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REMOVAL OF 5-FLUOROURACIL USING ACTIVATED CARBON PREPARED FROM BARK OF STERCULIA FOETIDA

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ABSTRACT

Pharmaceutical pollution is a growing concern today and our study focused on removal of environmentally hazardous pharmaceutical pollutant 5-flurouracil from synthetic fluorouracil wastewater using surface modified agricultural waste (Fluorouracil) and to study its adsorption characteristics.

The present study and analysis investigates the potential use of Activated Carbon extracted from Bark of Sterculia foetida in the removal of 5-fluorouracil from aqueous solution using the batch adsorption processes. Adsorption of 5-fluorouracil onto the adsorbent was found to be influenced by the adsorbent dose, 5-fluorouracil concentration, temperature and contact time. On the basis of the results obtained it can be concluded that the adsorbent have a very high potential on the removal of 5-fluorouracil molecules from its aqueous solution.

Keywords: Adsorbent, 5-Fluorouracil, drug, adsorbent dose, surface area

1.INTRODUCTION

1.1 INDUSTRIAL WASTEWATER

Water is an essential source for all living things to achieve their basic needs; also in the modern world water is very essential for the industries to carry out their production process as water finds vast application in various units of the industry. Water's properties such as high heat conductance, low viscosity, high density, compressibility (in vapor form), flow ability are utilized by industries to transport heat and materials, dilute chemicals, in mixing and cooling fluids and chemical synthesis, washing and for various other purposes[1]. Some industries that consume fresh water and produce waste water are Pharmaceutical industry, Iron industry, Steel industry, Mines, Quarries, Food industry, Dyeing industry, Oil refinery, petrochemical industry, Cement industry, Paint industry, Pulp and Paper industry. Large volumes of effluents are produced which in turn makes storing and treating it difficult and tedious, as a result of which toxic waste water is discarded as effluents into water bodies and land areas near the industries thereby causing severe environmental problems and associated health risks to life[2].

1.2 DRUG POLLUTION

Drug pollution or pharmaceutical pollution is pollution of the environment with pharmaceutical drugs and their metabolites, which reach the aquatic environment (groundwater, rivers, lakes, and oceans) through wastewater. Drug pollution is therefore mainly a form of water pollution. Pharmaceutical products for humans or animals, as well as their related metabolites (degradation products) end up in the aquatic environment after use. Recent investigations showed that low concentrations of pharmaceuticals are also detectable in municipal wastewater, surface water, and ground water and even in drinking water[2].

Pharmaceuticals do not usually persist in the environment but continuous input into the environment keeps concentrations relatively constant. Many drugs have been detected in various water streams throughout several countries. However, their concentrations are usually below the therapeutic level, the risks and effects of these substances in the environment have not been determined yet. However, this issue continues to be viewed with concern due to some of the properties that many pharmaceuticals have like biological activity, lipophilic nature and resistance to biodegradation etc[3].

SOURCE AND EFFECTS

Most simply from the drugs having been cleared and excreted in the urine. The portion that comes from expired or unneeded drugs that are flushed unused down the toilet is smaller, but it is also important, especially in hospitals (where its magnitude is greater than in residential contexts). This includes drug molecules that are too small to be filtered out by existing water treatment plants. The process of upgrading existing plants to use advanced oxidation processes that are able to remove these molecules can be expensive. Drugs such as antidepressants have been found in the United States Great Lakes. Researchers from the University of Buffalo have found high traces of antidepressants in the brains of fish. Fish behavior on antidepressants has been noted to have similar impacts and reducing risk-averse behavior, and thereby reducing survival through predation[3].

Other sources include agricultural runoff (because of antibiotic use in livestock) and pharmaceutical manufacturing. Drug pollution is implicated in the sex effects of water pollution. It is a suspected contributor (besides industrial pollution) in fish kills, amphibian die offs, and amphibian path morphology.

