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REMOVAL OF PARACETAMOL FROM AQUEOUS SOLUTION BY DRIED CELLULOSE AND ACTIVATED CARBON

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ABSTRACT

Paracetamol is favorable medicine which used by human. But if it is taken beyond the optimum limit, it can put that particular person be in danger. It also can cause effect to aquatic life if it is excessive in the aqueous solution. Therefore, it is important to remove paracetamol from aqueous solution to avoid any effect whether to the humans or even to the non-humans. The aim of this study is to compare the efficiency for removing paracetamol by using activated carbon and dried cellulose. The removal of paracetamol from the aqueous solution was investigated by using effect contact time and initial concentration. The treatment that was used in this experiment was adsorption batch method and the result was analyzed by using ultraviolet (UV) Visible Spectrophotometer. The percentage removal of paracetamol from aqueous solution by activated carbon is 94.5%, while dried cellulose is 58.1% with effect of the initial concentration. As for effect of contact time, the result of activated carbon is 98.6%; meanwhile dried cellulose is also 68.6%. The adsorption isotherm that was analyzed in this study is Langmuir model. It has indicated that percentage removal of paracetamol by using activated carbon is obeying the model, whereas unmodified dried cellulose does not.

Keywords: pharmaceutical, adsorption isotherm, langmuir isotherm model, isotherm model.

INTRODUCTION

Paracetamol is widely used for pain reliever and fever reducer, especially for humans and animals. Paracetamol is safe for use at suggested doses by the doctor or the specialist. But if somebody takes an overdose than the recommended dose, it will be harmful for that particular person or animal. This is similar to the aqueous solution such as rivers and lakes due to an overdose of paracetamol from the industrial waste matter from many sources into the river. It can lead to one of the environmental problems that made a big concern for everyone in the world. Human (disposal of unused medications, excretion of medicines or their metabolites that are not absorbed by the human body via the toilet), industry (pharmaceutical manufacturing residues), and agriculture (veterinary and aquaculture drugs, feed additives in livestock breeding via manure spread on land as fertilizer from which it can leach into local streams and rivers) are the example of different sources that causes the pharmaceuticals occurrence in the environment. Due to these occurrences that occur, much aquatic life and the environment had been affected badly and the worse part can lead to death. Lakes and rivers that are common sources of drinking water suppliers will be contaminated if the treatment is not effective. The substances of pharmaceutical origin are not biodegradable and often are not completely eliminated when by using conventional waste water treatment [1].

This serious environmental problem is due to the lack of appropriate treatment system that should be done to stop introducing medicines into diverse aquatic ecosystems [2]. Even though the removal of paracetamol treatment need a lot money, the particular industry must treat their wastewater properly [3]. There are many analyses for removing paracetamol from aqueous solution to determine possible treatment such as reverse osmosis, solvent extraction, neutralization and adsorption [4]. Adsorption has become one of the effective methods due to its simplicity of design, effectiveness and cost. It is also widely used in industrial application [5]. Activated carbon is one of the adsorbate used to remove paracetamol from aqueous solution in the industry. It is the highest efficient type of adsorbate for the removal along with dried cellulose. In addition, activated carbon can reach almost 100% removal efficiency in some application due to its high adsorption capacity as well as removal efficiency for certain organic substances [3]. Other than activated carbon, dried cellulose also one of the adsorbate to remove paracetamol from aqueous solution. It is cheaper and more effective adsorbent [4]. Cellulose is a popular substrate with adsorptive membranes with hydrophilic surfaces. It also have reactive hydroxyl groups with loss of rigidity and pore size changed [6]. It is also very environmentally benign nature and has attracted the attention due to its effectiveness [7].

In this study, commercial dried cellulose and activated carbon are used as the adsorbents to remove paracetamol from aqueous solution. It helps to determine the efficiency of unmodified dried cellulose and activated carbon to remove paracetamol from aqueous solution. It also will helpful for further study in producing appropriate biomaterial to remove paracetamol by using batch adsorption. The advantages of adsorption that exceed other method are the simple design and it can involve low investment of both lands required and initial cost [8]. Furthermore, new low cost adsorbent materials still arises a great with high adsorption capacity [9]. For this study, initial concentration and contact time were used as parameter to remove paracetamol form aqueous media.

