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Research Article

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Adsorption of Losartan onto Rice husk-derived Activated carbon to remove it from an Aqueous solution

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Abstract

Rice husk (RH) has been studied as a prospective adsorbent for the removal of pharmaceuticals like drugs, dyes from aqueous solution using batch technique. In this study we used rice husk as adsorbent for the removal of Losartan drug from aqueous solution. For this purpose, the activated carbon was prepared by the activation of rice husk which was an agricultural waste. At 5ppm concentration, dosage of 0.5g (500mg) and at 85°C temperature, the maximum percentage removal of losartan was found to be 86 %. Hence, we noticed that rice husk showed maximum removal of respective antihypertensive drug. The impact of adsorbent dose, contact time, stirring speed, pH, and temperature on the performance of the adsorption process was then investigated using the losartan. Removal percentage of respective drug were nearly same at pH 3 and pH 4. Over time, it was found that the rice husk's adsorption capability increased. The Removal efficiency of losartan drug was calculated with the help of UV- spectrophotometer and the prepared absorbent was characterized with the help of FTIR. An inexpensive and environmentally friendly absorbent was developed for the current investigation in order to look into the removal of pharmaceutical drug residues from wastewater. This study suggests that rice husk is the most effective low-cost adsorbent for removing losartan.

Keywords: Pharmaceuticals, Antihypertensive, Losartan, Adsorption, Adsorbent.

Introduction

The primary resource on Earth is water, and scientists are always working to guarantee its quality and safety [1]. Approximately 71% of the water on Earth is only 2.5% pure [2]. Life and the ecosystem depend on freshwater. Among the freshwater resources are rivers, ponds, lakes, groundwater, and streams [3, 4]. However, these resources are being depleted as a result of the ongoing population boom, fast industrialization, climate change, and interannual climate variability [5, 6].

Emerging pollutants that can have an impact on the ecosystem, such as pesticides, synthetic fertilizers, chemical compounds like dye, pharmaceuticals, heavy metals, hormones, personal care products, and detergents, are either directly or indirectly entering the aquatic system through the water supply system. These new pollutants are released from hospitals, sewage systems, research labs, and other food industries [7]. Even at low concentrations, the presence of these new pollutants in water could have detrimental toxicological effects [8].

In 2007, twelve thousand human medicines were discovered worldwide. The environment contains 850 active medicines [9, 10]. Various pharmaceutical classes, including analgesics, antidepressants, antihypertensives, contraceptives, antibiotics, steroids, and hormones, have been found in water bodies at concentrations ranging from ng/L to μg/L. Surface water, groundwater, household water, municipal waste water, and industrial water effluents have all been found to contain these pharmaceutical compounds [11].

Antihypertensive medication use has rapidly increased. In 2017, over 1.4 million antihypertensive prescriptions were written in the US [12]. A medication called losartan lowers the risk of heart attacks and strokes while treating hypertension [13, 14]. Human health issues such hypotension, kidney failure, kidney stone development, liver failure, and hyperkalemia are caused by an overdose of the antihypertensive medication losartan [15, 16]. These antihypertensives have been found in seas, rivers, and wetlands at concentrations of 0.60–08.70 ng/L, 149 ng/L, and 22.9 ng/L [17]. They are also found in surface water and wastewater effluents at environmentally relevant concentrations ranging from ng/L to mg/L. The typical biological process does not fully break down losartan [18]. Losartan may have negative effects on aquatic organisms, including cytotoxicity in the gills of brown mussels (Perna perna), fish (Astyanax altiparanae), and DNA damage in crustaceans (Daphnia Magna) [19, 20].

A variety of techniques, including membrane filtering, advanced oxidation, ozonation, fenton process, biodegradation, photocatalysis, reverse osmosis, and adsorption, have been employed to remove the drug from tainted water [21]. Adsorption is an inexpensive and environmentally beneficial physical remediation technique. In order to remove different contaminants from environmental media, different adsorbents, such as synthetic, natural, inorganic, and semi-synthetic adsorbents, have been used [22, 23].

In order to clean up the environment, natural organic adsorbents that are inexpensive, unconventional, and made from biomass from agricultural waste such as cotton fibers, rice husks, wheat husks, and kapok fibers as well as fruit and vegetable waste such as banana, orange, kiwi, and lemon peels have been employed [24, 25]. Activated carbon's processing and precursor material determine its physical and chemical properties [26].

This study focuses on an adsorbent that was made from agricultural waste and is used to remove pharmaceutical impurities. The manufacture of the adsorbents, operational parameters in the adsorption studies, such as pH, adsorbent dosage, contact time, and absorbance, and their effects on the adsorbent's absorption capability will all be covered.

