



Effective Granular Activated Carbon for Greywater Treatment Prepared from Corncobs

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Abstract

Human daily activities increase water consumption, which increases the quantities of wastewater. The produced wastewater must be treated thoroughly before discharging to protect the environment. Adsorption technique is one of the most common methods of treating water and wastewater. Graywater is resulting from human uses, as it is a mixture of bathroom and kitchen drainage. The main pollutants in greywater are oils&greases, total suspended solids (TSS), chemical and biological oxygen demand (COD and BOD₅), nitrogenous compounds, and phosphorus pollutants. This study aimed to verify the effectiveness of bio-adsorbents and the granular activated carbon produced from corncob as an agricultural waste. The corncob was converted into granular activated carbon. The removal efficiency of adsorbents depends on several factors such as contact time, the doses of adsorbents, pH of a solution, and depth of column bed. The maximum percent removal attained at the 120 min, 0.8 g/L of adsorbent. The results indicated that the second-order kinetic model gave a good fit of the data for adsorption of pollutants onto the GACC. The values of R² were very well in the pseudo-second-order kinetic model, The R² was 0.893, 0.986, 0.996, and 0.961 for COD, TSS, TKN, and phosphorus, respectively. Furthermore, the breakthrough study was conducted using GACC. The removal efficiency (R %) of GACC for COD, TSS, TKN, and T.P at 4 h were 88.18, 81.14, 87.11, and 99.57, respectively. The overall results in case study for greywater treatment by using GACC were complying with the Egyptian Environmental Association Affair (EEAA) limits.

Keywords; greywater; adsorption techniques; granular activated carbon; vertical column bed.

1. Introduction

The presence of water in the universe is necessary for the survival of humans. The amount of toxic pollutants increased as a result of industrialization as well as the increase in human activities. Due to the presence of many pollutants such as organic pollutants, pesticides, and, metals the water is unfit for human consumption. This affects the human health and the aquatic environment [1]. Greywater is generated in houses and offices without the presence of fecal contamination, kitchen drains, bathrooms, and laundry. Bathroom samples are represented by the drainage of showers and baths. Samples drawn from the greywater drainage are contaminated with soap, hair shampoos, and toothpaste. The kitchen drains are contaminated with food particles, cooking oils&grease, and dishwashing. The amount of

harmful pathogens in greywater is considerably lesser when compared to domestic sewage. Generally, greywater is safer to handle and simpler to treat. The treated greywater can be reused for flushing purposes, landscaping or crop irrigation, and other non-potable uses[2].

There is a large number of treatment methods that are used for the treat of wastewater generated from various industries like active sludge, bioremediation, photo-degradation, microfiltration, coagulation-flocculation, Fenton-oxidation, adsorption, and electrocoagulation. Among these, coagulation-flocculation has considerable attention for its high removal efficiency [1]. The adsorption technique has become a subject of considerable interest, due to the advantages it provides, such as the possible utilization of different materials as adsorbents, cheap

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technological steps, easy maintenance, no special operational skills, and local availability. Among the materials used as sorbents for pollutants removal, might be made of activated carbon, ion exchangers, polyamides, polyurethane foams, inorganic materials, and, more recently, natural lignocellulosic materials. The high cost of activated charcoal, the most commonly used sorbent, has redirected the research towards cheaper and more available materials with sorption properties. Within this context, the corncob as "non-conventional" or "low-cost" adsorbent was used [3]. The corncob has no significant re-use and even animals avoid eating it. Farmers resort to burning the corn cob for fuel, which contributes to air pollution and global warming. As a result of rigidity and high porous structure, corncobs possess adsorption properties [2,3]. The corncobs consist of 44.8 % cellulose, 46.1% hemicellulose, and 9.1% lignin [4, 5]. Cellulose is the β -1,4-polyacetal of cellobiose, which is considered as the polymer of glucose. The common polymer of hemicellulose is xylan which is mainly composed of five-carbon sugar monomers such as xylose and arabinose, and six-carbon sugar monomers such as glucose, mannose, and galactose. Lignin is a complex polymer that consists of three types of phenolic acids, coniferyl alcohol, and synapse alcohol. It plays an important role in the cell's endurance and development, as it affects the transport of water, nutrients, and metabolites in the plant cell [6]. In previous studies, it has been noted that many pollutants such as detergents, colored dyes, salts, suspended solids, oil&grease, and heavy metals adsorbed onto the active sites of corncobs. Corncob waste can be converted into activated carbon taking a step towards biomass utilization and bioresource recycling. The activated carbon thus prepared has an appreciably high surface area and is a promising adsorbent for pollution control [2]. This study aimed to evaluate the performance of activated carbon corncob (GACC) to remove the pollutants from greywater, and valorization of the treated effluent. The aims extended to study the equilibrium sorption, dose, contact time, and kinetic data were analyzed through different models, to understand the possible sorption mechanism of the pollutants molecules on GACC.

