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Comparison study for different adsorbents used to remove paracetamol from water sources

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***Abstract* –** The coronavirus disease 2019 (COVID-19) outbreak has induced an unprecedented increase in the use of several drugs, especially paracetamol, because, for COVID-19 symptoms, paracetamol is recommended as the first-line antipyretic and analgesic medication. The excretion of drugs and their metabolites in stools and urine release these drugs into wastewater, surface waters, and groundwater. This study intends to examine the adsorption of paracetamol, one of the most used medications used during the COVID-19 pandemic, from water using different adsorbents, commercial activated carbon (granular and powder form), clay, silica gel, and zeolite. The experiments were conducted under constant conditions at room temperature 27^{o}C, initial concentration of 10 ppm, speed of rotation is 200 rpm, the mass of adsorbents is 1 gm, pH 7.8, and volume of solution is 200 ml under different time intervals to study the effect of each adsorbent in the removal process. As a result of this work, activated carbon was the best adsorbent to remove paracetamol from water in both granular and powder forms, which gives the removal of 98.5% in 5 hours for granular and 93.33% in 1 hour for powder activated carbon. It is essential to improve the characteristics of activated carbon to reduce the process's time to make it more economical.

Keywords – Adsorption, Paracetamol, COVID-19, Activated Carbon, Wastewater Treatment.

1.0 Introduction

The four categories of drinking water, groundwater, surface water, and seawater are often used recently to categorize water resources. Humans have long been drawn to water, but increasingly, water scarcity has become a significant problem in all facets of human life [1]. Recent years have seen an increase in the detection of chemicals such as pesticides, food additives, pharmaceuticals and other contaminants in the water thanks to the development of new and more sensitive analytical techniques. The wastewater from homes and businesses, waste from treatment facilities, hospital effluents, and other sources all contribute to the environment's growing pollution problem. Because they have an impact on human health and animals, eliminating these contaminants has become one of the most urgent environmental issues [2].

Paracetamol is a widely used antipyretic and analgesic medicine in many nations throughout the world for the effective pain management of a variety of conditions such as headaches, fever, and minor aches [3]. Paracetamol is a widely used drug. After consumption by animals and humans, it is excreted through the urine to the sewer systems and natural aquatic environments [4] Its toxicity has been studied and is well-proven in microorganisms, algae, protozoans, and fishes. Developing technologies to remove paracetamol from natural and wastewater to avoid harmful health problems over living organisms is actual. The global consumption of paracetamol has grown. It ranks among the top 200 prescriptions written in the United States and among the top three pharmaceuticals prescribed in the United Kingdom. In the United Kingdom, around 3.2×10^9 paracetamol tablets are consumed annually, or 55 pills per person. In some industrialized countries, rates approached 20 g/person/year in the Nordic countries. Because of their high solubility and hydrophobicity, it is easily collected in aquatic environments [5].

Paracetamol is considered the first line of antipyretic and analgesia for COVID-19 symptoms. Also, because paracetamol has been suggested as a treatment for the COVID-19 pandemic's symptoms, it is anticipated that drug usage will rise significantly [6].

Many processes used to treat water sources from paracetamol can be classified as physiochemical processes. There have sand filtrations [7], Membrane separation [8], Fenton and Fenton-based processes [4], Advanced oxidation processes (AOPs) [9], Ozonation [10], and biological processes such as Anaerobic treatment [11], Fungal treatment [12], Membrane bioreactor (MBR) [13], and Bacterial treatment [14]. However, the most common process used is adsorption because it provides high efficiency and economical method [15], [16], [17], and [18].

This study focuses on choosing a suitable adsorbent to remove paracetamol from water to make the process more effective.

