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COMPARATIVE STUDY OF LEVOCETIRIZINE ELIMINATION BY PRISTINE AND POTASSIUM PERMANGANATE MODIFIED ACTIVATED CHARCOAL

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ABSTRACT: The elimination capacity of pristine powder activated charcoal PAC and powder activated charcoal modified with potassium permanganate MPAC to remove levocetirizine drug was investigated. The activated charcoal was impregnated with $KMnO_4$ 100 mg/L in order to improve the surface properties. The influence of concentration, time and pH on the adsorption of levocetirizine drug was conducted by means of UV-Vis spectroscopy. The experiments were carried out using two lines. Line 1; adsorption of the drug on pristine PAC and line 2; adsorption on MPAC. By batch experiments, the effect of levocetirizine concentrations of 25, 50, 75, and 100 mg/L, a reaction time of 15, 30, 45 and 60 min, and pH of 2, 4, 7 and 9 were investigated. The results showed that elimination capacity of both PAC and MPAC increased with increasing the drug concentration and time of contact and decreased with decreasing the pH. The elimination efficiency of MPAC found to be greater than that of PAC.

INTRODUCTION: Activated charcoal is one of the main effective adsorbents which has a high surface area, large pore volume, tunable pore size, chemical stability and high hydrophobicity [1, 2, 3]. It is in the shape of a fine black odorless, tasteless, microcrystalline powder [4]. Due to its high adsorptive capacity is used as an adsorbent for a large variety of compounds. It is used for removal of colors from solutions [5, 6]. Activated charcoal is used in the adsorption of gases and vapors such as ammonia, carbon tetrachloride, methane, nitrogen dioxide and carbon dioxide [7, 8, 9]. A large number of studies concern the adsorption of a verity of organic compounds on activated charcoal have been conducted.

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From these studies adsorption of phenol, diethyl phthalate, dibutyl phthalate and many other compounds [10, 11, 12]. In the pharmaceutical field, activated charcoal has been used as an antidote for the majority of poisons because of its ability to prevent the absorption of many toxic agents and enhance the removal of many absorbed drugs. It is used as an antiflatulent, and as a good treatment to reduce blood lipid concentrations in patients having uremia and diabetes [13], adsorption of odors from wounds [14].

Activated charcoal has been used as an adsorbent for a wide range of different drugs like paracetamol [15], tramadol hydrochloride [16], diazepam [17], acetaminophen [18] and many others. In recent years, many researchers have dedicated to research on modification of activated charcoal to improve the adsorption capacity [19, 20, 21, 22]. One of the important methods is to oxidize the structure by oxidizing agents [23, 24]. Oxidization with ozone [25], concentrated nitric acid [26, 27] and hydrogen peroxide [28] can enhance the adsorption of functional groups

of carboxylic acids and some other organic compounds. It was found that oxidizing the structure of activated charcoal change its inert and hydrophobic nature and enhance the wettability for polar solvents [29].

Granular activated charcoal modified with potassium permanganate was used to enhance the adsorption capacity for phenol. It was found that modification experiment increases the efficiency of activated charcoal adsorption by 20% [30]. Modification with potassium permanganate increases the removal efficiency of activated charcoal for removal of Cr(VI) [31, 32] and Cu^{2+} [33] from aqueous solution. The effect of oxidation of the organic matter by potassium permanganate before adsorption on activated charcoal and oxidizing activated charcoal and then adsorbing organic matter was also investigated [34]. Some researchers have found that permanganate is not adsorbed on the activated charcoal but reduced. The reduction process was found to be strongly pH dependent, and the product of reduction is mainly Mn^{2+}. The Mn^{2+} ion adsorbed onto the activated charcoal particles by surface complexation [35]. A study of adsorption of levofloxacin on granular activated carbon produced from date (*Phoenix dactylifera* L.) stones showed adsorption capacity of 100.3 mg/g [36].