2. Experimental

2.1 Sample sites

The greywater was collected from a site in the National Research Center (NRC), Giza, Egypt.



Figure 1: The manhole of the greywater storage tank

2.2 Analytical methods

Physicochemical characterization of influent, and the treated effluents were carried out according to the APHA. Characterizations include COD, BOD, total suspended solids (TSS), phosphates (TP), nitrates, nitrites, total Kjeldahl nitrogen (TKN), ammonia, and fecal coliform (FC) counts. All analyses were performed in complying with the standard methods of examination water and wastewater [22].

2.3 Characteristics of raw greywater

The greywater characteristics indicated that such wastewater is relatively strong as exhibited by the ammonia, COD, BOD₅, oil&grease, TSS, and phosphate. The characteristics of greywater are shown in Table 1 compared to the Egyptian Environmental Association Affair (EEAA) [7]. The COD and BOD₅ were 720 and 422 mg/L, respectively. The ratio of BOD₅/COD was 0.58. The TSS and oil&grease were varied from 438 mg/L and 114 mg/L, respectively. The total nitrogen in influent samples was 17 mg/L while, the total phosphate was 6.1 mg/L.

Table 1: Physicochemical characteristics of raw greywater

Test	Unit	*Ave. values
pH	Unit	7.40
TDS	mg/L	617
BOD ₅	mg/L	422
COD	mg/L	720
TSS	mg/L	438
Oils & greases	mg/L	114
T. phosphate	mg/L	6.10
T.N	mg/L	17

*Average for 20 samples

2.4 Preparation of adsorbents

2.4.1 Corncob (CC)

The CC was dried at 105°C for 24 h to reduce the moisture content. The dried CC was grounded

manually, then crushed with a mortar, and separated by sieving at a size ranging from 0.7 to 0.9 mm. The sieved CC was washed by distilled water (DW) many times, to remove color and dust [8].

2.4.2 Granular activated carbon (GACC)

The GACC was prepared from corncob using phosphoric acid (H_3PO_4 , 85.0 %) and boric acid (H_3BO_4 , 99.5 %) as a chemical activating agent [9]. The CC was added to the flask containing the activating agent (5.0 g/L). The pieces of CC are pre-carbonized then immersed in hydrochloric acid (HCl, 37.0 %), at 120 °C for 45 minutes, and then placed in an oven at 500 °C for 1 hour. The GACC obtained was washed several times using DW. After the pH of the solution reached neutral, the GACC was dried at the temperature of 105 °C for 24 hour [9].

2.5 Reagents

All chemicals were of analytical grade. The chemical reagents used included phosphoric acid (H_3PO_4 , 85.0%, Fischer scientific UK), potassium dichromate ($\text{K}_2\text{Cr}_2\text{O}_7$, 99.0 %, Merck, Germany), boric acid (H_3BO_3 , 99.5%, LOBA Chemie, India), hydrochloric acid (HCl, 37.0 %, Fischer scientific UK), nitric acid (HNO_3 , 70.0%), sodium hydroxide (NaOH, 99.0 %, Merck, Germany), and ammonia solution, (NH_3 , 35.0 %, Fischer scientific UK).

2.6 Instruments and characterization techniques

The instruments were: Furnace, the Drying oven (Fisher Scientific Equipment, American provisioner of scientific), Digital electronic balance (PCE-BSK 310 Instruments UK), DIW system (Millipore GER),

The pH AD110, ADWA, Hungary, Fourier-transform infrared spectroscopy (FTIR), (Thermo Fisher Scientific, UK), Orbital shakers instrument, Thermo Fisher Scientific, US), The COD digester instrument, (with auto time-controlled, MAC, India), The BOD₅ incubator, (Aircor, India), The UV-Vis Spectrophotometers, (PG Instruments, a UK company), Incubator, (Thermo Fisher Scientific, UK). Portable multiparameter water quality measurement, (HORIBA company. the USA),

Kjeldahl Digestion instrument, (ESEL, India), and The Scanning Electron Microscope (SEM) Model (Quanta 250 FEG – field emission gun – attached with accelerating voltage 30 K.V. FEI company (Netherlands).

