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A Study on Biosorption of Drug Using Green Mediated Synthesis of Nanocomposite

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ARTICLE DETAILS

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ABSTRACT

This study focused on the use of Indian leaf biomass, a local plant widely found in India, on removal of Aceclofenac from pharmaceutical waste water by biosorption. The biosorption characteristics of Aceclofenac were investigated in terms of equilibrium and kinetics. Optimum biosorption conditions were determined from pH, initial Aceclofenac concentration, biomass dosage, contact time, and temperature. The best interpretation for the experimental data was given by the Langmuir isotherm with correlation coefficient of 0.965. The results were found to tie in well with pseudo-second-order kinetics. Considering the cost- effectiveness, this biomass is considered to be suitable to remove Aceclofenac from pharmaceutical waste water

1. Introduction

Pharmaceuticals, particularly antibiotics, represent a concerning new class of pollutants in the environment, not to mention their effects on human health with regard to several infectious illnesses (Ahmadi et al. 2021; Li et al. 2022). Most of these chemicals are found in surface water at elevated amounts (Zare et al. 2022). Their bioresistant character and ability to elude traditional sewage treatment procedures are strongly tied together. It harms the ecology by making bacteria resistant and interfering naturally with the growth, development, and movement of a variety of microorganisms (Limousy et al. 2017). One of the biggest obstacles to a sustainable water future is the presence of this material in the aquatic environment, particularly in arid nations where water recycling is crucial. As a result of their extensive use in both human and animal medications, antibiotics play a significant role in water contamination. They are employed to treat bacterial infections and are extremely resistant to being inactivated until they have completed their intended function, which causes their incomplete metabolism in the organism (Naeini et al. 2022). More than 90% of medications taken orally don't decompose, thus they become active compounds. Since antibiotics are highly soluble in water, conventional treatment procedures cannot eliminate them, which poses a significant obstacle to their removal (Zhu et al. 2015).

Zerodol SP is a non-steroidal anti-inflammatory drug (NSAID) advocated for use in painful, inflammatory and swelling of muscles, joints and bones in arthritis. It is usable in a number of government forms which can be given orally, rectally or intramuscularly. Conveniently, dosage adjustments are not involved in the elderly or in those patients with renal or hepatic damage. The The main mechanism responsible for its anti-inflammatory, antipyretic, and analgesic action is believed to be inhibition of prostaglandin synthesis by inhibition of the transiently expressed prostaglandin-endoperoxide synthase-2 (PGES-2) also known as COX-2. Glandin synthesis by inhibition of the transiently expressed prostaglandin-endoperoxide synthase-2 (PGES-2) also known as cycloxygenase-2 (COX-2). It seems to exhibit bacteriostatic activity by inhibiting bacterial DNA synthesis. Then the inhibition of prostaglandin synthesis occurs various symptoms such as soreness of the gastric epithelium. This is the main side effect of Aceclofenac, Paracetamol and Serratiopeptidase. Aceclofenac, Paracetamol and Serratiopeptidase inhibits COX-2 with 20 times greater strength than the constitutively expressed isoenzyme COX-1and has, consequently, a somewhat lower incidence of gastrointestinal complaints than noted

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with aspirin which inhibits COX-1 to a larger extent. The action of one individual window pane is much longer (6 to 8 hr) than the very short 1.2–2hour half-life of the drug would indicate. This could be partially because it runs for over 11hours in synovial fluids.

The combination of Aceclofenac, Paracetamol and Serratiopeptidase may also be a singular member of the NSAIDs. Some result indicates it inhibits the lipoxygenase pathways, thus decreasing formation of the leukotrienes (also pro-inflammatory autacoids). It also may inhibit phospholipase A2 as part of its mechanism of activity. These additional activities may explain its high potency-it is the most potent NSAID on a wide base. There are some differences exist in NSAIDs in this present inhibition of the two subtypes of cyclooxygenase, COX-1 and COX-2. Much pharmaceutical drug design has tried to focus on selective COX-2 inhibition as a fashion to minimize the gastrointestinal side effects of NSAIDs such as acetylsalicylic acid. In drill, use of some COX-2 inhibitors with their adverse effects has led to massive numbers of patient, family lawsuits alleging wrongful death by heart attack, yet other significantly COX-selective

2. Materials and methods

2.1 Reactants

Powdered activated carbon (AC) (with surface area 893.83 m2 g-1), Acetic acid, Zinc oxide and Chitosan were purchased from Sigma- Aldrich Chemicals and used such as. The combination of Aceclofenac, Paracetamol and Serratiopeptidase (See Supplementary) were furnished by MedChem express and used without purification.

