UTILIZATION OF EGGSHELL AND ACTIVATED CARBON AS ADSORBENTS FOR REMOVAL SALICYLIC ACID FROM AQUEOUS SOLUTION

Zainab A. Hassan¹ and Marwa F. Abdul Jabbar¹,*

¹Chemical Engineering Department, Al-Nahrain University, Baghdad, Iraq.

*Corresponding author email: marwa84_2007@yahoo.com

ABSTRACT

Adsorption process has been proven to be one of the best water treatment technologies around the world. The present study attempts to use waste material (eggshell) as adsorbent for salicylic acid removal and compare its activity with activated carbon.

Many variables were studied to indicate their influence on removal efficiency. They have been included pH of solution were (2-11), salicylic acid concentration (10-50ppm) and adsorbent concentration (10g/l – 25 g/l) for eggshell while for activated carbon (1g/l -5g/l).

From the experimental results, there were noticed that the percentage removal increased with increasing adsorbent concentration, acidic solution while decreased with increasing salicylic acid concentration. Also, the results indicated that eggshell gave lower removal compared with activated carbon. Therefore the best conditions are pH solution was 2, salicylic acid concentration 10ppm, activated carbon amount 5g/l while for eggshell 25g/l that achieved percent removal reached 97 % and 24% for activated carbon and eggshell respectively.

KEYWORDS: Adsorption, Activated carbon, Eggshell, Salicylic acid, Waste as adsorbent
1. INTRODUCTION

Water pollution is one of many types of pollution which results from contaminants being come in the natural environment. The discharge of inadequately treated wastewater into natural bodies of water caused water pollution. Developments in medicine have resulted in an increasing use of pharmaceuticals, during drug treatment, the active components of pharmaceutical products are broadly excreted from the patient’s body, either unchanged (as the same compounds that are existing in the dose form) or as derivatives or metabolites of these compounds. Excretion from patients that way introduces pharmaceutical residues into the sewage disposal system and thus into wastewater. Another pathway whereby pharmaceutical residues can be disposal into the environment is through the improper disposal of unused or expired pharmaceuticals into the wastewater system or landfill (Kirstie, 2015).

The compounds that contain phenolic group are significant water contaminants. Their presence, even at low concentrations in water could be an obstacle to the use and the reuse of this resource. These pollutants appear in the water as a consequence of degradation of the phenolic compounds which are used in the synthesis of pesticides, insecticides, drugs, etc. (Sathishumar et.al, 2007 and Hussain et.al, 2009).

Among the pharmaceutical pollutants, salicylic acid (SA) is most considerably found in hospital waste and pharmaceutical industry effluents. It is aromatic organic compounds which becomes harmful to human beings beyond their threshold concentration level. SA is an anti-inflammatory drug generally employed to treat acne, wart and fungus infections. However, while exposure to high doses, it can give rise to severe health problems like salicylate poisoning, nausea, delirium, coma, severe stomach ache, gastric and death. The existence of SA in water resources is thus a significant concern, and its removal from industrial effluent is acute to safeguard human health and the environment (Xin et.al, 2015).

Numerous techniques have been used for treating water media contaminated with aromatic organic acids such as reverse osmosis, ion exchange, distillation, liquid extraction, bipolar membrane electrodialysis and reactive distillation. These methods are not effective when treating dilute solutions. Adsorption is a simple method and has been used for removal of organic carboxylic acids (Yan et.al, 2007, Yildiz et.al, 2005).

Different adsorbents such as activated carbon, natural zeolites, and clay are used for adsorption of salicylic acid. Activated carbons have a large adsorption capacity for a variety of organic pollutants, generally the adsorption capacity of activated carbon is concerning to the surface area, the pore structure, and the surface chemistry. In the liquid phase adsorption, the
capacity of activated carbon to uptake the aromatic compounds depends on some factors, these include the essential properties of the adsorbent such as (pore structure, functional group and ash content), the essential properties of the solute (polarity, solubility, molecular size and functional groups), the solution properties such as pH and initial concentration of the solute (Khenniche et al., 2009). In view of the high cost and tedious procedure for the preparation and regeneration of activated carbon, there is a continued search for the development of adsorbents using cheaper raw materials.

Egg shells are waste materials from hatcheries, households and fast food industries and can be readily collected in plenty. However, these egg shells can be used as an excellent adsorbent because of their porous nature; about 700 to 1,700 pores are present on the surface an egg shell. The egg shells are also attractive as adsorbent due to their good mechanical properties and thermal stability and already used as adsorbent for the removal of heavy metals, phenolic compounds, dyes, and pesticides (Ikram et al., 2016).