2.7 Adsorption batch studies

The kinetic study using adsorbent was investigated. The effect of operating parameters towards adsorption efficiency such as adsorbent amount, contact time, and column test experiment, were examined. When adsorbents were added into the greywater the effects of contact time, amount of adsorbent on removal efficiency (R%), and capacity q_{max} (mg/g) of adsorbents were measured [3]. The removal efficiency (R%), the amount of on an (mg/g) were calculated from the equations (1, 2).

$$R\% = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

$$q(\text{mg/g}) = \frac{(C_0 - C_e)V_L}{m} \quad (2)$$

where C_0 is the initial concentration (mg/L), C_e is the concentration after treatment (mg/L), m is the dose of adsorbents (g), and V_L is the volume of solution (Liter)

2.8 Factors affecting the removal efficiency

2.8.1 Effect of adsorbent amount

The investigation of the effect of the amount of adsorbent on the adsorption capacity of GACC was also part of batch studies [10]. Different amounts of GACC adsorbent were studied (0.1, 0.2, 0.5, 0.8, 1.0) g [6].

2.8.2 Effect of contact time

The effect of contact time (1.0, 2.0, 3.0, 4.0, 5.0 hours) on the adsorption efficiency of the GACC adsorbent in removing pollutants, were studied [11].

2.9 Adsorption kinetics models

The kinetic models can provide information about various factors such as the dose of adsorbents, and contact time. Two kinetic models were used to explain the mechanism of the adsorption processes, which were the pseudo-first-order model and the second-order model. These models determine the relationship between changes in concentrations of the pollutants with increasing the time [3].

2.9.1 The pseudo-first-order model

The linear form of the pseudo-first-order kinetic equation derived from the equation 3 [12]. Where q and q_i are the amounts of removed pollutants (mg/g) at equilibrium and at any time t (h), respectively, and k is the rate constant of the first-order. The constants of the model can be calculated from the linear plots of $\text{Ln}(q_e - q_t)$ vs. t [13].

2.9.2 The second-order model

The second-order adsorption may be fitted with a chemisorption mechanism, involving chemical forces between sorbent and adsorbate. The second-order reaction rate can be calculated by equation 4 [13]. Where k is the rate constant of second-order sorption (g/mg.h) and q_2 , k are the constants of the model were determined from plots t/q vs. t [13].

$$\frac{t}{q} = \frac{1}{Kq_2} + \frac{1}{q \cdot t} \quad (4)$$

Where k is the rate constant of second-order sorption (g/mg. hours) and q_2 , k are the constants of the model are determined from plots t/q vs. t [13].

2.1 Breakthrough curve experiments (Vertical column test)

2.10.1 Activated carbon corncob filter layer

The inner diameter of the column is 5 cm and length 50 cm, were packed with cotton (soft, fluffy, tough, and have high permeability) as supporting material at 5 cm and added adsorbent material (GACC) with a height of 15 cm, weight of GACC was 135 g. The greywater effluents were pumped upward through the column by the flow rate of 2.5 mL·min⁻¹. The cotton layer of 50 mm was used as the base for supporting the adsorbent layers above. Cotton was washed to remove dirt and then left to dry. The granular activated carbon was the second layer. The granular activated carbon was washed well to remove the ash and then allowed to dry. The impurities in the water get trapped in the voids of adsorbent particles.

2.10.2 Testing of the filter

The activated carbon (GACC) filter was checked to evaluate the removal efficiency. The greywater sample was poured through the top of the filter at a constant rate of flow. The time taken for complete filtration was 15 minutes. The effluent was then collected in a clean container and tested for its characteristics. The sampling intervals were 0, 1, 2, 3, 4, and 5 hours. The breakthrough curve for the column was determined by plotting the ratio of the R % against the time [14].

3. Results

3.1 Efficiency of the adsorbents

Laboratory batch studies were useful in obtaining and providing fundamental kinetic data for the applied adsorbents. The importance of obtaining kinetics curves lies in developing a model, which

accurately represents the obtained results, and could be used for design purposes. Moreover, the models were applied to describe the breakthrough curve. It was considered as a key role in the scaling-up procedure from laboratory experiments through pilot plant to industrial scale.