2.2 Collection of Samples and Preparation of Extract

Fresh green leaves (Mimusops elengi) of good health were collected from a nearby farm. They were rigorously washed to get rid of dust particles and then rinsed thrice with double distilled water and weighed afterwards. Five (5g) of powdered sample was soaked with 50 ml of water and acetone separately. The entire mixture was incubated at 4°C for 48 hr. After the incubation period was over, the mixture was filtered and centrifuged at 10,000rpm at 4°C. The extracts were dried and stored at 4°C until further used.

NSAIDs, such as Aceclofenac, Paracetamol and Serratiopeptidase have been well borne by most of the population. Then the COX-inhibition, a number of other targets Aceclofenac, Paracetamol and Serratiopeptidase like pain-relieving actions have recently been identified. These include the following:

Blockage of voltage-dependent sodium channels (after activation of the channel, the combination of Aceclofenac, Paracetamol and Serratiopeptidase inhibits its reactivation also known as phase inhibitory) Blockage of acid-sensing ion channels (ASICs) Positive allosteric modulation of KCNQ- and BK-potassium channels (The combination of Aceclofenac, Paracetamol and Serratiopeptidase opens these channels, leading to hyperpolarization of the cell membrane. Broad clinical experience has been gained with the combination of Aceclofenac, Paracetamol and Serratiopeptidase, clearly showing its safety profile. It is well tolerated compared with other NSAIDs and rarely produces gastrointestinal ulceration or other serious side effects. Thus, the combination of Aceclofenac, Paracetamol and Serratiopeptidase can be regarded as unitary of the few NSAIDs of' first choice' in the treatment of acute and chronic painful and inflammatory conditions such as upset stomach, nausea, heartburn, diarrhoea, irregularity, flatulence, headache, somnolence, dizziness may occur. This medication may cause your blood pressure to drop. This drug may rarely cause severe (perhaps fatal) liver disease. In the present study, the polymeric carbon/ CS- ZnO nanocomposites were prepared and characterized. These polymeric nanoadsorbents were applied to remove pharmaceuticals from aqueous solutions and the results proved to be very efficient for the combination of Aceclofenac, Paracetamol and Serratiopeptidase removal from aqueous solutions. Drug induces a relatively short elimination half-life, which defines the potential for drug accumulation. In numerous clinical tests the efficacy of Zerodol SP is equivalent to that of the many newer and established NSAIDs with which it has been likened. As an analgesic it has a quick attack and long continuance of activity.

2.3 Green-synthesis of Nanocomposite

40 ml of prepared Mimusops elengi extract at pH 4 was mixed with a solution containing 0.025 (M) Zinc oxide (5 ml) and 0.025 (M) Chitosan (5 ml) (1:1 vol ratio). The reaction starts as the solution was kept at 70oC for 10-15 min and after then without disturbing the content it was again left for 30-40 min. Few minutes later a clear phase separation was observed with light colour solvent at the top and green thick precipitates at the bottom. After filtering, the retentive green portion was freeze dried at 15oC. Powdered nanocomposite was formed by using a vacuumed lyophilizer chamber maintained at temperature 50°C and pressure 0.04 mbar. Finally the nanocomposite powder was homogenized to pH 7, dried and kept within vacuum desiccators for further use.