The removal of levofloxacin by means of alumina-doped coconut coir charcoal impregnated with alumina nanoparticles showed that the adsorption isotherm data were best represented by both Langmuir and Temkin isotherm [37]. Activated charcoal produced from sugarcane bagasse removes 37% from levofloxacin drug in 90 min reaction time. The reaction isotherm follows Langmuir model [38]. The adsorption of levofloxacin drug on goethite has been studied. The study concern the effects of nitrate, sulfate, small organic acids and humic acid on the adsorption process. It was shown that the presence of nitrate or sulfate had no effect, but the presence of humic acid increase the process [39]. The adsorption of levofloxacin on activated charcoal produced from rice husk was found to be spontaneous and endothermic while the adsorption on activated charcoal produced from a wood chip is spontaneous and exothermic [40]. A waste produced from corn bracts and later modified using zirconium cations were used for removal levofloxacin drug from waste water. The waste exhibited 73 mg/g adsorption capacity [41]. The adsorption of the gliclazide drug on activated charcoal was found to be pH, concentration and time-dependent [42]. The aim of this research is to compare the elimination levels of levocetirizine by pristine and modified activated charcoal and to study the effects of concentration, time and pH on the process.

MATERIALS AND METHODS: The activated charcoal used was obtained from Sigma-Aldrich, after repeatedly washed with distilled water and dried to constant weight at 105 ºC for over 3 h. The drug sample of levocetirizine is a production of TDA Pharma. All chemicals used in this work were of analytical grade obtained from Sigma-Aldrich.

The electronic spectra were recorded on PG instruments T80 UV/Vis double beam spectro-photometer. The pH was monitored using a Philips (PW-9409) digital pH meter.

The wavelength at which maximum absorbance occurs (λ_{max}) was recorded for the aqueous solutions of drug and found to be 230 nm.

An exact weight of 50 mg of levocetirizine was dissolved in 250 ml distilled water in a 500 ml volumetric flask. The solution was shaken to dissolve all the drug partials and the volume was adjusted up to the mark of the flask with distilled water to obtain a stock solution of 100 mg/L. The solution was filtered using Whatman filter paper no. 41.

Modification Method of Activated Charcoal: Modification of activated charcoal was conducted by immersing the powder in a solution of potassium permanganate 100 mg/L under a neutral condition for 4 h with slow stirring. The mixture then separated and washed repeatedly with distilled water to no color and dried to constant weight at 105 ºC for 4 h in order to remove the content of impregnation solvent and water.

Adsorption Studies: Adsorption experiments were performed to study the effects of important parameters such as concentration, time and pH. The experiments were performed on both pristine PAC and MPAC separately.

The flask containing 50 ml of levocetirizine solution of concentrations 25, 50, 75 and 100 mg/L were shaken with 0.5 gm activated charcoal for 1 hour for concentration study.

For time study, 50 ml of drug solution 100 mg/L were shaken with 0.5 gm activated charcoal for 15, 30, 45 and 60 min.

For pH study, 50 ml of drug solution 100 mg/L were shaken with 0.5 gram of activated charcoal for 1 h at pH of 2, 4, 7 and 9. The mixtures were shaken vigorously at room temperature on magnetic stirrer after which filtered through filter paper and the filtrate solutions were analyzed spectrophotometrically at λ = 230 for residual drug concentration. The process was repeated for PAC and MPAC under different equilibrium conditions of concentration, time and pH. The pH was adjusted using 0.1 N solutions of HCl and NaOH.

The amount of the drug adsorbed was calculated using the equation:

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

Where Q_e is the maximum quantity of the drug in mg/gm adsorbed on the activated charcoal, C_o is the initial concentration (mg/L) of the drug solution, C_e is the concentration of the drug (mg/L) in the supernatant at the equilibrium stage, V is the volume of the drug solution in liter and m is the mass of adsorbent employed in grams.

The calibration curve was conducted by measuring the absorbance of solutions for each experiment, then plotting the absorbance versus concentrations, time and pH.

RESULTS AND DISCUSSION: Levocetirizine is a selective H_1-antihistamine drug used for the treatment of allergic rhinitis and chronic idiopathic urticaria. It is an off-white crystalline powder soluble in water. The chemical formula is $C_{21}H_{25}ClN_2O_3$. It is the *R*-enantiomer of racemic cetirizine. The chemical name of the drug is 2-[2-[4-[(*R*)-(4-chlorophenyl)-phenyl methyl] piperazinyl-1-yl]ethoxy] acetic acid [43, 44].