Materials and Methods

Material

Losartan ($C_{22}H_{22}ClN_6O$) with purity, Sodium Hydroxide (NaOH), Sodium Chloride (NaCl), Hydrochloric acid (HCl), Rice husk.

Adsorbent preparation

The husk of the rice was gathered from the marketplace. Dust and other surface contaminants were removed from the collected rice husk (RH) by washing it with water. After washing, the rice husk (RH) was left to dry in the sun until its weight remained consistent. For 50 minutes, 200g of rice husk was thermally activated at 80±5^{0}C in a muffle furnace. Following activation, rice husk was crushed using a mortar and pestle and sieved to a mesh size of 32. Demineralized water was used to rinse the activated rice husk (ARH) until it was colorless. A filtrate was produced. The cleaned ARH was then dried for approximately 12 hours at 100±5^{0}C in a hot air oven. ARHs were stored in a desiccator for further use after drying.

Proximal analysis of adsorbent

Moisture content

The sample was heated in muffle furnace at 103±2^{0}C until constant weight. The percentage of moisture content was calculated as follows.

% Moisture content for rice husk =

$$\frac{\text{Initial weight} - \text{Final weight}}{\text{Initial weight}} \times 100$$

% Moisture content for rice husk powder =

$$\frac{\text{Initial weight} - \text{Final weight}}{\text{Final weight}} \times 100$$

Volatile content

The sample was heated in muffle furnace at 650^{o}C for 10 minutes. The percentage of volatile content was calculated as follows:

% Volatile content =

$$\frac{\text{Initial weight} - \text{Final weight}}{\text{Final weight}} \times 100$$

Ash content

Ash content was determined by heating the sample at 750^{o}C for 3 hours. The percentage of ash content was calculated as follows:

% Ash content =

$$\frac{\text{Initial weight} - \text{Final weight}}{\text{Final weight}} \times 100$$

Fixed carbon

The fixed carbon was calculated as follows:

% Fixed carbon = 100 - (% of Volatile content + % of Ash content)

Adsorbate preparation

The solution of losartan was prepared by dissolving 2.5mg and 5mg for 5 ppm and 10 ppm respectively in 500ml distilled water. Then, the pH of solution was adjusted at pH 4 and pH 9 using 0.1N NaOH/HCl solutions with the help of pH meter. After that the initial concentration of losartan was determined by finding out absorbance of the characteristic wavelength (233nm) using double beam UV-visible spectrophotometer, Thermo Fisher Scientific, Shanghai, China.

Activation of adsorbent

The ARH was placed in the muffle furnace and rising the temperature from 30^{o}C to 50^{o}C within 10 minutes. It is stand for one hour and temperature recorded as 100^{o}C.

Batch adsorption studies

A 500ml beaker was taken to carry out the batch adsorption experiment with 200ml adsorbate solution of 5 ppm and 10 ppm concentration. The pre-determined amount of adsorbent 0.5g was added to prepared solutions. After that the beaker were placed in the temperature controlled magnetic stirrer at constant speed. The effect of variation of different parameter like pH (4 and 9), concentration of the solution (5 ppm and 10 ppm), contact time (1, 5, 10, 15, 20, 25, 30min.) at temperature 30 to 35^{o}C and adsorbent dosage 0.5g. After different time intervals, 5ml of the sample were withdrawn from the beaker and the adsorbent were separated from the solution by microfilter. The absorbance of the filtered solution was estimated to determine the residual concentration measured at λ_{max} =233nm spectrophotometrically using UV- visible spectrophotometer. The amount of the adsorbate adsorbed at equilibrium condition q_e(mg/g) was calculated using the following equation:

$$q_e = (C_o - C_e)\ V/m$$

where C_o and C_e are the initial and equilibrium adsorbate concentration (mg/L) respectively V is volume of the solution (ml) and m is the mass of adsorbent (mg). The drug removal percentage can be calculated as follows.