MATERIALS AND METHOD

Materials

Materials used in this study are paracetamol, dried cellulose and activated carbon which is already in the laboratory. Dried cellulose and paracetamol are from Acros organics while activated carbon is from R&M chemicals.

Batch adsorption studies

Paracetamol solution as adsorbate was prepared in a beaker. Measure the pH of the solution by adding either 1M of hydrochloric acid (HCl) or 1M of sodium hydroxide (NaOH) for pH adjustment by using a pH meter and constant it at 3. Next, weight the amount of adsorbent which is dried cellulose activated carbon to 0.1g and put it inside a conical flask that has paracetamol solution ready in the flask. After that, the solution was put inside the incubator shaker at a controlled temperature of 30°C, the agitation time of 150rpm and several samples were collected at the desired contact time which is 2 hours. After it has collected, the solution is then filtered by using the filter paper and solution that has been filtered is analyzed by UV-Visible Spectrophotomer Varian 50.

Effect of initial concentration

In this parameter, adsorbent dosage is kept constant at 1g/L. The solutions of 1M of HCl and 1M of NaOH were used to achieve the pH adjustment of 3. About 100mL of paracetamol was prepared in the beaker. Next, the solution was stirred at 0.4ppm of initial concentration at 30°C and 150rpm agitation speed for 2 hours. The experiment then was repeated with the different initial concentration of adsorbate which is 0.6, 0.8, 1.0, 1.2 and 1.4ppm. After that, the solution was filtered to separate the suspended solid in the solution. Lastly, the product was tested with UV-Visible Spectrophotometer at 245nm.

Effect of contact time

For this study, about 1g/L of adsorbent dosage and 100ppm of initial concentration of adsorbate was constant. The mixture was stirred at 150rpm of 30°C for different contact time which is 10, 20, 30, 40, 90, 120, 180, 240 minutes. Lastly, the product was analyzed for paracetamol concentration by using UV Visible Spectrophotometer at 245nm.

he adsorption percentage (removal (%)) of paracetamols from aqueous solution is computed as follows:

$$\text{Percentage of removal } (\%) = \left(\frac{C_0 - C_n}{C_0}\right) x100\ \% \qquad (1)$$

where C_0 is initial concentration of paracetamol solution and C_n is the final concentration of paracetamol solution.

RESULTS AND DISCUSSIONS

Effect of initial concentration

Based on the Figure-1, the red line indicates percent removal of paracetamol by activated carbon while the blue line is dried cellulose. As for the activated carbon adsorbent, the percentage removal for 1.4ppm concentration of paracetamol is the highest that is 94.5% which is a lot of the concentration is being removed, while for 0.4ppm concentration of paracetamol give lowest percentage removal which is 55.6%. As for the dried cellulose adsorbent which is the blue line, the percentage removal for 1.4ppm concentration of paracetamol is the highest that is 58.1%, which has only half of the concentration is removed, while for 0.4ppm concentration of paracetamol give lowest percentage removal which is 38.01%. There is a significant increasing shown in Figure-1 [10]. It indicates that as initial concentration is increasing, the percent removal of paracetamol of dried cellulose also increasing as same as the activated carbon. But, the difference between these two adsorbents is that the percent removal of paracetamol solution by activated carbon give a higher percent removal than dried cellulose. The reason for a low efficiency of the dried cellulose to remove paracetamol is where it is not modified yet to become modified cellulose, which will give a higher percent removal of paracetamol from aqueous solution. The two adsorbent shows the same flow of graph which is increasing. The more initial concentration the higher the percentage removal of adsorbate from aqueous solution. This obtained result is in line with the result reported by the other researchers [10].