The chemical structure of the drug is shown in **Fig. 1**. The UV spectrum of the levocetirizine solution is shown in **Fig. 2**.

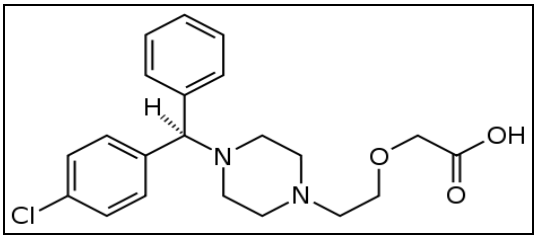

FIG. 1: THE CHEMICAL STRUCTURE OF LEVOCETIRIZINE

The results of concentration for both PAC and MPAC study are listed in **Table 1**.

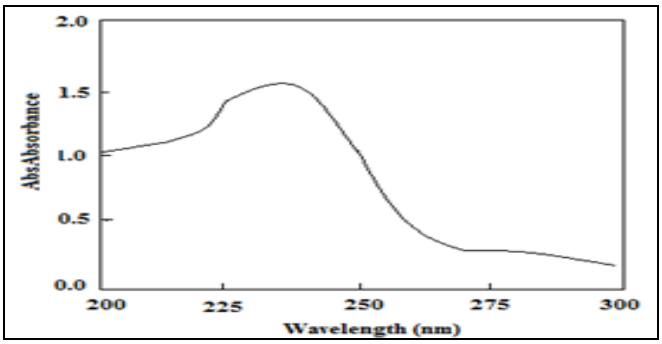

FIG. 2: THE UV SPECTRUM OF LEVOCETIRIZINE

TABLE 1: PARAMETERS OF CONCENTRATION STUDY

Concentration (mg/L)	Levocetirizine Adsorbed on PAC (mg/g)	Levocetirizine Adsorbed on MPAC (mg/g)
25	1.1	1.8
50	2.2	2.8
75	3.2	3.8
100	4.7	4.9

To simulate the adsorption process in the environment of the gastric fluid a sample was used with its additives.

It is clear from **Table 1** that the amounts of the adsorbed drug in mg for each gm of both PAC and MPAC increases almost steadily with increased concentration of the drug solution. At the same time, it seems clear that MPAC adsorbs larger amounts than PAC. The amounts of 4.7 and 4.9 mg of the levocetirizine adsorbed on 1 gm PAC and MPAC respectively in the 100 mg/L concentration seem reasonable.

Fig. 3 is a plot of the amounts of the drug adsorbed on activated charcoal in mg/gm versus concentration for concentration study. The steady increscent in the amount of the levocetirizine drug adsorption on PAC and MPAC is very obvious from the plot.

The parameters for time study are listed in **Table 2**. When the time duration of the reaction was changed from 15, 30, 45 to 60 min, the amounts of the drug adsorbed increased with increasing the time for both PAC and MPAC. This time incensement gives the indication that the time of contact is also an important factor for the elimination of the drug. Again the adsorption rate of MPAC was greater that of PAC. The enhancement of adsorption when MPAC is used is very obvious. The relationship between time and the amounts of the adsorbed drug is shown in **Fig 4**. The steady increscent of the adsorption with increasing time is clear.

TABLE 2: PARAMETERS OF TIME STUDY

Time (min)	Levocetirizine Adsorbed on PAC (mg/gm)	Levocetirizine Adsorbed on MPAC (mg/gm)
15	3	4
30	3.2	4.3
45	3.9	5
60	4.2	5.4

Table 3 contains the parameters for pH study. The amounts of the levocetirizine in mg adsorbed on 1gm activated charcoal increasing as the pH of the solution decreased. The highest values of 5.4 and 6.6 mg/gm for the PAC and MPAC respectively found at pH 2, which seems a good result since the pH of the gastric fluid in this rang. The relationship between the pH and the amounts of the adsorbed drug gave a straight line as shown in **Fig 5.** Only the value at pH 7 for the PAC showed deviation from the straight line.