1.2.1 PREVENTION

The main action for preventing drug pollution is to incinerate unwanted pharmaceutical drugs rather than flushing them down the drain. Burning them chemically degrades their active molecules, with few exceptions. The resulting ash can be further processed before land filling, such as to remove and recycle any heavy metals that may be present.

There are now programs in many cities that provide collection points at places including drug stores, grocery stores, and police stations. People can bring their unwanted pharmaceuticals there for safe disposal, instead of flushing them (externalizing them to the waterways) or throwing them in the trash (externalizing them to the landfill, where they can become leachate)[4].

1.2.2 DRUGS CAUSING POLLUTION

Antineoplastic drugs are employed during chemotherapy all over the world. They pollute water courses and have 'mutagenic, cytostatic, and ecotoxicological effects on the micro-organisms that are in the aquatic environment.' The wastewater treatment process is not able to remove antineoplastic drugs due to the intractable nature of them. Bodies of water that are contaminated with antineoplastic drugs possess grave consequences on the aquatic environment and even human health. Chemotherapy drugs such as cyclophosphamide 1, fluorouracil, doxorubicin, cisplatin and mitomycin C were discovered to cause genotoxicity in aquatic organisms[5].

Antibiotics are widely produced and consumed to treat bacterial and fungal diseases. Since antibiotics are only partially metabolized, the non-metabolized antibiotics are released into the environment. Due to this, antibiotics are discovered in sludge, drinking water, wastewater, surface water, soil, groundwater and sediments. Residual antibiotics are not easily degraded so they can survive in environments for long periods of time. There is an urgent push to eradicate antibiotics from the environment because they could cause a generation of antibiotics resistance bacteria and antibiotics resistance genes, which would pose an immense threat to the ecological system and human health[6].The excessive use and excretion of antibiotics to waterways makes the problem of antimicrobial resistance worse and will gradually affect the human population, possibly causing more deaths[5]. Antibiotics were found to reduce growth in algae, aquatic plants and environmental bacteria.

1.2.3 5-FLUOROURACIL

Fluorouracil is a nucleobase analogue that is uracil in which the hydrogen at position 5 is replaced by fluorine. It has a role as a xenobiotic, an environmental contaminant, a radiosensitizing agent, an antineoplastic agent, an immunosuppressive agent and an antimetabolite. It belongs to a group of anti- cancer medicines called 'antimetabolite' used to treat breast, skin, colon, rectum and stomach cancer. Cancer is a disease in which abnormal cells divide uncontrollably and destroy body tissue. Some of the body's cells begin to divide into all cancer types without stopping and spread into surrounding tissues.Cancer can be localized (benign) or spread to the whole body (metastasized). FLUOROURACIL contains 'Fluorouracil' that works by interfering with the growth of genetic material (DNA and RNA) of the cancer cells[7]. This prevents the cancer cells from multiplying and growing and eventually kills them.

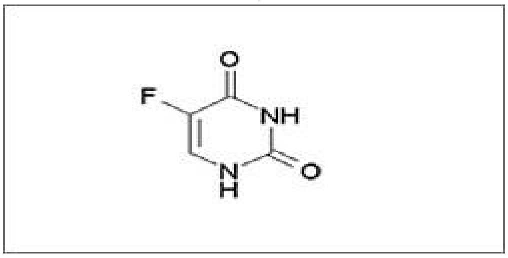

Fig 1: Structure of Fluorouracil

1.2.4 FLUOROURACIL POLLUTION

Fluorouracil (Fig. 1) belongs to the subclass of antimetabolites and its consumption reaches the range of tonnes in Europe. Several studies have reported its presence in natural water at concentrations ranging from mg L-1 to µg L-1. Conventional water treatment methods based on biological processes have been shown as insufficient to eliminate the majority of target pharmaceuticals because of their high recalcitrance and toxicity to bacteria. 5-Fluorouracil is not an exception and, furthermore, it has been demonstrated that photodegradation by sun is not suitable for its removal either, showing partial degradation and no mineralization. Therefore, it does not undergo natural attenuation in the environment and cannot be removed in traditional treatment plants.