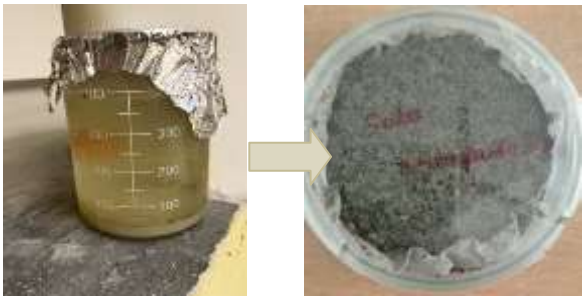


Fig 1. CS/ZNO Nanocomposite

The Acetic acid, Zinc oxide, Chitosan and Charcoal in 25ml of distilled water each. Both the solution were mixed and kept in a magnetic stirrer for 30 minutes at pH 7 in room temperature at 650 rpm. Then add 5g of Activated Carbon and green leaves (Mimusops elengi) powder extract to the solution and again kept in the magnetic stirrer for 4 hrs in room temperature at 1100-1300 rpm. The solution was filtered and dried overnight and further heated at 400°C in a Muffle furnace for 10 minutes. Then it is converted into powder form, the nanocomposites are synthesized.

1g of Acetic acid in 99 ml of distilled water +4.21g of Zinc oxide and Chitosan each are mixed into 25ml of distilled water
↓
Both the solution were mixed and kept in a Magnetic stirrer at room temperature for 30 minutes at 650 rpm
↓
Add 5g of Activated carbon and green leaves powder extract to the solution and
↓
again, kept in a Magnetic stirrer for 4hrs at room temperature at 1100-1300 rpm
↓
Then the solution was filtered and dried for overnight and further heated in a Muffle furnace for 10 minutes at 400°C
↓
Nanocomposite

3. Schematic Diagram of Preparation of Nanocomposite

3.1 Characterization of Nanocomposite

The silver Scanning Electron Microscopy-Energy Dispersive X-ray Spectrometry (SEM-EDX) Analysis

The microstructure and composite homogeneity of the obtained samples were investigated using a SEM/EDX scanning microscope JEOL-JSM 64000 LV. Energy dispersive X-ray analysis measurements were performed under nanoparticles were centrifuged at 10,000 rpm for 30 min and the pellet was redispersed in 10 mL ethanol and washed 3 times with sterile distilled water to obtain the pellet. The pellet was dried in an oven and thin films of dried samples (10 mg/mL) were used for compositional analysis.

4. Result

*4.1 Fourier Transform Infrared Spectroscopy (FTIR) analysis***.**

The vacuum dried samples were mixed with potassium bromide (KBr) and the spectra were recorded with a Perkin Elmer Spectrum Express version 1.03.00. The scanning data were obtained from the average of 47 scans in the range 4000–400cm-1 with the resolution of 4cm-1.

4.2 Adsorption studies

The experiments on the drugs were conducted in 100 mL Erlenmeyer flasks containing 50 mL of the drug solution of known concentration and one gram of the adsorbent at the appropriate pH and agitated by a mechanical shaker at room temperature. After a definite time interval, the solution was filtered and the filtrate thus obtained was analyzed spectrophotometrically by measuring the absorbance at λmax of drug solution. All the experiments were duplicated and the mean values were reported. The maximum deviation observed was less than ±5 %. In the adsorption kinetic experiments samples were taken to measure the drug removal at predetermined time intervals. The amount of drug biosorbed per unit mass was calculated using the following equation:

$$Q = (Co-Ce)*V/W \quad (1)$$

where q is the amount of adsorbed dye (mg/g), Co and Ce are the initial and equilibrium concentration, respectively. V is the volume of the solution (mL) and W is the amount of the biosorbent.

4.3 Synthesis and Characterization of Nanocomposite

The present study revealed that the adsorption capacity of the metal activated nanocomposites. The synthesized nanocomposites were initially confirmed by visual observation by colour change. It was observed that the colour of the

reaction mixture was changed from light colour to dark colour. Further, the synthesized nanocomposites were characterized by Fourier Transform Infrared Spectroscopy (FT-IR), and Scanning Electron Microscope (SEM)-EDAX.

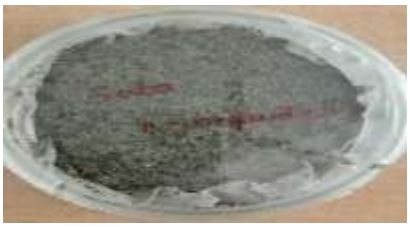


Fig 2 CS/ZNO Nanocomposite

4.4 SEM-EDX Analysis

The size and surface morphology of the CS/ZNO Nanocomposite were obtained by Scanning Electron Microscopy (SEM) analysis is shown in Figure 3. This study shows that white diminutive particles were well diffused in the pores of the carbon shell and were aggregated well because of its paramagnetic nature. From this study, we could not find exact size and shape of the particles in carbon shell, which suggests that the presences of metal oxides are in the nanosize range. The formation of nanocomposite as well as their morphological dimensions in the SEM study demonstrated that the average size of 12-15nm and 15-20nm respectively. EDX analysis confirmed the presence of Zn, Ca, and C and O elements within nanocomposite.