TABLE 3: PARAMETERS OF pH STUDY

pH	Levocetirizine Adsorbed on PAC (mg/gm)	Levocetirizine Adsorbed on MPAC (mg/gm)
2	5.4	6.6
4	4.3	5.8
7	4	4.5
9	2.5	3.3

From the parameters of the three factors, it is quite clear that the elimination of the levocetirizine drug from the solutions depends on concentration, time and pH. Modification of the activated charcoal by potassium permanganate enhance its elimination capacity in all the experiments factors used.

The enhancement capacity reaches 63% for the 25% concentration as is evident from **Table 4**. A quick glance at the table reveals that the MPAC adsorption capacity was enhanced by about 30% in general.

TABLE 4: % OF ADSORPTION CAPACITY ENHANCEMENT IN MPAC

% Adsorption Enhancement in concentration study	25 63%	50 27%	75 18%	100 4%
% Adsorption Enhancement in Time Study	15 33%	30 34%	45 28%	60 28%
% Adsorption Enhancement in pH Study	2 22%	4 34%	7 12%	9 32%

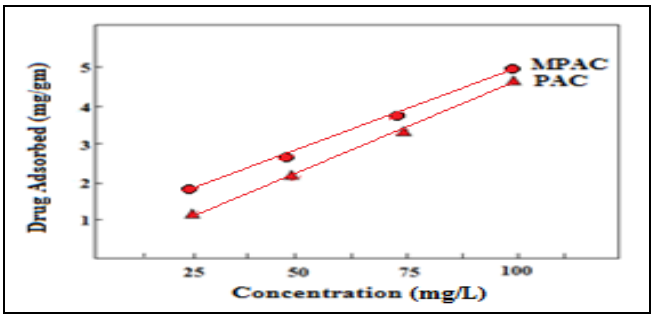

FIG. 3: PLOT OF ADSORBED DRUG (mg/gm) VERSUS CONCENTRATION FOR CONCENTRATION STUDY

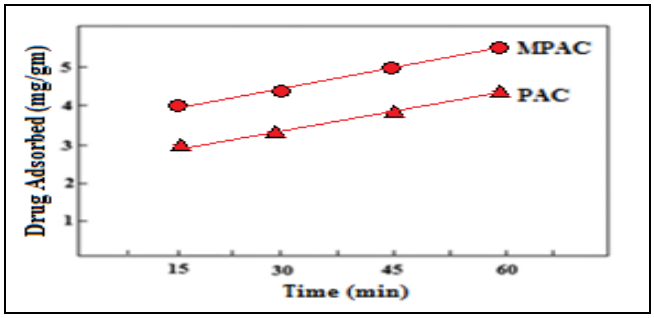

FIG. 4: PLOT OF ADSORBED DRUG (mg/g) VERSUS TIME FOR TIME STUDY

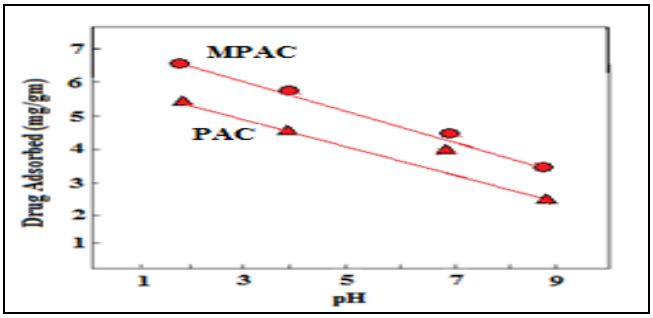

FIG. 5: PLOT OF ADSORBED DRUG (mg/g) VERSUS pH FOR pH STUDY

CONCLUSION: As a comparative investigation, powder activated charcoal PAC and powder activated charcoal impregnated with potassium permanganate MPAC were used to eliminate the levocetirizine drug from solutions. The process include different concentrations (25, 50, 75 and 100 mg/L), different times (15, 30, 45, and 60 min) and different pH (2, 4, 7, and 9). It was found that the elimination rate depends on the concentration, time and pH. The greater the concentration and time the greater the elimination capacity, and the lower the pH the greater the elimination capacity. The enhancement in elimination capacity of MPAC by about 30% was evident in all the variable factors of the experiments.

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