Journal of ENVIRONMENTAL Treatment Techniques

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

Evaluation of Formaldehyde Adsorption by Human Hair and Sheep Wool in Industrial Wastewater with High Concentration

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Received: 29/01/2014 Revised: 04/03/2014 Published: 09/03/2014

Abstract

One of such pollutants which can be detected in many different industrial wastewaters is formaldehyde. The biological treatment of wastewater contaminated with formaldehyde is very difficult due to its antibacterial property. However the removal of formaldehyde using adsorption process such as activated carbon is passable but it may be expensive. The main aim of this study is the adsorption of formaldehyde on human hair and sheep wool as an adsorbent. Human hair and sheep wool are cheap and they can be disposed by common methods such as incineration. In this study, the concentration of formaldehyde in wastewater was measured during the adsorption process using chemical oxygen demand method (COD). Then the effects of different parameters such as pH, temperature, hydraulic retention time, formaldehyde concentration, and weight of human hair and sheep wool were evaluated. Finally, based on our results, optimum values of the parameters were investigated. The obtained results showed that formaldehyde can be adsorbed on human hair significantly. Also it was revealed that formaldehyde can be removed from wastewater during 5 min, and the increase of hydraulic retention time from 5 to 25 min is not effective on formaldehyde adsorption efficiency. The results elaborated that, formaldehyde adsorption efficiency using white hair, colored hair by chemical colors or natural ones (e g. Henna), and sheep wool is as efficient as common human hair. The results attained from this research showed that using human hair or sheep woolfor removing formaldehyde from industrial wastewater is possible.

Keyword: Formaldehyde, Adsorption process, Human hair, Formalin, Sheep wool.

1 Introduction

Nowadays, wastewater produced by various industrial activities has caused several environmental problems. Some industries produce wastewater containing dangerous substances which have a negative impact on humans and animals [1, 2]. Therefore, much attention has been given to development of techniques for treatment of this kind of wastewater.

Formaldehyde (CH_2O) is a dangerous and toxic substance detected in the wastewater produced by adhesive, chemical and petrochemical industries. Approximately formaldehyde concentration in industrial wastewaters is more than 10 mg/l [3] and it has been ranked firstly in the environmental effects ranking of 45 harmful chemicals by Edwards et al, [4]. As a disinfectant, formaldehyde 0.5% solution is capable of killing microorganisms within 6 to 12 hours [5, 6]. Formaldehyde vapor is a nasty gas which can stimulate the skin, mucous membranes and eyes and causes complications such as respiratory problems, asthma and respiratory carcinogenicity on experimental animals. Therefore, it can probably be highly carcinogenic for humans [4, 5, 6, and 7].

Many physical and biological techniques have been developed for formaldehyde removal. Physical techniques for removal of formaldehyde, has particularly been attracted for environmentalists due to complexity in biological removal of this compound [7].

In many industries, the wastewater contaminated with formaldehyde is collected into lagoons to evaporate over time. This causes the formaldehyde ingression into the atmosphere and further pollution of the air [8].

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So using adsorbents could be a suitable solution for removal of wastewater, containing highly toxic compounds such as formaldehyde [9]. In the adsorption process, the materials are adsorbed on solid surface, and thus are separated from the solution due to gravity force between functional groups on a solid surface and molecules of the adsorbed materials; thus, the adsorption intensity depends on the nature of the adsorbent and the adsorbed materials [9]. This process can also be used for the separation of gas and liquid.

Usually solid adsorbents are used as granule or as powder and their diameters vary from 12 to 50 mm. these adsorbents are capable of available adsorbing impurities in gas or a liquid. Properties of a suitable adsorbent are as following: high adsorption rate in the environment, capable of disposing the adsorbed materials at a temperature above ambient temperature [9].

The amount of a substance adsorbed on an adsorbent depends on the amount and the quality of the contact area, temperature, density and the type of the adsorbed material [9]. If the adsorption is accompanied by chemical reaction and altered by the chemicals nature of the adsorbent, the contamination properties of the produced material should be much lesser than the contamination properties of the adsorbed materials [9]. Important industrial adsorbents have a contact area of 1000 m^2/g of the adsorbent. This amount of contact area will only be possible when the adsorbent has a porous structure. Also, the porosity network should be in a way which provides the adsorbing material with access to all the inner parts of the adsorbent [9].