The Aim of present work using chemical adsorbent (activated carbon) and natural material (Egg shell) to test their activity for treatment water containing salicylic acid by study the effect of adsorbent concentration, adsorbate concentration and pH of solution on adsorption process.

2. EXPERIMENTAL WORK

2.1 Materials

Salicylic acid was purchased from pharmacy, sodium hydroxide and sulfuric acid for adjust pH solution, activated carbon and eggshell used as adsorbents, ethanol as solvent for salicylic acid. Distilled water was used throughout this study.

2.2 Experimental Work

Chicken eggshells were used as the adsorbent in this study. The chicken eggshells were collected from kitchen waste and washed by deionized water for several times to remove the dirt particles. The eggshells were then dried in oven at 80°C for 4 hr. Eggshells were ground to a powder in a grinder, and sieved to obtain diameter of 75μm. For adsorption process prepare stock solution of SA with concentration (100 ppm) by dissolving (0.1 g) of SA with drops of ethanol and after that adding (1000 mL) of distilled water. In order to prepare (10 ppm, 25 ppm, 35 ppm and 50 ppm), taking amount of stock solution and diluted to require concentration. Adding the adsorbent (egg shell) with different concentration (10 g/L, 15 g/L, 20 g/L and 25 g/L) to flask of SA solution and put into shaker. Withdraw a sample at different period of time for (2 hr) and filtrate to separate the adsorbent from solution. The salicylic acid concentration
was measured by visible ultraviolet spectrophotometer and then the removal was calculated via the formula 1:

\[ X\% = \frac{C_0 - C}{C_0} \times 100 \]

Where \( C_0 \) is the initial concentration of salicylic acid and \( C \) is the concentration at a specified time. Repeat the same procedure above for using activated carbon as adsorbent with amount (1 g/L -5 g/L).

3. RESULTS AND DISCUSSION

3.1 Effect of pH

The pH value of a solution is a major controlling parameter in the adsorption process because it affects the surface charge of adsorbent as well as degree of ionization of adsorbate. The effect of the pH was studied in the interval 2-11 with initial concentration of 50 ppm of salicylic acid. The results obtained are presented on Fig. 1 and Fig. 2 for activated carbon and eggshell respectively.

The results show that best removal occur at pH =2 which was 85% and 8% by activated carbon and eggshell respectively. This can be explicated the surfaces had a positive charge when the pH value was low, this makes a strong electrostatic interaction between the adsorbate molecule and the surface of material. At higher pH, fraction of ion in solution increases, the relative positive charge on adsorbent decreases, and hence, electrostatic force of attraction weakens. The surface becomes negative and exhibits repulsion to SA ions (Raoul et.al, 2015).

![Fig. 1. Influence of pH on the adsorption, activated carbon 5g/l.](image-url)
3.2 Effect of contact time and initial concentration of adsorbate

The effect of contact time on removal salicylic acid by activated carbon and eggshell can be seen in Fig. 3 and Fig. 4 respectively. Experimental studies were carried out with varying initial concentration from 10 to 50 ppm using 5 g/l and 10 g/l of adsorbent dose for activated carbon and eggshell respectively at pH= 2. Equilibrium established within 30 min for activated carbon while for eggshell there are no equilibrium time till 120 min since it may be need more time because not activated. This behavior agrees with (Pathak et. al, 2015) who used banana peel and need 14 h to reach equilibrium for removal salicylic acid.

From these figures also can be seen influence of different concentration of salicylic acid. The amount of adsorbate adsorbed per unit mass of adsorbent increased with the decrease in initial concentration. The percent adsorption using activated carbon and eggshell were reduced from 97% to 84% (30 min) and 23% to 5% (120 min) for concentration increased from 10 to 50 ppm respectively. These results may be explained by the fact that, at low adsorbate concentration, the ratio of surface active sites is high hence salicylic acid ions could interact with sorbent to occupy the active sites and can be removed from solution (Pathak et. al, 2015).
3.3 Effect of Adsorbent Dose

To estimate the influence of mass of adsorbing material on the adsorption process, different masses of the activated carbon and eggshell were used varying between 1-5g/l and 10-25g/l for activated carbon and eggshell respectively. Initial concentration of adsorbate was kept fixed at 25ppm and pH = 2. The results are presented in Fig. 5 and Fig. 6.