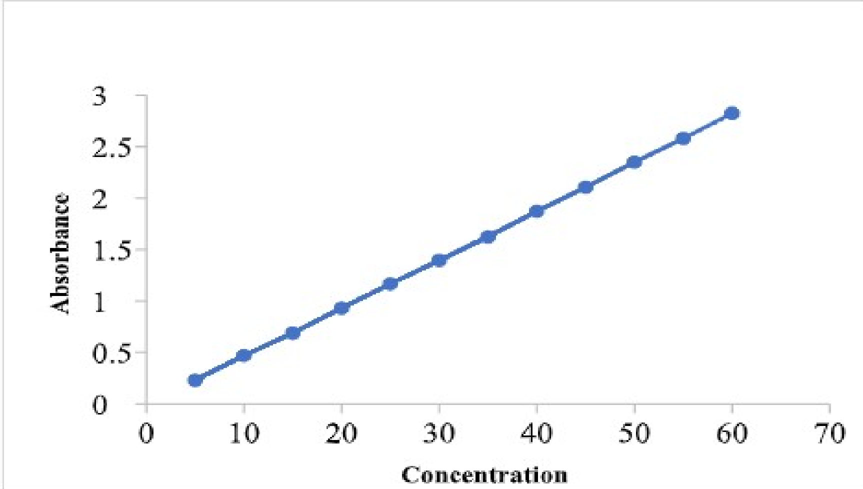
$$\%\ \text{of removal} = (C_o - C_e)\ /C_o \times 100$$

Plotting calibration curve

At first, the stock solution of losartan was prepared. The stock solution of losartan was prepared by dissolving 10mg drug in 100ml water for given concentration 100ppm. The successive dilution of the stock solution was diluted with different concentration at 5mg/L, 10mg/L, 15mg/L, 20mg/L, 25mg/L, 30mg/L, 35mg/L, 40mg/L, 45mg/L, 50mg/L, 55mg/L and 60mg/L and their absorbance were found out by UV-Spectrophotometer (λ_{max} =233nm). With these values a standard calibration curve was plotted. The equation of the curve was used to calculated the concentration for various absorbances.

Table 1: Absorbance of losartan at different concentration.

Concentration (ppm)	Absorbance
5	0.230
10	0.470
15	0.688
20	0.930
25	1.166
30	1.395
35	1.622
40	1.873
45	2.105
50	2.350
55	2.580
60	2.824
65	2.935



Results and Discussion

Adsorbent Characterization:

In this study the physical parameters of rice husk (RH) and activated rice husk (ARH) such as moisture content, volatile content, ash content and fixed carbon was determined by using muffle furnace. The values of various parameters shown in table no.2.

Table 2: Physical parameters of adsorbent.

Material	% Moisture content	% Volatile content	% Ash content	% Fixed carbon
RH	1.13	0.36	0.37	99.24
ARH	0.20	1.52	1.61	96.87

Effect of contact time and pH with different concentration on adsorption capacity and removal efficiency:

The amount of losartan absorbed by activated rice husk was studied as a function of adsorbate - adsorbent contact time at different concentration (5 and 10ppm) and pH (4 and 9) of losartan. Rate of removal of drug were observed at various contact time 't' (1, 5, 10, 15, 20, 25, 30min.). In this process, the maximum adsorption capacity and percentage removal was found at 20min. for 5ppm at pH 4 and pH 9, at 25min. for 10ppm at pH 4 and pH 9. The effect of contact time and different initial concentration of losartan on adsorption capacity (q_e) and removal efficiency are shown in table (3, 4, 5, 6) and figure (2, 3, 4, 5).

Table 3: Effect of contact time and pH with 5 ppm concentration on adsorption capacity and removal efficiency [At optimum condition pH 4].

Time	Absorbance	Final concentration C_e (mg/L)	Adsorption capacity q_e (mg/g)	% Removal
1 min.	0.184	4.094	0.092	5.38
5 min.	0.073	1.863	0.987	56.98
10 min.	0.045	1.125	1.280	74.00
15 min.	0.023	0.764	1.426	82.35
20 min.	0.021	0.700	1.452	83.83
25 min.	0.071	1.762	1.027	59.31

C_o = 4.331, (Absorbance = 0.192)