Previously, the adsorption process of wastewater treatment was not widely used, but today, due to the increasing demand for quality by treated wastewater, the adsorption process has received much more attention. Usually the wastewater treatment process is finally considered with an adsorbent material which is applied to the water that has been previously subjected to conventional biological treatment.

Today, much research has been carried out to utilize various natural materials as adsorbents, especially wastewater generated by human activities. Such materials are cheaper and effective. For example, Samar Ghandyet al. evaluated the use of sawdust for removing chrome from wastewater. They reported that pH, contact time, amount of adsorbent material and initial concentration of chrome are the most effective factors on chrome removal by sawdust [8]. Particle size of adsorbent was introduced as another effective factor on removal of pollutants from wastewater using sawdust by Rahmani et al. [12]. Many natural adsorbents such as charcoaled bones, olive kernel, human hair and biological sludge were used to remove pollutants from wastewater [9, 10, 11, 12 and 17]. Although several researches on pollutants removal from wastewater using various adsorbents were conducted, but no comprehensive study was ever carried out on human and animal hair as a natural adsorbent.

The main aim of this research is to evaluate the adsorption rate of formaldehyde (as a hazardous industrial pollutant) by taking advantage of different types of hair (e.g. human or animal) as inexpensive adsorbents. In this study, the wastewater containing formaldehyde was generated in laboratory conditions. After that, the adsorption ability of formaldehyde onto human and animal hair and their effective factors were scrutinized.

2 Material and Methods

2.1 Production of synthetic wastewater with different concentrations

In this study, the synthetic contaminated wastewater was produced usinga mixture of distilled water and different amount of 37% stock solution of formaldehyde. So 7.5, 5.11 and 3.17 ml of 37% formaldehyde was delivered up to the volume of 250 ml distilled water to generate the synthetic contaminated wastewater with concentrations of 9000, 18000 and 27000 milligrams per liter and chemical oxygen demand (COD) of 9600, 32000 and 41600 mg/l respectively.

2.2 Evaluation of hair quantityfor adsorbing formaldehyde

Since amount of hair used as adsorbent is a very important factor in removal efficiency of formaldehyde, it is crucial to evaluate this factor. At this stage of the experiment, the formaldehyde adsorption from synthetic wastewater was conducted by using 0.5, 1, 1.5, and 2 gram of hair. The experiment was carried out under the following conditions: 30°C reaction temperature and 27000 mg of formaldehyde concentration with pH equal to 5, retention time of 10 minutes. Before starting the experiment the hair was washed with distilled water. After that, hair was added to a 250 ml Erlenmeyer containing 100 ml of synthetic wastewater. At the end of the experiment, COD of synthetic wastewater was measured and adsorption efficiency was determined.

2.3 Evaluation the effect of retention time on formaldehyde removal

Retention time can affect the formaldehyde adsorption efficiency. Therefore, for this purpose, the retention times of 10, 15, 20, and 25 minutes were evaluated. The temperature during the experiment was adjusted on 30°C and the pH of the environment was equal to 5. At this stage of experiment 0.5 g of hair was added into a 250 ml Erlenmeyer flask containing 100 ml synthetic wastewater with COD of 27000 mg/l. After 5 minutes, the amount of formaldehyde remained on the Erlenmeyer flask was evaluated using COD test.

2.4 Evaluation of pH effect on formaldehyde removal

Previous studies have indicated that the pH of the wastewater is one of the important parameters in the adsorption process. In this part of the experiment, the effect of various pHs (3, 5, 6, 7, 9 and 11) on ability of formaldehyde removal using human hair was studied. In all the tests, the following conditions were chosen: Temperature 30 ° C, 5 min retention time, concentration of 27000 mg/ l and 0.5 g of hair. To adjust the pH, nitric acid (0.2 M) and NaOH (0.2 M) solutions were used. 6 separated 250 ml Erlenmeyer flasks containing 100 ml of synthetic wastewater with the above mentioned pH were prepared. The amount of 0.5g of hair was washed in distilled water and was added on each Erlenmeyer flask.

After 5 minutes the amount of adsorbed formaldehyde in different pHs were evaluated using COD test.