They show that the percentage of elimination increased with the mass of adsorbent but with activated carbon the difference very low this may be the amounts used were very high therefore gave high removal, it increase from 92% to 97% as amount increase from 1g to 5g after 30min. While for eggshell it increase from 11% to 24% as amount of adsorbent increase from 10g to 25g after 120min. Although amount of activated carbon was low but gave higher removal compared with eggshell this due to active site of it. This can be explained by the fact that by increasing the mass of adsorbent, there is an increase in the number of adsorption sites on the surface of material (Abd Ali et.al, 2016).

3.4 Comparison between activated carbon and eggshell

After experiment were done, the best results for two types of adsorbent were shown in Fig. 7. It can be seen although concentration of eggshell was high but it gave low removal compared to activated carbon. The maximum removal approximately 24% at maximum amount 25g while for activated carbon reach 97% although its amount was little (5g) at the same conditions of salicylic acid concentration 25ppm and time 120min.
Fig. 5. Influence of activated carbon concentration on the adsorption, SA concentration 25ppm, pH=2.

Fig. 6. Influence of eggshell concentration on the adsorption, SA concentration 25ppm, pH=2.

Fig. 7. Comparison between activated carbon and eggshell.
4. CONCLUSIONS

1. The maximum removal obtained from experiments was 97% that achieved by used activated carbon as adsorbent with amount 5g, salicylic acid concentration 10ppm and pH=2.

2. Performance of eggshell as adsorbent compared to activated carbon was low although its amount was fifth order. Since it gave 16% removal (25g eggshell) while this percent increase to reach 97% removal (5g activated carbon) after 60min.

3. The percent removal increases as Salicylic acid concentration decreases.

4. As amount of adsorbents increase the removal increase as a result of more available active site.

5. REFERENCES

Abd Ali Z.T., M. A. Ibrahim and H. M. Madhloom, 2016, “Egg Shell Powder as an Adsorbent for Removal of Cu (II) and Cd (II) from Aqueous Solution: Equilibrium, Kinetic and Thermodynamic Studies”, Al-Nahrain University, College of Engineering Journal (NUCEJ), 91(2).

Hussain A., T. Parveen, P. Kumar and I. Mehrotra, 2009, Desalination Water Treat, 2, 254–259.

Ilkram M., A. Rehman, S. Ali, S. Ali, S. U. Bakhtiar and S. Alam, 2016, “The Adsorptive Potential of Chicken Egg Shells for the Removal of Oxalic Acid from Waste Water”, Journal of Biomedical Engineering and Informatics, 2(2).

Khenniche L. and A. Farida, 2009, “Characterization and Utilization of Activated Carbons Prepared from Coffee Residue for Adsorptive Removal of Salicylic Acid and Phenol: Kinetic and Isotherm Study”, Desalination and water treatment, 11, 192-203.

Kirstie M., 2015, Pharmaceutical Pollution in the Environment: Issues for Australia, new Zealand and pacific island countries.

Pathak P. D., S. A. Mandalganea and B.D. Kulkarni, 2015, “Utilization of Banana Peel for the Removal of Benzoic and Salicylic acid from Aqueous Solutions and its Potential Reuse”, Desalination and Water Treatment, 11,,1-13

Raoul D., A. S. Gabche, N. G. Ndifor, K. J. Mbadcam, T.D. Raoul, A. S. Gabche, N. G. Ndifor and J. M. Ketcha, 2015, “Adsorption of Salicylic Acid and SulfoSalicylic Acid onto Powdered Activated Carbon Prepared from Rice and Coffee Husks”, International Journal of Current Engineering and Technology, 5(3).
Sathishkumar M., A.R. Binupriya, D. Kavitha and S.E. Yun, 2007, Bioresour Technol., 98(4), 866–873.

Xin J. L., N. Chemmangattuvalappil and L. Y. Lee., 2015, ‘Adsorptive Removal of Salicylic Acid from Aqueous Solutions using New Graphene-Based Nanosorbents”, chemical engineering transactions, 45.

Yan L. G., J. Wang, H. Q. Yu, Q. Wei, B. Du and X. Q. Shan., 2007, “Adsorption of Benzoic Acid by CTAB Exchanged Montmorillonite”, Appl. Clay Sci, 37, 226-230.

Yildiz N., R. Gonulsena, H. Koyuncu and A. Calimli., 2005, “Adsorption of Benzoic Acid and Hydroquinone by Organically Modified Bentonites”, Colloids Surf. A, 260, 87-94.