Due to the prevalence of 5-FU in all forms of water, conventional wastewater treatment is sometimes ineffective and insufficient in eliminating it. Even though they appear to be inefficient at removing 5-FU, a number of tertiary treatment technologies, including microfiltration, ultrafiltration, reverse osmosis, and sand filters, are widely utilized at waste water treatment plants. As many process fail to eliminate the substantial amount of fluorouracil present in water, adsorption is chosen as a better method as it is predominant in separation processes due to its low cost and high selectivity[7].

1.2.6 ADSORPTION

Adsorption is the adhesion of atoms, ions, or molecules from a gas, liquid, or dissolved solid to a surface. This process creates a film of the adsorbate on the surface of the adsorbent. This process differs from absorption, in which a fluid (the absorbate) permeates or is dissolved by a liquid or solid (the absorbent)[8]. Adsorption is a surface- based process while absorption involves the whole volume of the material. The term sorption encompasses both processes, while desorption is the reverse of it. Adsorption is a surface phenomenon. Similar to surface tension, adsorption is a consequence of surface energy. In a bulk material, all the bonding requirements (be they ionic, covalent, or metallic) of the constituent atoms of the material are filled by other atoms in the material. However, atoms on the surface of the adsorbent are not wholly surrounded by other adsorbent atoms and therefore can attract adsorbates. The exact nature of the bonding depends on the details of the species involved, but the adsorption process is generally classified as physisorption (characteristic of weak van der Waals forces) or chemisorption (characteristic of covalent bonding). It may also occur due to electrostatic attraction[9].Adsorption is present in many natural, physical, biological, and chemical systems, and is widely used in industrial applications such as activated charcoal, capturing and using waste heat to provide cold water for air conditioning and other process requirements (adsorption chillers), synthetic resins, increase storage capacity of carbide-derived carbons, and water purification. Adsorption, ion exchange, and chromatography are sorption processes in which certain adsorbates are selectively transferred from the fluid phase to the surface of insoluble, rigid particles suspended in a vessel or packed in a column. Lesser known, are the pharmaceutical industry applications as a means to prolong neurological exposure to specific drugs or parts thereof.

1.2.6.1ADSORBENT

Adsorbents are used usually in the form of spherical pellets, rods, moldings, or monoliths with a hydrodynamic radius between 0.25 and 5 mm. They must have high abrasion resistance, high thermal stability and small pore diameters, which results in higher exposed surface area and hence high capacity for adsorption. The adsorbents must also have a distinct pore structure that enables fast transport of the gaseous vapors. Activated carbon has been widely used as an effective adsorbent for the removal of dyes from the aqueous solutions because of their high adsorption capacities and atmospheric properties. However, the cost of production and the regeneration of the activated carbon are high, and its application was unjustified for the most pollution control applications.

It gives an idea for the search of alternatives and low-cost adsorbents[8].

1.2.6.2.STERCULIA FOETIDA

Sterculia Foetida is a plant of Sterculiacaea family. It has numerous medicinal uses. Several studies are available to prove the effect of sterculia foetida fruit shell ,gum, leaves as an adsorbent. The bark of the tree is understudied part. So the main focus of this article is to determine the performance and properties of an adsorbent prepared from the bark of Sterculia foetida and to prove its effectiveness in removing the pharmaceutical pollutant 5-flurouracil[9].

2. MATERIALS AND METHODS

2.1 PREPARATION OF ADSORBENT

Sterculia foetida bark was collected from Veltech High-tech College campus, Tamil nadu. The bark sample was washed with water to remove the debris present on their surface. The bark was then sun dried for three days to remove the free moisture content and this was followed by drying in a tray drier at 90^{o}C for 12 hours. The outer epidermis was removed manually after drying and the bark. The bark was then crushed in a jaw crusher which was the primary size reducing operation. The broken bark was further crushed in a roll crusher followed by a pulveriser. The powdered materials are sieved to 100 mesh particle size. All the oversize materials were again sent back to the pulveriser[10].