2.5 Evaluation of temperature effect on formaldehyde removal usinghuman hair

Temperature is another effective factor on adsorption process. In this part of experiment, the effect of different temperatures (30, 35,40,45,50 and 55 ℃) on formaldehyde removal by human hair were evaluated. In all the tests the following conditions were chosen: wastewater pH of 5, wastewater COD concentration of 27000 mg/l, retention time of 5 minutes and 0.5 g of hair. Before using the hair, it was washed with distilled water. Then 0.5 g of hair was added into a 250 ml Erlenmeyer containing 100 ml of synthetic wastewater contaminated with formaldehyde. After 5 minutes, the amount of reminded formaldehyde was measured using COD test.

2.6 Evaluation of human white hair (No pigment) in formaldehyde adsorption

Based on our previous studies, no research was conducted on hairs without melanin pigments; therefore, this type of hair was studied in this part of experiment. In this test, 0.25 g of white hair was added into a 250 ml Erlenmeyer flask containing formaldehyde contaminated synthetic wastewater after washing with distilled water. The Erlenmeyer flask was placed on following conditions. Retention time of 5 minutes, temperature of 60C, pH of 5, COD of 27000 mg/l are considered, At the end of the experiment, the concentration of the remained formaldehyde was measured based on COD test.

2.7 Evaluating the colored human hair in formaldehyde adsorption

Various substances with chemical compounds observed in dyed hair could decrease or increase the formaldehyde removal efficiency. Thus, a combination of these types of hair were collected and studied. The amount of 0.25 g of hair was shed in a 1 liter of Erlenmeyer flask containing 100 ml synthetic wastewater with concentration of 27000 mg / l and pH of 5 for 5 minutes at temperature of 60 C. At the end of the test, the COD of the solution was measured and the formaldehyde efficiency was indicated.

2.8 Evaluation of dyed human hair with Henna in formaldehyde adsorption

Natural substances like Henna could also alter the chemical structure of human hair. Thus, evaluation of the formaldehyde removal efficiency by this type of hair has a great importance. In this test, 0.5 g of Henna dyed hair was added in a 250 ml Erlenmeyer flask containing 100 ml synthetic wastewater with COD of 27000 mg/l with pH equal to 5. The mixture of Henna dyed hair and wastewater were kept in contact for 5 minutes. At the end, the solution COD was measured and the removal efficiency was indicated.

2.9 Evaluation of Sheep wool in formaldehyde adsorption

In this stage of experiment, the sheep wool efficiency in the removal of wastewater contaminated with formaldehyde was evaluated. A certain amount of sheep wool was rinsed with 2 Molar nitric acid and then completely dried by distilled water. 0.5 g of sheep wool was placed in a 250 ml Erlenmeyer flask containing 100 ml of synthetic wastewater with the concentration of 27000 mg/l and pH of 5. This Erlenmeyer flask was placed at temperature of 60 ℃ for 5 minutes. At the end, the amount of adsorbed formaldehyde was measured by using COD test.

2.10 Methods of Experiment

In this research, a digital pH meter was used to determine the pH rate. Astandard method of COD determination was used in this study which is mentioned in the Standard methods book [11]. To stabilize the temperature in the various tests, a steam bath device was used controlling the temperature by indirect heat.

3 Results

3.1 The Effects of retention time in formaldehyde adsorption

As indicated in figure one, increments in hydraulic retention times from 5 to 25 minutes, the formaldehyde adsorption by human hair, did not increase. Changes in retention time have only led to a few tenths of percent increase in adsorption efficiency. In a research carried out by Talaeiet. al, it was reported that changes more than 5 minutes in retention time, not only has no positive impacts on adsorption efficiency of human hair, but could also lead to negative impacts in longer retention times [7]. However, this effect is not observed in other types of adsorbent. For example Samarghandy et. al, reported that increments in retention time has led to increased pollutant adsorption on the adsorbent [6]. The test results show that human hair is capable of removing more than 98% of formaldehyde from wastewaters in slight amounts of time. Slight retention time makes the human hair suitable for use in complementary treatment of wastewater contaminated with formaldehyde in industrial scale.