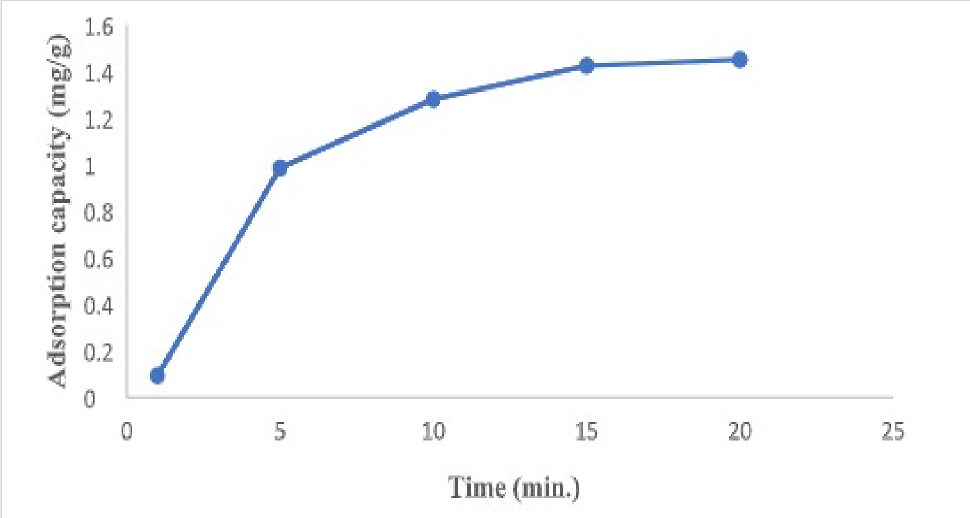


Fig 2. Adsorption capacity of losartan adsorbed by adsorbent as a function of time.

Table 4: Effect of contact time and pH with 10 ppm concentration on adsorption capacity and removal efficiency [At optimum condition pH 4].

Time	Absorbance	Final concentration C_e (mg/L)	Adsorption capacity q_e (mg/g)	% Removal
1 min.	0.250	5.605	0.543	19.50
5 min.	0.231	5.371	0.636	22.86
10 min.	0.216	4.840	0.849	30.48
15 min.	0.173	3.885	1.231	44.20
20 min.	0.110	2.738	1.690	60.67
25 min.	0.099	2.356	1.842	66.16

C_o = 6.963, (Absorbance = 0.316)

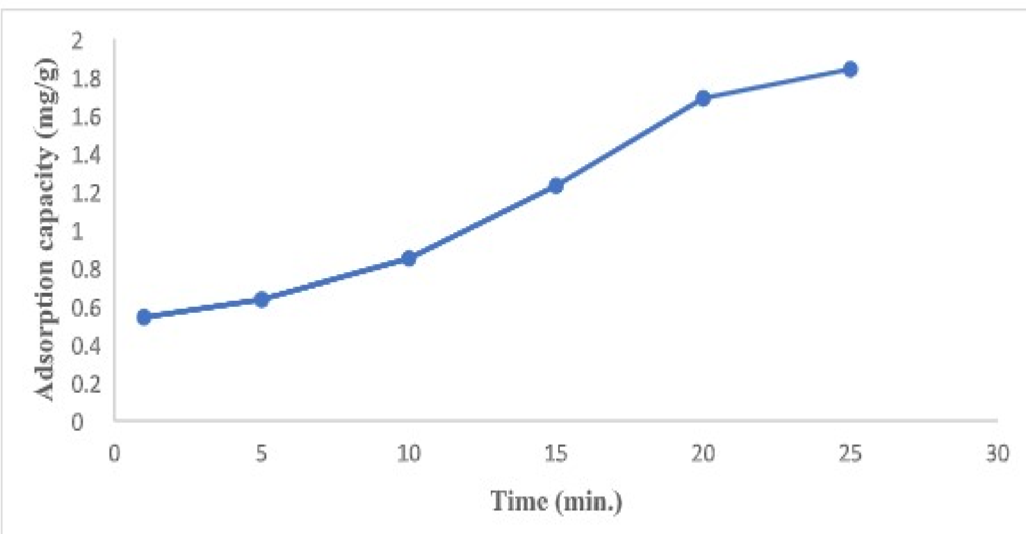


Fig 3. Adsorption capacity of losartan onto adsorbent as a function of time.

Table 5: Effect of contact time and pH with 5 ppm concentration on adsorption capacity and removal efficiency [At optimum condition pH 9].

Time	Absorbance	Final concentration C_e (mg/L)	Adsorption capacity q_e (mg/g)	% Removal
1 min.	0.176	3.864	0.772	33.33
5 min.	0.119	2.738	1.223	52.76
10 min.	0.106	2.590	1.282	56.86
15 min.	0.100	2.484	1.324	57.14
20 min.	0.115	2.696	1.240	53.48
25 min.	0.117	2.738	1.223	52.76

C_O= 5.796, (Absorbance = 0.261)