2.0 Experiment

2.1 Materials and Method

Paracetamol (acetaminophen) with assay 98% - 102% supplied by SIGMA-ALDRICH, U.K., Commercial granular activated carbon provided by R&M Marketing, Essex, U.K with particle size 1.5-10 mm., Commercial powder activated carbon supplied by SIGMA-ALDRICH, U.K., Clay (bentonite) produced by SIGMA-ALDRICH, U.K., Silica gel made by R&M Marketing, Essex, U.K. and Zeolite provided by SIGMA-ALDRICH, Germany.

2.2 Adsorption process

First, a paracetamol solution with a concentration is 10 ppm was prepared by adding 0.01 g of paracetamol to 1 L of distilled water with mixing for 1 hour. Then 200 ml was taken from this solution as a sample. The experimental conditions were measured, giving the temperature a lab temperature of 27 °C and the pH value of after adding paracetamol in distilled water 7.8. Then stirring was used with a rotation speed of 200 rpm to mix the solution for 30 minutes to make a homogenous solution. After that, the initial concentration was measured by UV-VIS and recorded before starting the adsorption process. Then 1 gm of adsorbent was added to adsorb paracetamol from water under stable conditions, and the samples were taken to check the concentration of paracetamol in solution every 10 minutes for the first hour; after one hour, the samples were taken each 30 minutes and measured the concentration in the UV-VIS device.

The effect of four paracetamol adsorption materials (adsorbents) from the water was studied (activated carbon powder and granular, Silica gel, Bentonite, and Zeolite), and the same procedures were repeated for each adsorbent. Before starting the experiment, 10 ppm of paracetamol solution was prepared by adding 0.01g of paracetamol in 1 L distilled water to determine the maximum wavelength for paracetamol in UV-VIS. The maximum wavelength determined is 243.3 nm; it was too close to the theoretical value is 243 nm.

Next, five different concentration solutions were prepared to determine the calibration curve of the adsorption process by diluting the first solution with a concentration of 10 ppm by using the following formula:

$$M_1 \times V_1 = M_2 \times V_2 \quad (1)$$

The solutions have concentrations of 2 ppm, 4 ppm, 6 ppm, 8 ppm, and 10 ppm. The calibration curve was determined for wavelength is 243.3 nm and the value of R^2 is 0.998.

After those four adsorbents were experimented with to remove paracetamol from water under constant conditions as the above procedures.

3.0 Results and discussion

In this study, the effect of different adsorbents which studied under stable conditions which initial concentration = 10 ppm, temperature = at room Temp around 27 °C, the mass of adsorbents = 1 gm, the volume of solution = 200 ml, speed of rotation = 200 rpm and under natural pH = 7.8 under different periods. Different results are obtained as the following: -

3.1 Commercial Granular Activated Carbon

When used granular activated carbon with particle size (1.5 mm – 10 mm) adsorb paracetamol for 5 hours, the following results were found: -

Table 1: Commercial Granular Activated Carbon

Number	Time (Min.)	Concentration(ppm)	Adsorbate
1	0	9.847 Initial Concentration	0.582
2	10	8.250	0.488
3	20	7.259	0.429
4	30	6.054	0.358
5	40	5.253	0.310
6	50	4.730	0.280
7	60	3.967	0.234
8	90	2.546	0.150
9	120	1.882	0.111
10	150	0.965	0.057
11	180	0.611	0.036
12	210	0.506	0.030
13	240	0.269	0.016
14	270	0.184	0.011
15	300	0.148	0.009

Table 1 and Figure 1 show the findings of the adsorption process using commercial granular activated carbon. The adsorption process is quick at the beginning of the experiment. In the first hour, the removal percentage was 59.7%, reaching 80.88% in the second hour. The removal rate becomes slower after that because the activated carbon surface, more precisely its carbon outer layer, which has numerous vacant sites, gradually occupies over time as a result of the adsorption process [19]. After 2 hours of adsorption, there is no substantial increment concerning the removal of the drug due to the absence of free adsorption sites. The free spaces are occupied by paracetamol molecules, leading to no further rise in adsorption even with the increase in paracetamol quantity, so after 2 hours slight increase in removal percentage [18].