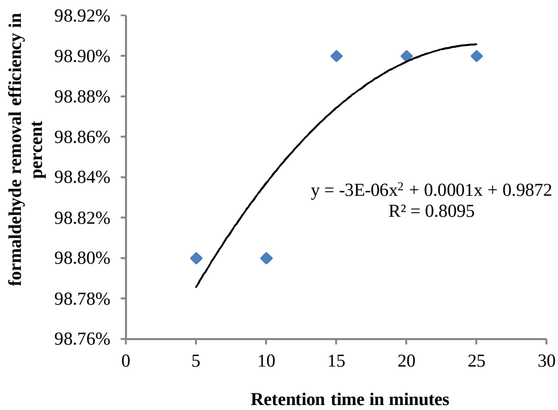

Figure.1: Formaldehyde removal rate in different hydraulic retention times

3.2 Evaluation of temperature effects

Test results for formaldehyde adsorption in different retention times and stable temperatures have been shown in figure.2. Last studies on different types of adsorbents indicate that temperature could have positive or negative effects on the adsorption rate. Basically, if the adsorption is a chemical type, increments in temperature could have positive effects on the adsorption efficiency. However, in physical adsorption, increases in temperature will have potentially negative effect on the adsorption efficiency [7]. As indicated in figure.2, by increasing the temperature, the adsorption rate could be increased. As it is shown, the highest adsorption efficiencies belong to temperatures of 60 and 65℃. However, this rise is very small, in a way which the removal efficiency differences of the lowest (30℃) and the highest (65℃) temperatures were only 3%. The result of non-linear regression between formaldehyde removal efficiency and the temperature was ended to the following equation 1.

$$y = 0.924x^{0.016} \qquad (1)$$

Which y is formaldehyde removal efficiency in percent and x is the temperature in ℃. Using this equation to predict the removal efficiencies in temperatures close to boiling point (99℃) shows that the efficiency will remain constant on 99.4%. Test results indicate that temperature has a little effect on formaldehyde removal by human hair. However, previous research on this issue reported that, a sharp increase in removal efficiency of formaldehyde by human hair was observed. Therefore, formaldehyde adsorption on human hair must be a chemical adsorption [7]. However, such conditions were not observed in this study which could be related to the chosen hair type and other unknown conditions which requires further study.

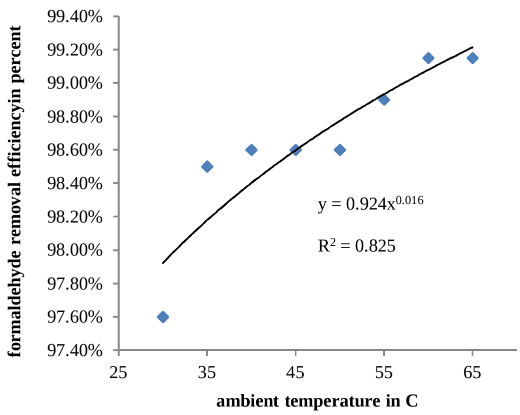

Figure.2: Formaldehyde adsorption rate changes in different temperatures

3.3 Evaluating the pH effects

Another effective parameter on the adsorption process is pH. Numerous reports have stated the effects of this parameter on the adsorption process [8, 9]. This effect is evident in Figure.3. Different pH rates between 3 and 11 were evaluated in this study. Test results indicated that the adsorption efficiency is higher in acidic conditions than alkali conditions. Adsorption efficiency was increased from 3 to 5 by pH increments and the highest adsorption efficiency was observed in the pH rate of 5. With the increases in pH from 5 to 9, the system efficiency was decreased, and finally it was stabilized in the pH rates of 5 and 9. Thus, according to the test results, the optimum pH rate for formaldehyde removal by human hair is between 4 and 5.

Researchers such as Asgari et al also demonstrated that neutral pH is the most efficient pH for adsorbing the pollutants by many different natural adsorbents [7]. It is worth mentioning that there is no possibility of proper comparison between similar studies that have been carried out using other types of adsorbents and pollutant materials. That is, because in most of these experiments, the adsorption mechanisms (physical or chemical) were not evaluated. Thus, the comparison of adsorption efficiency with shifts in pH, temperature, etc, is not a correct action without knowledge of the adsorption mechanism.