The fine powder was again dried in a hot air oven at 110^{o}C in order to remove the excess moisture present in it. This raw bark powder was converted to carbon based adsorbent by surface treatment with sulphuric acid.

The weighted materials were taken in a ceramic bowl and sulphuric acid about twice its weight was added to it gradually with constant mixing using a glass rod. The entire setup was placed in fume hood so that the toxic vapours generated during the process could be vented. The acid treatment process time was 24 hours which was carried out with hourly mixing to ensure proper surface contact of the acid.After 24 hours the prepared adsorbent was washed with distilled water till the pH of the wash water was 7. It was ensured that all the excess acid was removed and the adsorbent didn’t possess any acidity in its surface. The adsorbent was then dried in a tray drier at 90^{0}C for about 2 hours so that the moisture is completely removed. Due to the surface treatment process mild agglomeration was observed hence the material was again crushed in a mortar and pestle and the dried adsorbent was stored in airtight container[11].

2.1.1 ULTRASONIC SURFACE TREATMENT

Ultrasonic waves are sound waves transmitted above the human-detectable frequency range, usually above 20,000 Hz. They are high energy waves and when they are passed through a liquid medium such as water due to the propagation of longitudinal waves high energy bubbles are created and due to the collapse of these bubbles energy is liberated, this energy is utilized for modifying the surface of the adsorbent.

The dried surface modified bark was taken in a beaker and twice its weight water was added and an ultrasonic probe was inserted into the liquid suspension. The entire setup was enclosed in a wooden box and an ultrasonicator was switched on[12].

Ultrasonic waves at frequency of 22 KHz and a power of 750 watts were passed into the suspension intermittently with cycle duration of 10 seconds for 30 mins. The beaker was jacketed with a cooling water beaker so that the heat produced during the process is dissipated effectively. The Ultrasonicated powdered material was then filtered using a filter paper to remove water. The filtrate was then dried in a hot air oven at 90°C for 120 minutes to remove the excess moisture content present in it[13].

2.2 ADSORPTION STUDIES

The surface modified bark was preserved in the container. The adsorption studies would be further continued by the preparation of fluorouracil solution by adding different concentrations of drug. The adsorbent also would be prepared at various concentrations and the effect of drug on the adsorbent would be studied. The qualitative analysis would be carried out in which the effect of fluorouracil on the adsorbent would be investigated on varying the drug concentration, contact time, adsorbent concentration, and temperature[14]. The concentration of 5-fluorouracil in the supernatant was analyzed by using the UV–vis spectrophotometer. The percentage removal of 5-fluorouracil was calculated by using the following equation:

$$\% \text{ Removal} = \frac{c_i - c_f}{c_i} X\, 100$$

Where c_i is the initial diclofenac concentration (mg/l) and c_f is the final 5-fluorouracil concentration (mg/l)[15].

3.RESULTS AND DISCUSSION

3.1 EFFECT OF ADSORBENT DOSE

The effect of adsorbent dose on the adsorption of fluorouracil, batch adsorption studies would be carried out using an initial 5-fluorouracil concentration of 50ppm. The adsorbent dose would be varied from 2g/l to 18g/l and the adsorption process would be allowed to reach equilibrium while maintaining isothermal conditions throughout the process. The effect of adsorbent dose on the percentage removal will be analyzed[16].