3.2 Infrared study of corncob (CC) and granular activated carbon (GACC)

More information on CC and GACC was obtained by infrared analysis. The FTIR spectra of CC and GACC are plotted in Figures 2, and 3. The CC spectra presents many overlapping bands. A comparison between the spectra of CC and GACC show that, the occurrence of the bands at 3300 cm⁻¹ corresponds to O–H stretching vibrations that indicates the presence of hydroxyl groups, while that near 2844 cm⁻¹ depicts the C–H stretching that corresponds to the presence of alkanes [3]. The band at 2517.1 cm⁻¹ corresponds to C=C bond [15]. The occurrence of bands from 1710 cm⁻¹ indicates the presence of lignin [3]. The band at 1000 cm⁻¹ corresponds to C–O stretching. While that at 1517 and 600 cm⁻¹ showing characteristics of –N–O stretching and C–H bending, respectively [4]. These functional groups represent the chemically active components. The presence of the –OH group will initiate and accelerate the rate of condensation reactions created by dehydroxylation as a result of thermal decomposition of the cellulose content of the corncobs. While the C–H presence is due to alkanes is connected to the reactions leading to hemicellulose degradation [4]. The existence of the C=C group, which is an indication of the presence of alkenes, facilitates the reactions leading to lignin decomposition; while the group C–O, which is assigned to carboxylic groups in cellulose and hemicellulose [3,16]. Figures 2 and 3 reveal that the bands at 3300 cm⁻¹ and 1000 cm⁻¹ in GACC are higher than CC which is assigned to a high density of oxygen atom in GACC groups (–OH group, and C–O stretching), that refers to the chemically active components increased in GACC [3]. The FT-IR spectra suggest the treatment of pollutants from greywater by adsorption processes such as physical sorption pattern [3].

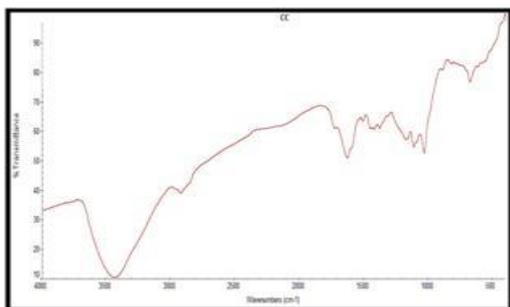


Figure 2: The FT-IR spectrum of the Raw CC

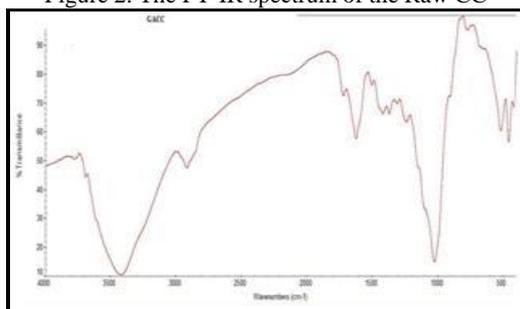


Figure 3: The FT-IR spectrum of the raw GACC

3.3 The morphology of surface by SEM

The SEM images of the GACC before and after treatment are shown in Figures 4, 5. The activated carbon corncob is soft, and pores appeared Figure 4. But the activated carbon after treatment were clear and no pore due to the pore occupied by pollutants.

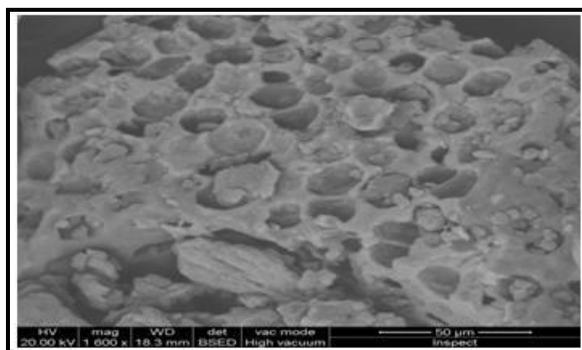


Figure 4: Scanning electron microscope images of activated carbon corncobs before treatment

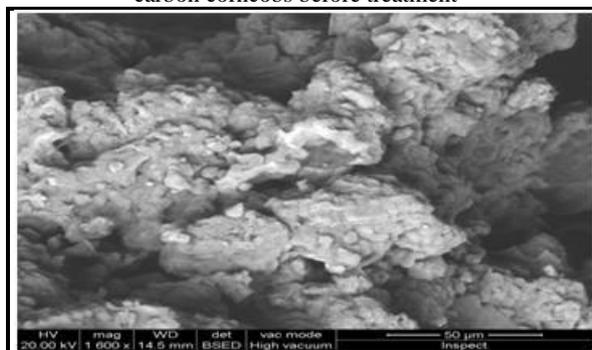


Figure 5: Scanning electron microscope images of activated carbon corncobs after treatment