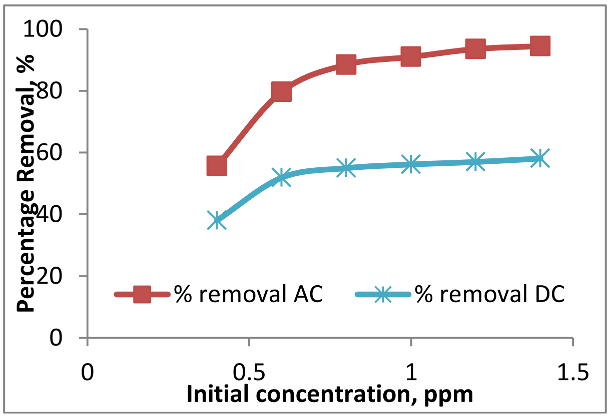

Figure-1. Effect of initial concentration on percentage removal of paracetamol solution concentration.

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Effect of contact time

To establish the equilibrium time for maximum uptake and to know the kinetics of the sorption process, the sorption of paracetamol solution by two adsorbents which is activated carbon and dried cellulose was carried out by using contact times which ranging from 10 to 240 min [11]. Figure-2 shows that green line is percentage removal paracetamol by activated carbon, while the blue line is indicated percentage removal of paracetamol by dried cellulose. For adsorbent of activated carbon which is the green line, the effect of contact time is increasing in terms of percent removal of paracetamol solution from aqueous solution. But, it is not as smooth as the percent removal of paracetamol by activated carbon adsorbent on the effect of the initial concentration. Generally, this study showed that percentage removal of paracetamol solution concentration time by activated carbon is slightly increased as contact time increased for both types of adsorbent [10]. It shows that the percentage removal of paracetamol is lower compared to activated carbon. However, this result agreed with the result obtained from [2]. These researchers reported that sugar cane based which is contained cellulose has 60% efficiency of adsorbent. Furthermore, unmodified cellulose was used as an adsorbent in this study. The efficiency can be enhanced if the modified cellulose is used [12].

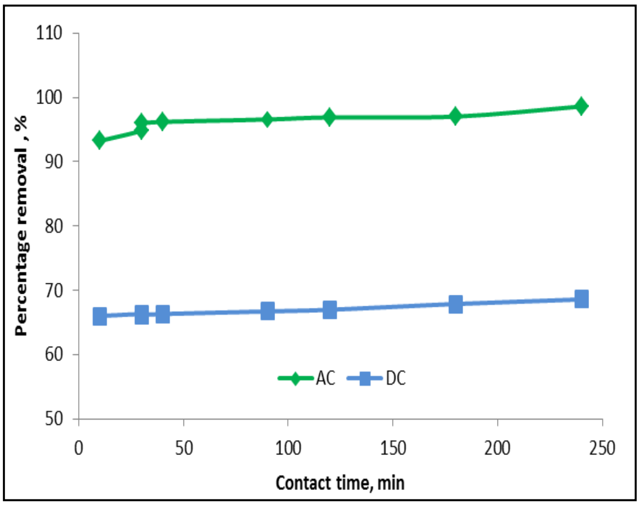

Figure-2. Effect of contact time on percentage removal of paracetamol solution concentration.

Adsorption isotherm

Adsorption isotherm describes the equilibrium of the sorption of a material on a surface [13]. The function of the material present in the gas phase and/or in the solution is the amount of material bound on the surface (the sorbate) which is represented by adsorption isotherm. To obtain the sorption capacity, the amount of paracetamol sorbed per mass unit of activated carbon/dried cellulose (qe) is evaluated by using the following expression:

$$q_e = (c_0 - c_e)\ v/m \qquad (2)$$

where C_0 is the initial metal ion concentration ($mg.L^{-1}$), C_e is the equilibrium metal ion concentration ($mg.L^{-1}$), V is the volume of the aqueous phase (L) and M is the weight of date tree leaves used (g).