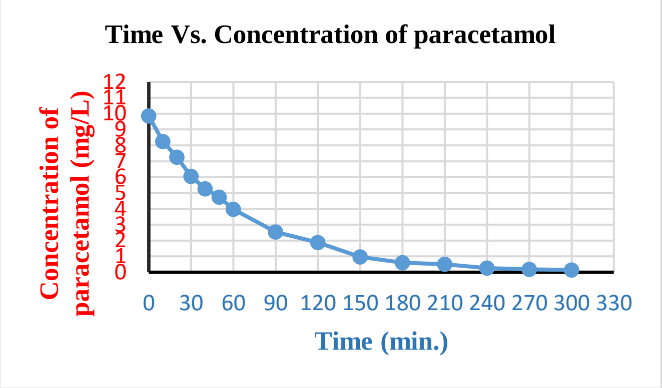

Figure 1: The removal of paracetamol by Commercial granular Activated Carbon

3.2 Commercial Powder Activated Carbon

The experiment used for 1 hr by using a vacuum pump and filter paper in the separation process, and the following results were found: -

Table 2: Commercial Powder Activated Carbon

Number	Time (Min.)	Concentration(ppm)	Adsorbate
1	0	10.633 Initial Concentration	0.629
2	10	1.642	0.097
3	20	1.291	0.076
4	30	0.949	0.056
5	40	0.953	0.056
6	50	0.914	0.054
7	60	0.709	0.042

Table 2 and Figure 2 show the findings of the adsorption process using commercial powder-activated carbon. Significant removal of paracetamol was observed in the first 10 minutes with 84.55%, and the total removal of paracetamol was 93.33% for 1 hour of the adsorption process. From the findings, powder-activated carbon has a significant affinity for paracetamol in solution. There is no significant increase in elimination % after 30 minutes of adsorption. After 30 minutes, the equilibrium is reached.

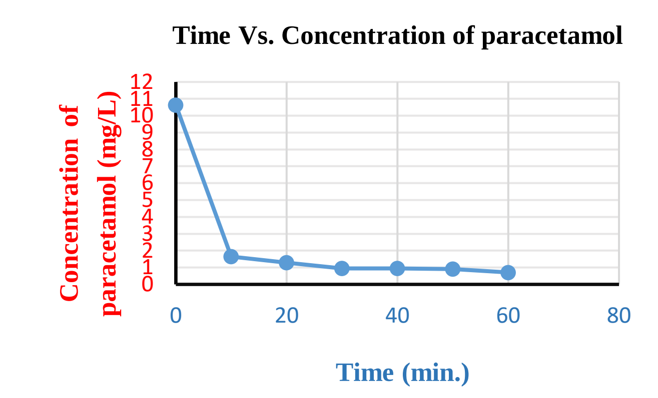

Figure 2: The removal of paracetamol by Commercial powder Activated Carbon

3.3 Clay (Bentonite)

Bentonite did not give any result because the particles were suspended in the solution. So, the experiment was repeated by using separation with a single-use membrane filter of 0.45 µg, but also the particle was still suspended. After that, the adsorption process was repeated using a vacuum pump and filter paper to separate, but the same problem continued. Also, the swelling process used bentonite for 24 hr. to avoid particle suspension, but the same problem occurred. So, bentonite is not a suitable adsorbent to adsorb paracetamol from water.

3.4 Zeolite

When zeolite is used to adsorb paracetamol from the aqueous solution, the same problem with clay is repeated because the particles are also suspended, and different methods were used to separate the particles. However, it is still suspended even after centrifugal force for 10 minutes. So, zeolite is also not a suitable adsorbent to adsorb paracetamol from water.

3.5 Silica Gel

When used silica gel was to adsorb paracetamol for 5 hours, the results were obtained as shown in Table 3.