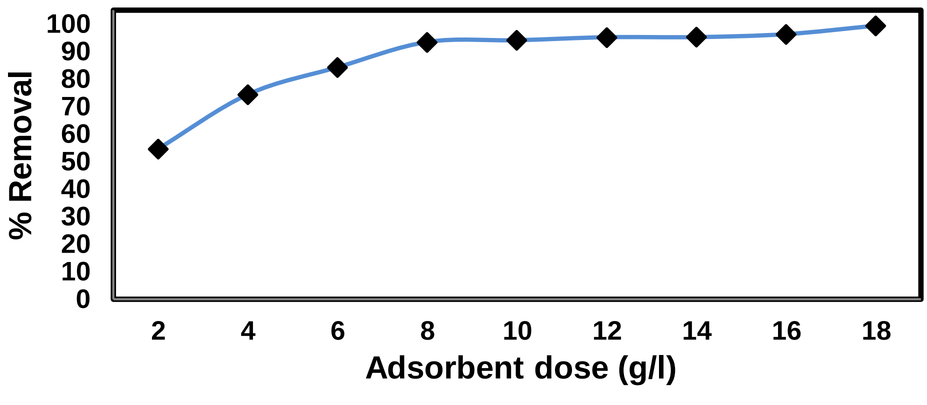

Fig 2. EFFECT OF ADSORBENT DOSE

It is observed that as the dose of the adsorbent is increased, the amount of 5-Fluorouracil removed from the solution also increases. Also it is noted from the graph that after a certain dose there is a negligible change in the percentage removal. This is possibly due to the relatively low equilibrium concentration, low driving force, and the available active sites gradually decreased.

3.2 EFFECT OF INITIAL FLUOROURACIL CONCENTRATION

Initial fluorouracil concentration plays a significant role in the removal of metal ions from aqueous solution as it gives the driving force that overcomes mass transfer resistance of absorbate from aqueous and solid phase. To study the effect of initial concentration on adsorption, the initial concentration of fluorouracil solution would be varied from 50 ppm-250 ppm and the adsorbent concentration was fixed to 10g/l. The effect of varying concentration would be noted[17].

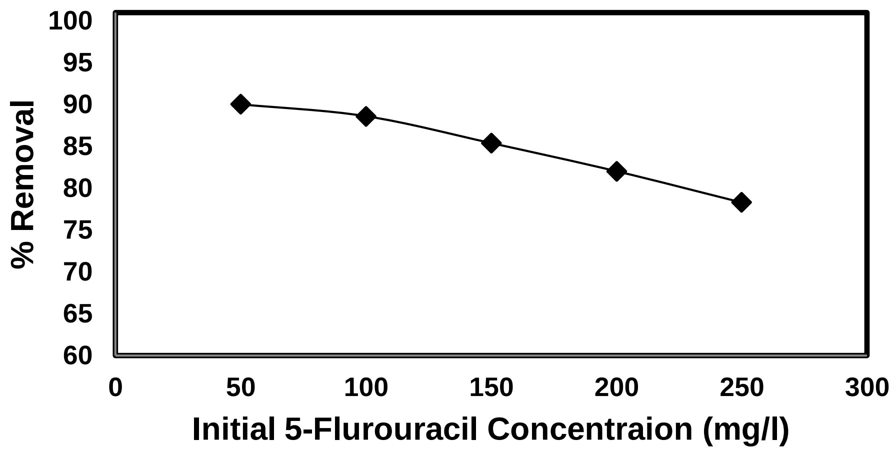

Fig 3 .EFFECT OF INITIAL FLUOROURACIL CONCENTRATION

It could be seen that as the initial concentration is increased the percentage removal has decreased but the maximum adsorption increases as the driving force increases.

3.3 EFFECT OF THE CONTACT TIME

The effect of contact time on adsorption would be studied by analyzing the concentration of fluorouracil at regular time intervals. The dose of adsorbent was fixed at 10g/l and the experiment was carried out at various initial concentrations to find the optimum contact for maximum removal.