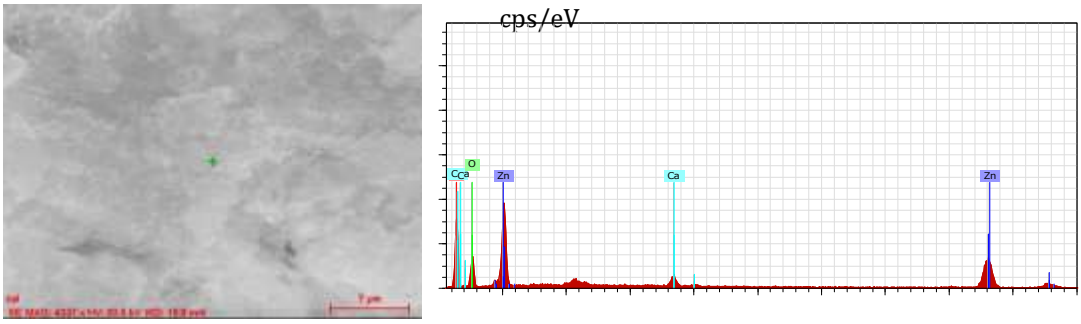
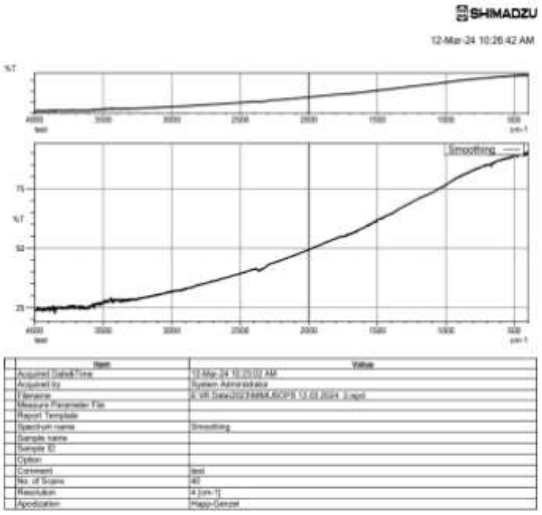


Fig 3 SEM-EDX analysis of CS/ZNO Nanocomposite

4.5 FTIR Analysis

FT-IR spectra analysis revealed that the characteristic peaks in the range of 1500–1570 and 2320–2370cm-1 due to AC and this corroborates that the carbon was not decomposed in the nanocomposites during the thermal decomposition of the carboxylates. Moreover, untreated AC and the nanocomposite do not show any carboxylate stretching in their spectra. This reveals that the entrapped metal carboxylates are well decomposed in carbon shell. These results are similar to other reported values (Ranjithkumar and Vairam, 2012).

4.6 FTIR analysis of CS/ZNO Nanocomposite



5. Optimization of Adsorption studies

5.1 Effect of pH on Adsorption

The pH of the solution is a key factor that could affect the adsorption of drug on an adsorbent (Arya and Philip, 2016; Saucier et al. 2015b; Shan et al. 2016; Zhao et al. 2016). The optimum pH of organics is a function of the chemical nature of adsorbent, and the solubility of the organics also depends on the pH of the solution. In this study, it was observed that for pH values 1,4,7,10 and 14, the percentage of removal for Aceclofenac was practically constant within this pH interval. For this reason, all the solutions were prepared in pH 7.0, considering that a wastewater contaminated with these pharmaceuticals would need to be neutralized before being released to the environment. For the treatment with adsorption, it is not necessary to make any pH adjustments if the solution of the effluent is within pH-1,4,7,10 and 14. This result also reveals that the mechanism of adsorption of Aceclofenac onto nanocomposite should not be electrostatic attraction, since this mechanism is pH dependent as may be observed several times in the literature (Calvete et al. 2010; Dos Santos et al. 2015).

