The equation used in this study is shown in eq.1.

$$\frac{\mathrm{c}}{\mathrm{f}} = \left(\frac{1}{\mathrm{f_{max}}}\right) c + \frac{1}{\mathrm{k} \times \mathrm{f_{max}}} \qquad (1)$$

Where c is the initial formaldehyde concentration in mg/l, f is the adsorbed formaldehyde onto the adsorbent in mg/l, f_{max} is the maximum adsorbed formaldehyde onto the adsorbent at any concentration in mg/l and k is the Langmuir constant coefficient in mg/l. as it is clear; the above equation is a linear equation similar to $y = ax + b$.

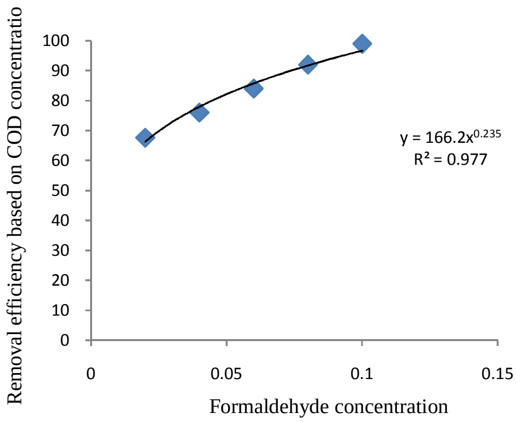

Fig.4: removal efficiency shifts in various concentrations

So in eq.1 y is equal to C/F factor and x is equal to c factor. By plotting these factors against one another, constants of (a) and (b) which are respectively equal to $\frac{1}{f_{max}}$ and $\frac{1}{k\times f_{max}}$ could be calculated. The diagram shown in fiq.5 illustrates the result of the plotted factors and the regression between them. As it is clear, the linear equation resulted from the regression is $y = -0.174x + 1.548$ in which (a) and (b) coefficients are respectively equal to 0/174 and 1/548. According to the conducted calculations f_{max} is equal to 5/74 mg/l and k is equal to 0/112 mg/l.

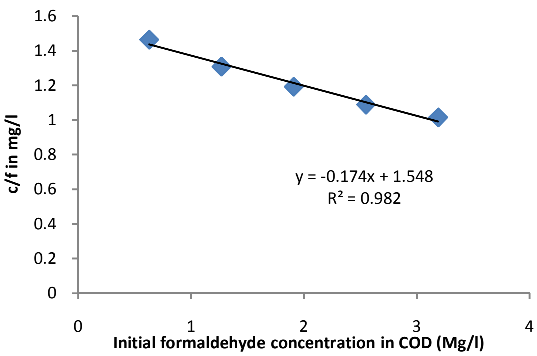

Fig.5: Resulting plotted regression of $\frac{1}{k\times f_{max}}$ against $\frac{1}{f_{max}}$

By using eq.1 and the calculated constant coefficients, formaldehyde adsorption efficiencies in various concentrations are anticipated and the results are shown in fig.6. As it is shown in this figure, in low formaldehyde concentrations the adsorbed formaldehyde increases linearly with increases in initial formaldehyde concentration. However with increases in the initial formaldehyde concentration, the relation between the adsorption efficiency and the initial concentration will no longer remain linear and will drift toward a fixed number. Given that 500mg activated carbon, attained from cypress tree leaves, were used on 100ml of water contaminated water, it could be concluded that 500mg of this type of activated carbon is capable of removing 5/5 mg formaldehyde COD. With the help of this proportion the equation.2 was offered to calculate the required amount of activated carbon to remove 100% of the formaldehyde in various concentrations.

$$a = \frac{(0.5 \times c)}{5.5} \qquad (2)$$

Where a is the required amount of the adsorbent in gram and c is the formaldehyde concentration. The amount of required adsorbent on various formaldehyde concentrations were calculated using eq.2 and were shown in fig.6. As it is clear from this figure, the formaldehyde removal efficiency drifts toward a fixed number when the required adsorbent raises proportional to the growth of the formaldehyde concentration.

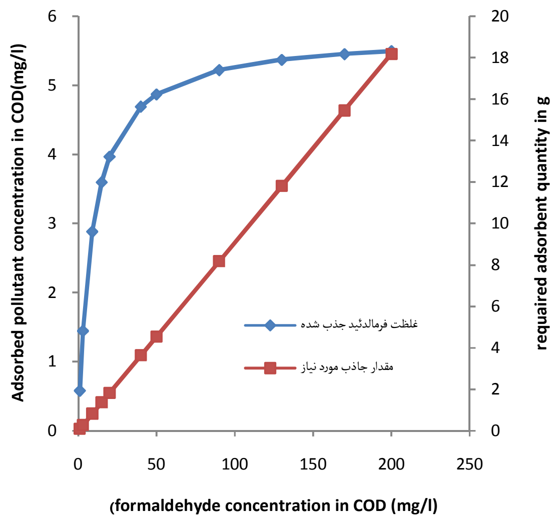

Fig.6: Prediction of removable formaldehyde and the required adsorbent rate in various formaldehyde concentrations