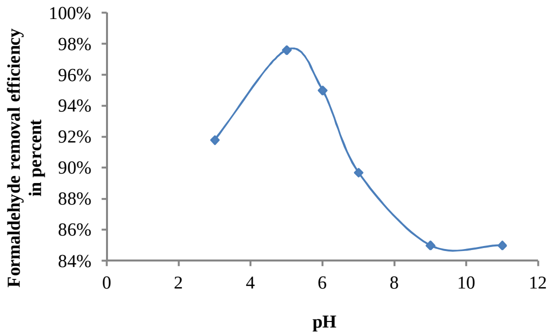

Figure.3: Formaldehyde adsorption shifts on human hair based on pH

3.4 The effects of formaldehyde concentration in adsorption efficiency

Changes in formaldehyde concentration in stable amounts of hair can affect the adsorption process. As it is shown in fig.4, by increasing the formaldehyde concentration, the adsorption rate is also increased. This increase is entirely logarithmic and follows the equation 2 with a correlation coefficient of 0.999.

$$y = 0.0131\ln(x) + 0.956 \qquad (2)$$

In the equation 2, y is the formaldehyde adsorption efficiency in percent, and x is the formaldehyde concentration rate in mg/l. In similar conditions used in this study, this equation is applicable for prediction of formaldehyde adsorption rate. This equation can be used for evaluating the formaldehyde adsorption efficiency in different concentrations by interpolation and extrapolation. This equation was used to provide the required information in the modeling section using Langmuir equation.

3.5 The effects of different hair quantities in formaldehyde adsorption efficiency

In this study, the quantities between 0.5 to 2 g of hair were applied for treatment of synthetic wastewater contaminated with 27000 mg/l of formaldehyde. As it could be deduced from figure.5, changes of hair quantities in formaldehyde adsorption are not meaningful in the mentioned human hair quantity and in all conditions, shows the steady amount of 98%. Other researchers have discovered different results in their experiments. For example Samarghandy has mentioned that increases in adsorbent dose can positively affect the adsorption efficiency [12].

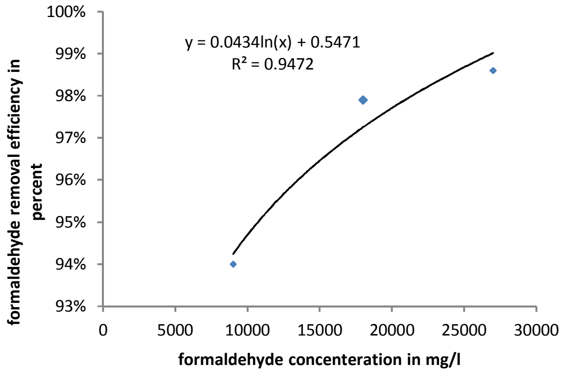

Figure.4: Formaldehyde adsorption shifts on human hair based on different concentrations

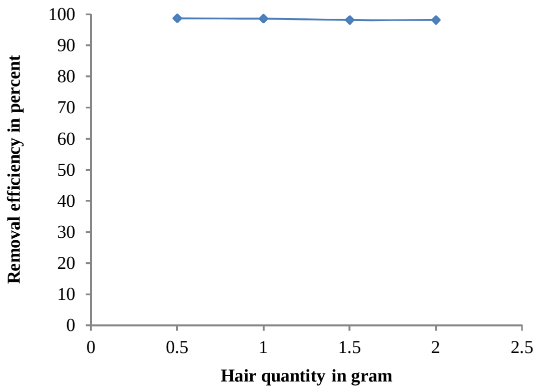

Figure.5: Changes in formaldehyde removal efficiency in different hair quantities

3.6 Evaluation of dyed hair, Henna dyed hair, white hair and sheep wool in the adsorption process

Many different chemical or natural pigments are used for dyeing human hair. Dyed hair or henna dyed hair could have different potencies in formaldehyde adsorption. It is clear that, there is no possibility for isolating the different types of hair when it is applied to remove of formaldehyde from wastewater in large scale. Thus, effect of using dyed hair was evaluated in this part of study. Moreover, in many cases, collecting and using sheep wool is much easier than human hair, therefore, limited studies were carried out to evaluate the application of sheep wool in formaldehyde adsorption. As it could be deduced from Table.1, the adsorption efficiency of sheep wool and dyed human hair are not much different from natural human hair.