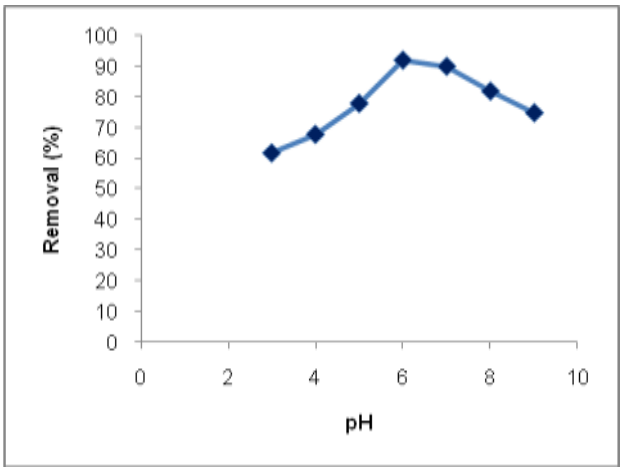


Fig 4 Effect of pH on Adsorbent

6. Equilibrium Models

The isothermal models and adsorption kinetics are shown in Figure 5 and 6. The results showed that drug fitted according to Freundlich isotherm model (R2=0.99). The R2 of kinetic models suggested that the pseudo-second-order model mechanism is predominant which means the uptake process follows the pseudo second-order expression with correlation coefficients were always greater of 0.9379.