4 *Conclusions*

The utilized activated carbon in this study was attained by Heating cypress tree leaf at the presence of limited oxygen and ultimately by using 15atm steam pressure. Studies on the attained activated carbon indicate that the most optimum pH rate is neutral pH (about 7). It was also indicated that there is a converse relation between temperature and the removal efficiency which confirms that the adsorption process is of physical type. Thus, the produced adsorbent in this study could be an inexpensive replacement for the more expensive formaldehyde contaminated wastewater treatment methods.

References

1- Goli A, Talaiekhozani A, (2013) Evaluation of biological treatment of air stream contaminated with

formaldehyde using bio-trickling filter, *Tolue behdasht journal of Yazd*, In press.
2- Mohamad Ali Fulazzaky, Amirreza Talaiekhozani, Mohanadoss Ponraj, M.Z. Abd Majid, Tony Hadibarata, Amin Goli, (2013) Biofiltration process as an ideal approach to remove pollutants from polluted air, In Press in *Desalination and Water Treatment*, 1-16.
3- Mohamad Ali Fulazzaky, Amirreza Talaiekhozani, Mohanadoss Ponraj, M.Z. Abd Majid, Tony Hadibarata, Amin Goli, (2013) Biofiltration process as an ideal approach to remove pollutants from polluted air, In Press in *Desalination and Water Treatment*, 1-16.
4- Talaiekhozani, A., Ali Fulazzaky, M., Ponraj, M., & Abd Majid, M. Z. (2013). Removal of formaldehyde from polluted air in a biotrickling filter reactor. In Press in *Desalination and Water Treatment*, 1-9
5- Zhogjun xu, na qin, jinggang wang, Hua Ton., 2010. Formaldehyde biofilteration as affected by spider plant. Bioresource technology 101, 6930-6934.
6- Squire, R.A., Cameron, L.L., 1984. An analysis if potential carcinogenic risk from formaldehyde. Reul. Toxicol. PHarmacol. 4, 107-129.
7- Wolverton, B.C., wolverton, J.D ,. 1993. Plant and soil microorganisms: removal of formaldehyde, xylesne, and ammonia from the indoor environment. J. miss. Acad. Sci. 38, 11-15.
8- Oliseria svwb etal.formaldehyde degraddition anan anaerobic packed-bed bioreactor – water research 2004 – vol .38-pp, 1685-1694.
9- Rahmani. A., Samarghandi. M. R., Evaluation of electerochemical in COD removal from wastewater, water&wastewater journal 2007 64(18):9-12.
10- Walker .G. M., Weatherley, L.R.(1997).Adsorption of Acid Dyes On To Granular Activated Carbon In Fixed BEDS wat, 31 , 2093-2101.
11- Yamada , T. and T sumli, K., Adsorbent Body Including Activated Carbon and Inorganic Binder for Bonding Together Activated Carbon Grains, and Metod of Producing the Adsorbent Body. US Patent 5,685,986(1997).
12- Duong . D.D.(1998). Ed .eds. Adsorption Analysis : Equilibria And Kinetics:Imperial college Press.
13- MacDowall , j., Metod of Producing Granular Activated Carbon.US Patent 5,162,286(1992).
14- Yan, Z.Q.,McCue, j., and Tolles, E., Preparation of High Activity, High Density Activated Carbon with Activatable Binder. US Patent 5,538,932(1996).
15- A. C. Deiana , D. L. Granados, L. M. Petkovic, M. F . Sardella and H. S. Silva.
16- Darvishi, R., Shams, Gh., Jonidi, A., Jorfi. S., Salehi, S., Evaluation of activator materials on bioabsorption of cadmium by biosolid, *The 12th National Congress of Environmental Health*, 2008.
17- Samadi M T, Khodabandelo Z, Khodabakhsh F, Kashitarash Z, Investigating fluoride Removal from Water by Usingofbone char, *The 12th National Congress of Environmental Health*, 2008.
18- Rahmani A R, Samadi M T, Ghimali, M, Motaghipor H, Mirzaee S. comparison of arsenic & cyanide removal from effluents by using of Holly, Sallow and Poplar Hamadan sawdust, *The 12th National Congress of Environmental Health*, 2008.
19- APHA. Standard methods for the Examination of water&wastewater. 21 Edition, Washington DC,USA. 2005.
20- ASTM D 3172-3175, 1999.Annual Book of ASTM Standard, Section 5 petroleum products, and Fossil Fuels, Vol05.05 Gaseous Fuels, Coal and Coke.