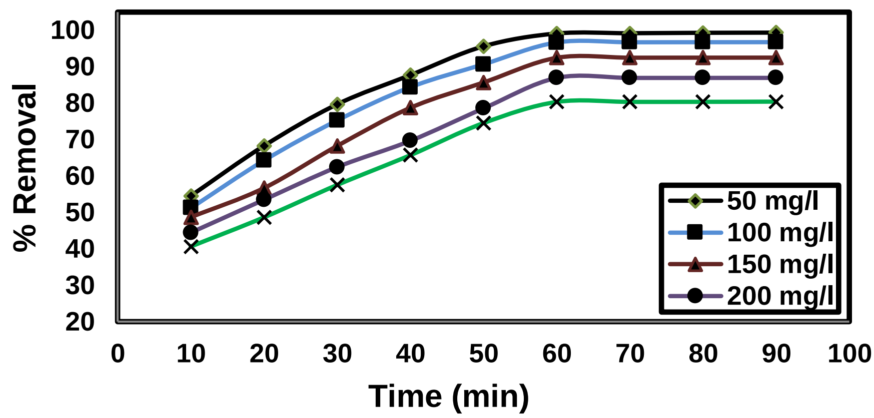

Fig 4. EFFECT OF THE CONTACT TIME

Maximum adsorption was observed at 60mins after which there was no considerable removal. Hence 60mins was found to be the optimum time for 99% removal[18].

3.4 EFFECT OF TEMPERATURE

Majority of the adsorption process is exothermic with a few exceptions and it is thus important to study the effect of the temperature at which adsorption is carried out. The effect of temperature on adsorption would be studied in the temperature range 303K-313K and for different concentrations ranging from 50 mg/l to 250 mg/l. The adsorbent concentration would however fix to 10g/l.

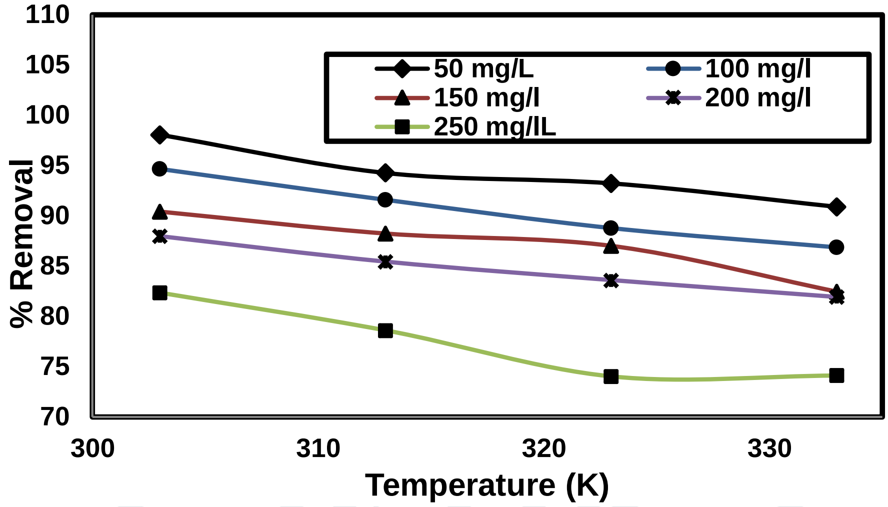

Fig 5. EFFECT OF TEMPERATURE

It was observed that the removal was high at lower temperatures and removal decreased with increase in temperature. Thus the exothermic nature of the adsorption process is evident[19]. The data thus obtained was used for calculating the thermodynamic properties for the adsorption process.

4.CONCLUSION

Pharmaceutical pollution is a growing concern today and our study focused on removal of environmentally hazardous pharmaceutical pollutant 5-fluorouracil from waste water using surface modified agricultural waste (Sterculia foetida)[20]. The adsorption behavior of Sterculia foetida on the drug is analyzed by the effect of various parameters like adsorbent dose, drug concentration, time, temperature. The results obtained prove us that the bark of Sterculia foetida bark can be used as a best source to eradicate the effect of drug pollutant like 5-Fluorouracil[21].

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