3.4. Kinetics studies

Adsorption of pollutants were measured by pseudo-first-order and second-order kinetic equations. The first-order model described the sorption capacity of solids in solid-liquid systems, while the second-order model has been applied for the analysis of kinetics from liquid solutions [13]. The non-linear form of the pseudo-first-order model is given in Figures 7 and 8. While the linear form of the second-order model was shown in Figures 9 and 10 [13].

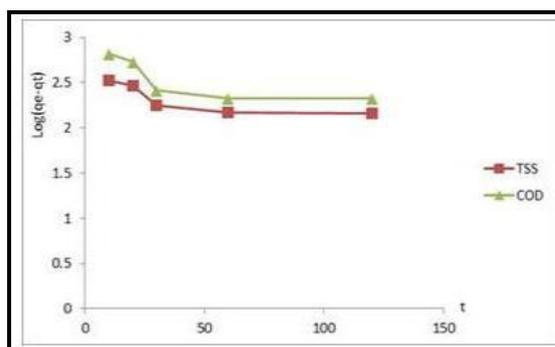


Figure 6: The pseudo-first-order kinetic models of TSS, COD using GACC.

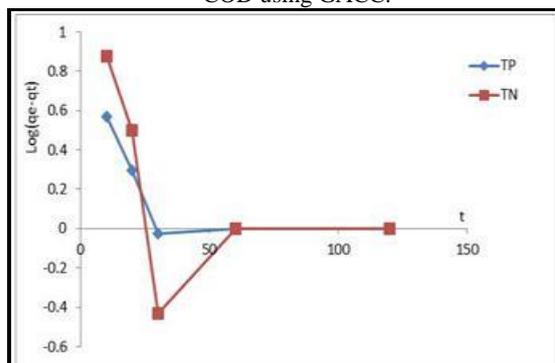


Figure 7: The pseudo-first-order kinetic models of TKN, T.P using GACC

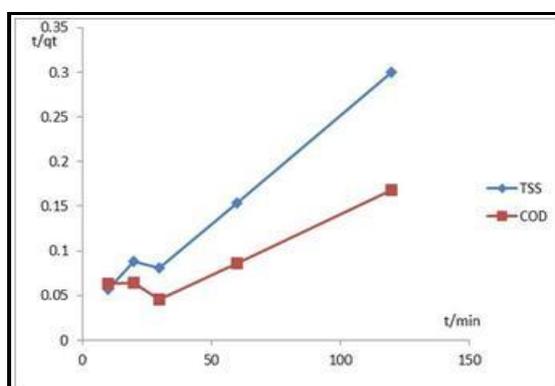


Figure 8: The second-order kinetic models of TSS, and COD using GACC

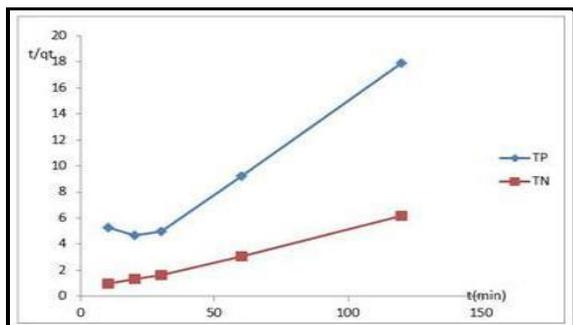


Figure 9: The second-order kinetic models of TKN, and T.P using GACC

Figures 6-9 show a fit of the sorption kinetics of pollutants at different times. The values of the rate constant and the correlation coefficient are listed in Table 2. The results indicated that the second-order kinetic model gave a good fit of the data for adsorption of pollutants onto the GACC. The theoretical values of R^2 are very well in the pseudo-second-order kinetic model. The R^2 was 0.893, 0.986, 0.996, and 0.961 for COD, TSS, TKN, and phosphorus, respectively. The adsorption of pollutants on GACC follows the pseudo-second-order kinetic model, which relies on the assumption that chemisorption may be the rate-limiting step. In chemisorption, the metals ions stick to the adsorbent surface by forming a chemical bond and tend to find sites that maximize their coordination number with the surface [16].