The Langmuir isotherm is based on the monolayer sorption of paracetamol on the surface activated carbon or dried cellulose sites [11]. It is represented by the following equation where q_{max} and K_L are the Langmuir constants which related to sorption capacity and the energy of sorption:

$$C_e/q_e = 1/q_{max}K_L + 1/q_{max}\ (C_e) \qquad (3)$$

where q_e is the amount adsorbed at equilibrium, Ce is the equilibrium concentration and q_{max} and K_L are the Langmuir constants related to sorption capacity and the energy of sorption

Adsorption is usually describes by using isotherm. Adsorption isotherm describes the equilibrium of the sorption of a material on a surface [13]. The only isotherm that will be discussed in this study is Langmuir model. The graph of Langmuir model is made based on the effect of initial concentration by activated carbon and dried cellulose. Langmuir constants which are q_{max} and K_L that related to adsorption capacity and the energy of sorption that can be calculated based on Equation. (3) in the section above.

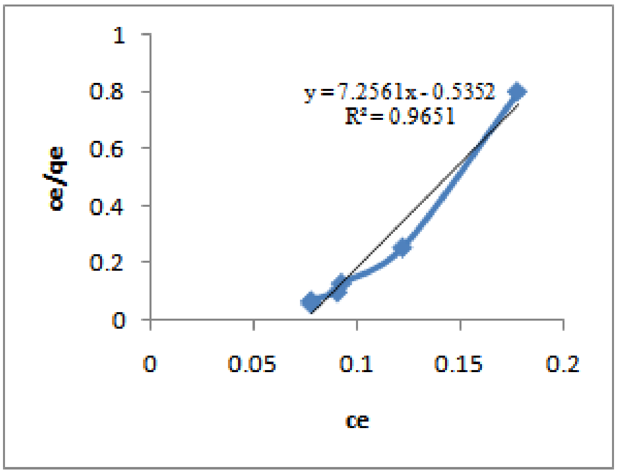

Figure-3. Adsorption isotherms on effect of initial concentration of paracetamol removal by activated carbon.

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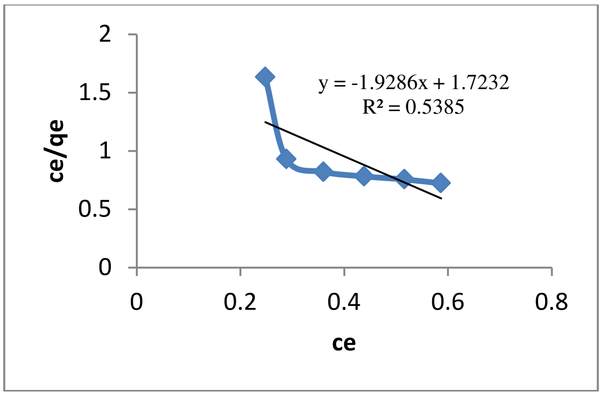

Figure-4. Adsorption isotherms on effect of initial concentration of paracetamol removal by dried cellulose.

Figure-3 shows a graph of Langmuir model on the effect of initial concentration of paracetamol removal by activated carbon. It indicates an increased linear where the value of R^2 is high which is 0.9651, so it is obedient and favorable the Langmuir model [5]. As for the Figure-4 which is the graph of the Langmuir model on the effect of initial concentration of paracetamol removal by dried cellulose, it shows a decreasing linearly with the R^2 is low which is 0.5385. Therefore, it does not obey the Langmuir model. It might obey the other model of adsorption isotherms such as Freundlich or Tempkin. For Figure-3, the value of K_L which is Langmuir constant is -13.54 while q_{max} is 0.138. As for Figure-4, the K_L is -0.893 while q_{max} is -0.519.

CONCLUSIONS

Based on the percentage removal of paracetamol between two adsorbents, activated carbon shows the highest removal compared to unmodified dried cellulose in two parameters which is effected of initial concentration and contact time. The adsorption isotherm in this study showed that the activated carbon is favorable the Langmuir model, but dried cellulose is not favorable the Langmuir model where it might obey other isotherms such as Freundlich or Tempkin. Thus, it can be concluded that activated carbon is more efficient to remove paracetamol than unmodified dried cellulose in aqueous solution.

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