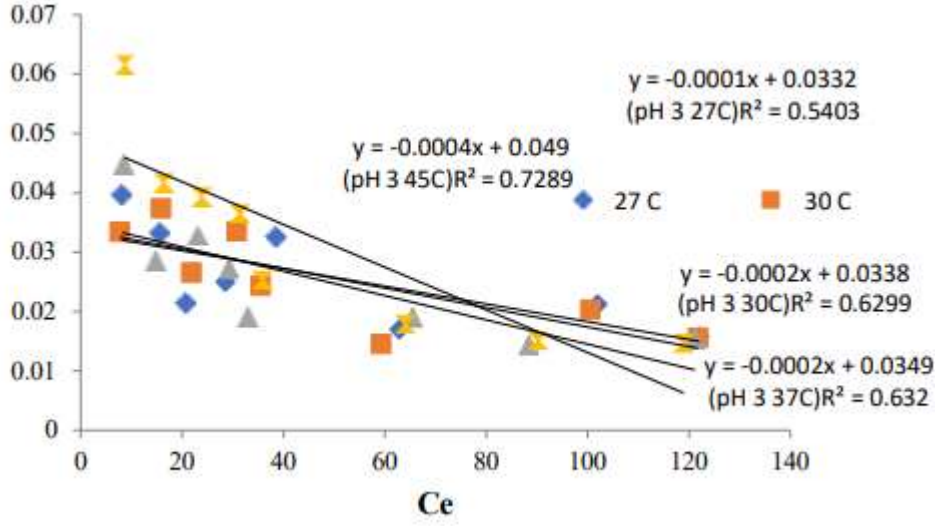


Fig 5. Langmuir isotherm

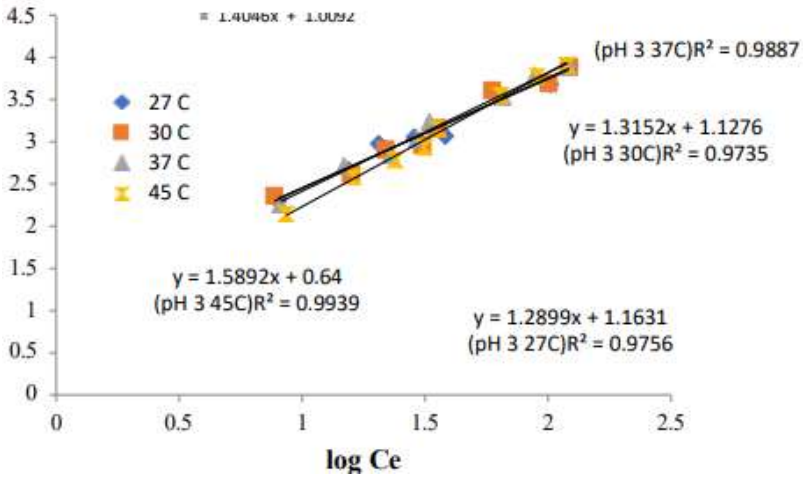


Fig 6 Freundlich isotherm

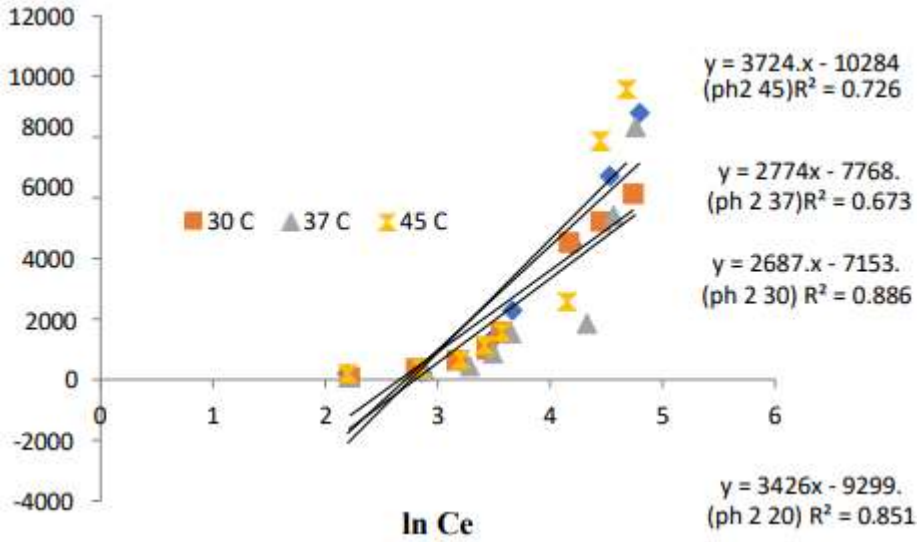
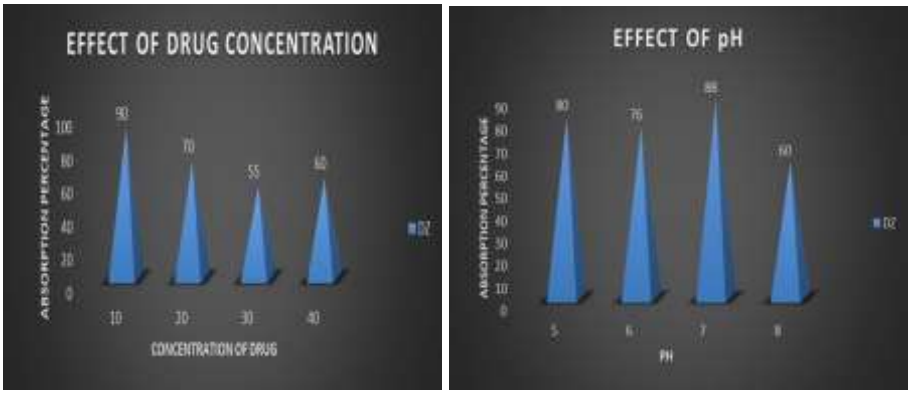
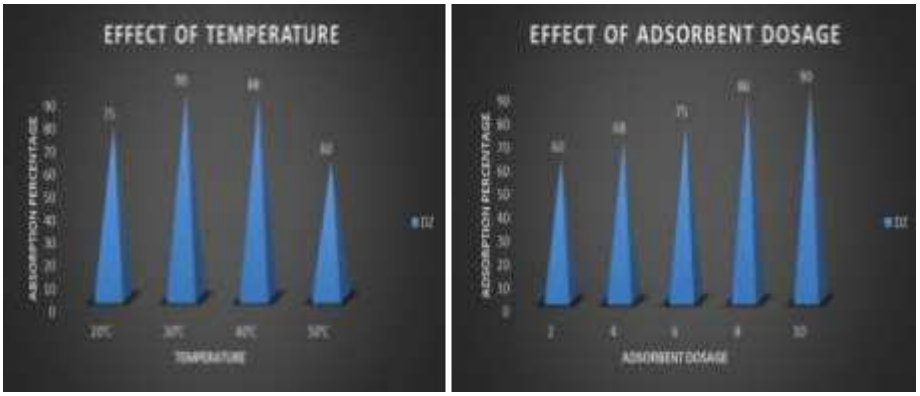