Table 3: Silica Gel

Number	Time (Min.)	Concentration(ppm)	Adsorbate
1	0	9.854 Initial Concentration	0.582
2	10	9.779	0.578
3	20	9.449	0.559
4	30	9.721	0.575
5	40	9.964	0.589
6	50	10.161	0.601
7	60	10.273	0.607
8	90	10.307	0.609
9	120	10.274	0.607
10	150	10.059	0.595
11	180	10.360	0.612
12	240	10.631	0.626
13	300	12.058	0.713

The amount of Silica gel was found to be dissolved in water, so the concentration could not be measured correctly, and the absorbance value increased. The minimum absorption of Silica gel in water is 22%, according to the material's properties. The dissolving of silica gel in distilled water was proved by adding a small amount of silica gel in distilled water and noticing the concentration increase from 0.00 to 0.2 in two minutes.

4.0 Conclusion

This study is aimed to choose a suitable adsorbent which can be used to adsorb paracetamol from an aqueous solution. From this study, activated carbon is the best adsorbent to remove paracetamol which gives a high percentage of removal in granular and powder shapes, 98.5% in 5 hours and 93.33% in 1 hour, respectively, because activated carbon structure has porosity with different pore size and the active sites is another factor in achieving the adsorption process. So activated carbon can be further tested for improvement by increasing the surface area or porosity to reduce the treatment time.

References

[1] A. Hojjati-Najafabadi, M. Mansoorianfar, T. Liang, K. Shahin, and H. Karimi-Maleh, "A review on magnetic sensors for monitoring of hazardous pollutants in water resources," *Science of the Total Environment*, vol. 824, no. 86, p. 153844, 2022, doi: 10.1016/j.scitotenv.2022.153844.

[2] A. Spaltro *et al.*, "Removal of paracetamol from aqueous solution by activated carbon and silica. Experimental and computational study," *Journal of*

Contaminant Hydrology, vol. 236, no. July 2020, 2021, doi: 10.1016/j.jconhyd.2020.103739.

[3] H. A. E. Wadhah, "a Review on Comparative Study Between the Physicochemical and Biological Processes for Paracetamol Degradation," *Universal Journal of Pharmaceutical Research*, vol. 2, no. 2, pp. 12–21, 2017, doi: 10.22270/ujpr.v2i2.rw4.

[4] M. Pacheco-Álvarez, R. Picos Benítez, O. M. Rodríguez-Narváez, E. Brillas, and J. M. Peralta-Hernández, "A critical review on paracetamol removal from different aqueous matrices by Fenton and Fenton-based processes, and their combined methods," *Chemosphere*, vol. 303, no. May, 2022, doi: 10.1016/j.chemosphere.2022.134883.

[5] Ali Gamal Al-Kaf, Khalid Mohammed Naji, Qais Yusuf Abdullah, and Wadhah Hassan Edrees, "Occurrence of Paracetamol in Aquatic Environments and Transformation by Microorganisms: A Review Citation: Wadhah Hassan A Edrees., et al. 'Occurrence of Paracetamol in Aquatic Environments and Transformation by Microorgan-isms: A Review,'" *Chronicles of Pharmaceutical Science*, vol. 1, no. 6, pp. 341–355, 2017, [Online]. Available: https://www.scientiaricerca.com/srcops/SRCOPS-01-00033.php

[6] L. H. K. Alfhaid, "Adsorption of paracetamol in contaminated water through pH-responsive polymer-brush-grafted mesoporous silica nanoparticles," *International Journal of Environmental Analytical Chemistry*, vol. 00, no. 00, pp. 1–17, 2022, doi: 10.1080/03067319.2022.2025789.

[7] J. Li, X. Han, B. W. Brandt, Q. Zhou, L. Ciric, and L. C. Campos, "Physico-chemical and biological aspects of a serially connected lab-scale constructed wetland-stabilization tank-GAC slow sand filtration system during removal of selected PPCPs," *Chemical Engineering Journal*, vol. 369, no. March, pp. 1109–1118, 2019, doi: 10.1016/j.cej.2019.03.105.