Table1: Removal efficiency

Sheep wool	White hair	Henna dyed hair	Dyed hair	Factors
98.4	98.8	98.3	98.7	Formaldehyde removal efficiency

4.7 Approximate optimal conditions

The optimal conditions that were determined in these series of tests are as following: temperature of 60 C, retention time of 5 minutes, pH equal to 5, concentration of 27000 mg per liter and 0.5 g of hair for each 100 ml of wastewater. It is worth mentioning that the method used to determine the optimal conditions is the one factor at a time. But this method can somewhat contain errors and ignore the different levels of various factors. It is recommended that in further studies on formaldehyde adsorption by human hair, the optimization process should be carried out using experimental design methods to investigate the interaction effects between different factors as well.

3.8 Modeling formaldehyde adsorption on human hair

In this study, formaldehyde adsorption on human hair modeling was designed based on Langmuir isotherm. For this purpose, formaldehyde adsorption was conducted on six concentrations mentioned in fig. 6 using predictions made by equation.1 and the results were used in the modeling. The result of the modeling was the determination of equation constants which could be used to the design this system in industrial scale. The modeling was carried out using equation 3.

$$\frac{\mathrm{c}}{\mathrm{f}} = \left(\frac{1}{\mathrm{f_{max}}}\right) c + \frac{1}{\mathrm{k} \times \mathrm{f_{max}}} \qquad (3)$$

In the above equation, *c* is the initial formaldehyde concentration in mg/l, *f* is adsorbed formaldehyde in each formaldehyde concentration in mg/l, *k* is Langmuir constant coefficient in l/mg and eventually, f_{max} is the maximum of adsorbed formaldehyde in each formaldehyde concentration in mg/l. The constant coefficients of *k* and f_{max} could be calculated versus *c*. The resulting regression is a linear equation in which *a* is the slope and *b* is the interception. The result of the regression between *f* versus *c* is shown in fig. 6.

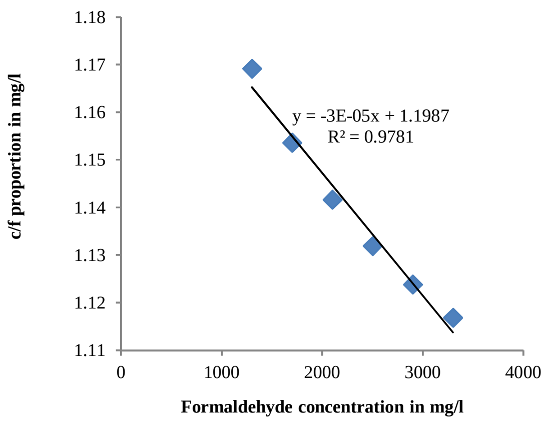

Figure.6: The regression result between formaldehyde concentration and Langmuir constant coefficient

As indicated in this figure, the result of the regression is the following linear equation.

$$y = -3 \times 10^{-5} x + 1.198$$

The slope (a) in this equation is equal to 3×10^{-5} and the interception (b) is equal to 198.1. Thus, based on equation.2, $1/f_{max}$ is the slope which is equal to 3×10^{-5} and $\frac{1}{k} \times f_{max}$ is the interception which is equal to 1.198. Therefore, the calculation of constant coefficients of k and f_{max} is equal to 2000 l/mg and 4.173 mg/l, respectively.

4 Conclusions

In this study, human hair, as a low-cost natural sorbent, was applied to remove formaldehyde from the wastewater. It was indicated that human hair is a suitable and efficient adsorbent for decreasing formaldehyde concentration in the environment. This natural adsorbent provides 99% formaldehyde removal in a slight retention time of 5 minutes. In this study other types of human hair (dyed, Henna dyed and white hair) and also sheep wool were studied to remove formaldehyde from aquatic environments. The test results indicated that the adsorption efficiency in all cases is identical to human hair. This adsorbent requires no initial preparations (unlike activated carbon production process) for adsorption purposes. The usage possibility of low-cost adsorbents in waste treatment could be an incentive for industries to prevent the release of harmful pollutants, such as formaldehyde in the environment by taking advantage of such cheap technologies.

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