Fig 7 Tempkin isotherm

In order to test the performance of adsorbents on wastewater treatment, they were applied on simulated hospital effluents. The spectra of the simulated effluents before and after treatment with the activated carbons were recorded from 200 to 800 nm by UV-Vis spectrophotometer. The areas under the absorption bands give the percentages of the mixture of pharmaceuticals removed from the effluents. It provides a slightly better performance for the treatment of simulated hospital effluents. These results are in agreement with the maximum sorption capacities.

*6.1 Effect of various parameters on Adsorption of drugs using Nanocomposi*te





6.2 Determination of Ash and Moisture content

The ash and moisture content of Commiphora wightii are listed below (Table 1 and Figure 8). The moisture content was 40±0.26 and ash was 37±0.22. The similar result was reported by Saraswathy et al. (2022). Dried products containing nutrient content less than 10% indicate a good keeping quality. Hence it was clear that moisture content of the plants was within the acceptable range and indicates a good keeping quality.

Table 1.1 Ash and Moisture content of *Mimusops elengi*

Parameters	***Mimusops elengi***
Moisture (%)	40 ± 0.26
Ash (%)	37 ± 0.22

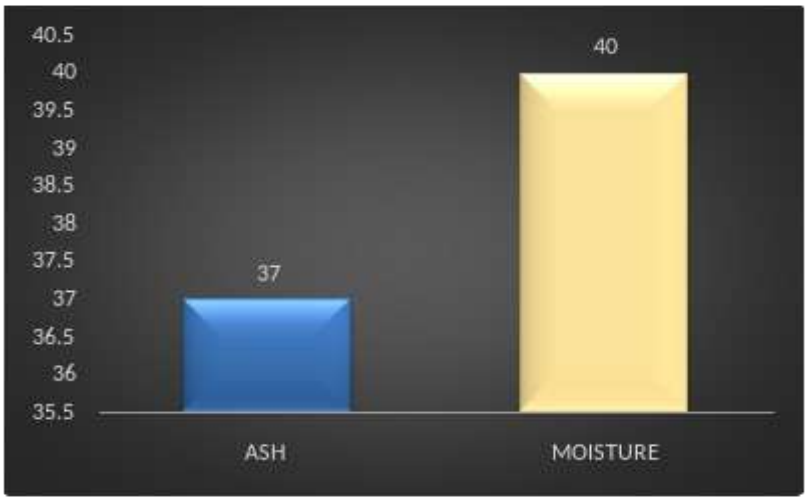


Fig 8 Ash and Moisture content of *Mimusops elengi*

7. Conclusion

The present study provides evidence that CS/ZnO nanocomposites were prepared by a simple pyrolytic method using a mixture of Chitosan, Activated carbon, Zinc oxide, were developed by mixture of extracts, respectively, and they were used as adsorbents for the successful removal of Aceclofenac (ACF) from aqueous effluents.

The synthesized nanocomposites were characterized by Fourier transform infrared spectroscopy (FT-IR) and Scanning electron microscopy (SEM)-Energy dispersive X-ray spectroscopy (EDX). The kinetics of adsorption was evaluated by using nonlinear pseudo first-order, pseudo-second-order and general order kinetic models. For Aceclofenac, the general order kinetic model was the best model that was adjusted to the experimental data. The equilibrium data were also fit using the nonlinear Langmuir, Freundlich and Tempkin isotherm models. For pharmaceuticals using the adsorbents, the best fitting of the equilibrium data occurred for the Freundlich isotherm model, it is possible to propose a mechanism of adsorption of Aceiclofenac onto the adsorbents. Also, the CS/ZnO nanocomposites were successfully used as adsorbents for treatment of simulated hospital effluents removed. From this study, it can be concluded that the synthesis of nanocomposite is effective in adsorption removal of Aceclofenac.

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