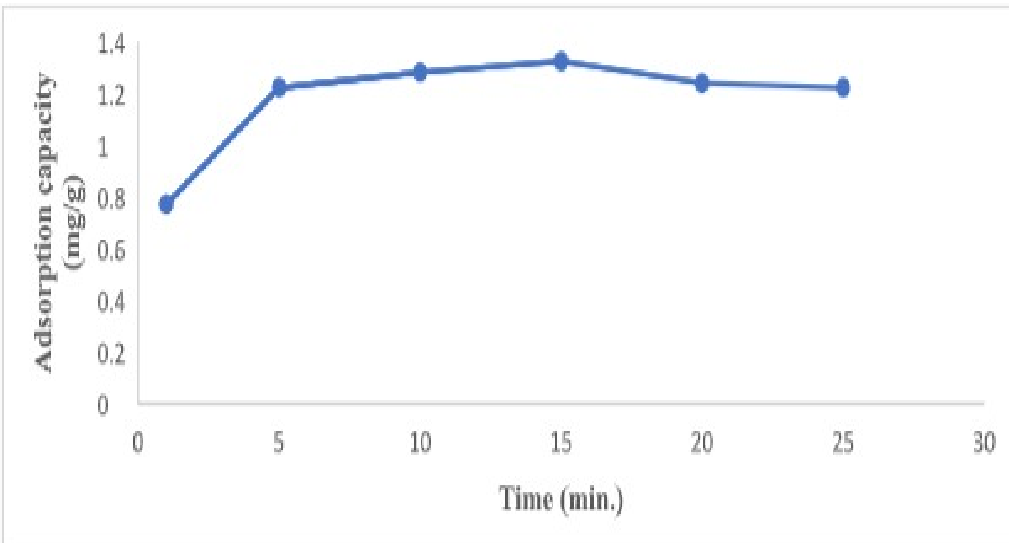


Fig 4. Adsorption capacity of losartan onto adsorbent as a function of time

Table 6: Effect of contact time and pH with 10 ppm concentration on adsorption capacity and removal efficiency [At optimum condition pH 9].

Time	Absorbance	Final concentration C_e (mg/L)	Adsorption capacity q_e (mg/g)	% Removal
1 min.	0.260	6.029	1.681	41.08
5 min.	0.237	5.265	1.987	48.54
10 min.	0.206	4.692	2.216	54.14
15 min.	0.172	3.927	2.522	61.62
20 min.	0.132	3.014	2.887	70.54
25 min.	0.113	2.696	3.014	73.65

C_0 = 10.233, (Absorbance = 0.470)

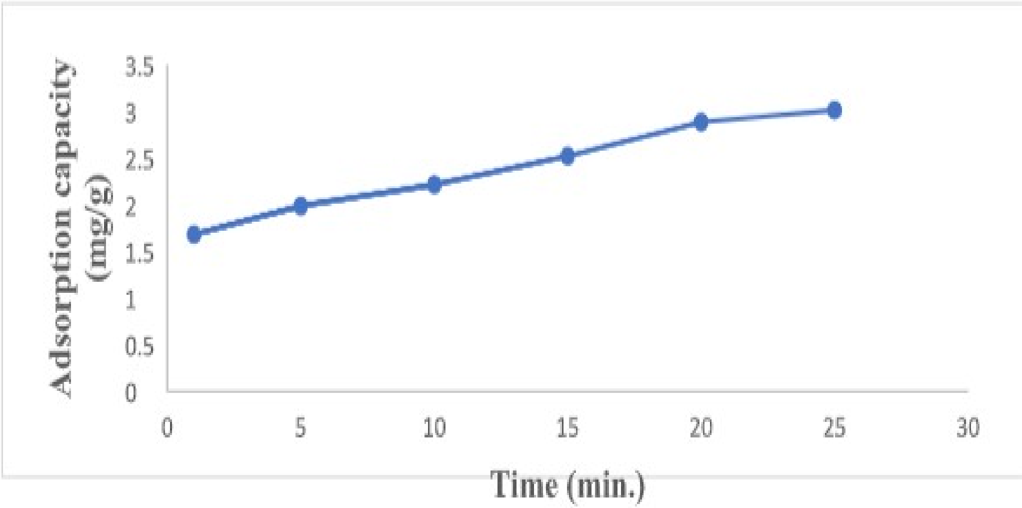


Fig 5. Adsorption capacity of losartan onto adsorbent as a function of time.

Conclusion

Removal of losartan from aqueous solution by adsorption process using rice husk adsorbent has been experimentally determined and the following observation are made:

From the above experiment, Proximate analysis showed good carbon content by adsorption which is show in table 2. Removal efficiency of the losartan decreases with increases pH and the concentration of the solution. Maximum adsorption took place at pH=4. Using the adsorbent mass of 500mg and concentration of solution is 5mg/L. the recorded percentage removal efficiency is 83.83% within 20minutes.

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