[8] E. H. El Atmani *et al.*, "The oriented processes for extraction and recovery of paracetamol compound across different affinity polymer membranes. Parameters and mechanisms," *European Journal of Pharmaceutics and Biopharmaceutics*, vol. 126, pp. 201–210, 2018, doi: 10.1016/j.ejpb.2017.06.001.

[9] C. Yu *et al.*, "Marriage of membrane filtration and sulfate radical-advanced oxidation processes (SR-AOPs) for water purification: Current developments, challenges and prospects," *Chemical Engineering Journal*, vol. 433, no. P3, p. 133802, 2022, doi: 10.1016/j.cej.2021.133802.

[10] M. H. Abdurahman and A. Z. Abdullah, "Mechanism and reaction kinetic of hybrid ozonation-ultrasonication treatment for intensified degradation of emerging organic contaminants in water: A critical review," *Chemical Engineering and Processing - Process Intensification*, vol. 154, no. July, p. 108047, 2020, doi: 10.1016/j.cep.2020.108047.

[11] S. Zhang, Z. Lei, M. Dzakpasu, Q. Li, Y. Y. Li, and R. Chen, "Removal of trace organic contaminants in municipal wastewater by anaerobic membrane bioreactor: Efficiencies, fates and impact factors," *Journal of Water Process Engineering*, vol. 40, no. 13, p. 101953, 2021, doi: 10.1016/j.jwpe.2021.101953.

[12] D. P. de Lima *et al.*, "Fungal bioremediation of pollutant aromatic amines," *Current Opinion in Green and Sustainable Chemistry*, vol. 11, pp. 34–44, 2018, doi: 10.1016/j.cogsc.2018.03.012.

[13] W. Liu, X. Song, Z. Na, G. Li, and W. Luo, "Strategies to enhance micropollutant removal from wastewater by membrane bioreactors: Recent advances and future perspectives," *Bioresource Technology*, vol. 344, no. PB, p. 126322, 2022, doi: 10.1016/j.biortech.2021.126322.

[14] Y. Ravikumar, J. Yun, G. Zhang, H. M. Zabed, and X. Qi, "A review on constructed wetlands-based removal of pharmaceutical contaminants derived from non-point source pollution," *Environmental Technology & Innovation*, vol. 26, p. 102504, 2022, doi: 10.1016/j.eti.2022.102504.

[15] I. C. Afolabi, S. I. Popoola, and O. S. Bello, "Machine learning approach for prediction of paracetamol adsorption efficiency on chemically modified orange peel," *Spectrochimica Acta - Part A: Molecular and Biomolecular Spectroscopy*, vol. 243, p. 118769, 2020, doi: 10.1016/j.saa.2020.118769.

[16] N. Benyekkou, M. R. Ghezzar, F. Abdelmalek, and A. Addou, "Elimination of paracetamol from water by a spent coffee grounds biomaterial," *Environmental Nanotechnology, Monitoring and Management*, vol. 14, no. April, p. 100396, 2020, doi: 10.1016/j.enmm.2020.100396.

[17] M. R. Elamin, B. Y. Abdulkhair, F. K. Algethami, and L. Khezami, "Linear and nonlinear investigations for the adsorption of paracetamol and metformin from water on acid-treated clay," *Scientific Reports*, vol. 11, no. 1, pp. 1–13, 2021, doi: 10.1038/s41598-021-93040-y.

[18] R. Natarajan *et al.*, "Performance study on adsorptive removal of acetaminophen from wastewater using silica microspheres: Kinetic and isotherm studies," *Chemosphere*, vol. 272, p. 129896, 2021, doi: 10.1016/j.chemosphere.2021.129896.

[19] N. Dhiman and N. Sharma, "Removal of pharmaceutical drugs from binary mixtures by use of ZnO nanoparticles: (Competitive adsorption of

drugs)," *Environmental Technology and Innovation*, vol. 15, p. 100392, 2019, doi: 10.1016/j.eti.2019.100392.