Table 2: The kinetic adsorption of pollutants using GACC

Pollutant	Adsorbent	R^2	
		Second-order	First-order
COD	GACC	0.893	0.57
TSS		0.986	0.61
TKN		0.996	0.193
TP		0.961	0.4

3.4.1 Effect of contact time

Figure 10 depicts the effect of contact time for the pollutants on the GACC surface. In the adsorption test, the rate of pollutants removal was relatively fast at the beginning due to the greater availability of the surface area. The pollutants sorption for GACC, shows a quick tendency to reach equilibrium. Figure 10 shows that the maximum percent of removal attained at the 60 min. While between 60 to 120 min there were no significant improvement in removal

efficiency. This occurred due to a large number of vacant surface sites available for adsorption [17], over time. The adsorbent surface becomes occupied, so it was difficult to adsorb other materials onto the surface but, when T.P is removed, the maximum removal percent was reached after 120 min. After 60 min, the COD, TKN, and TSS are remained relatively constant due to the surface becoming occupied [18]. The removal of COD, TSS, TKN, and T.P at optimum contact time is 88.40%, 88.37%, 97.05%, and 99.22%, respectively.

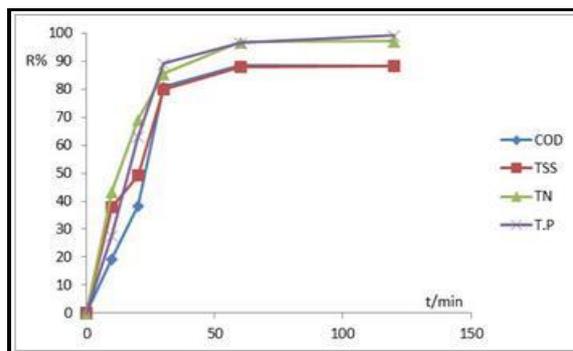


Figure 10: The effect of contact time on COD, TSS, TKN, and T.P adsorption using GACC

3.4.2 Effect of adsorbent dose

The effect of the adsorbent dose on the removal of COD, TSS, TKN, and T.P by GACC is depicted in Figure 11. The COD, TSS, TKN, and T.P removal using GACC is 88.6%, 86.4%, 87%, and 99.2%, respectively. The Increase in removal percentage is due to an increase in the adsorbent surface area as well as the active sites available [18]. Optimum doses of adsorbent (GACC) for removal of pollutants was 0.8 g. The percentages of removal for pollutants were relatively constant at 1.0 g of adsorbent due to the saturation of adsorption sites [18]. At dosages beyond the optimum, the removal rates decrease due to the destabilization of the adsorption process takes place [19, 20].

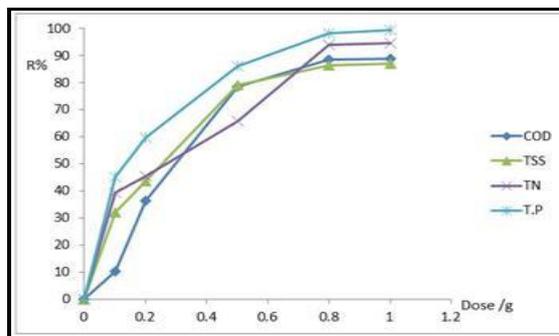


Figure 11: The effect of adsorbent dose on COD, TSS, TKN, and T.P adsorption using GACC

3.5. Continuous studies using GACC

The GACC columns were used to investigate the removal capacity of pollutants from greywater samples under continuous flow conditions. This study can throw light on the data necessary for the optimization of the design experiments of adsorbents. Different contact times (1, 2, 3, 4, and 5 h) were examined to study the effect of the adsorptive capacity of GACC. The rest of the other variables were kept constant. The concentrations of pollutants in the raw samples are determined in Table 1. The experiment for each system was carried out for 5 h. The operating conditions are given in Table 3. The removal efficiency (R %) of GACC for COD, TSS, TKN, and T.P at different contact times are shown in Figure 12 and Table 3. The removal efficiency of GACC reaches a maximum of 4 h then the R% decreases with increased contact time due to the adsorption capacity of GACC being increased by increasing the time [14]. The effect of contact time on the adsorption capacity showed the adsorption of **COD, TSS, TKN, and T.P** on GACC. It was found that the adsorption of **COD, TSS, TKN, and T.P** on GACC was reversible when desorption was undertaken after 4 h [14].

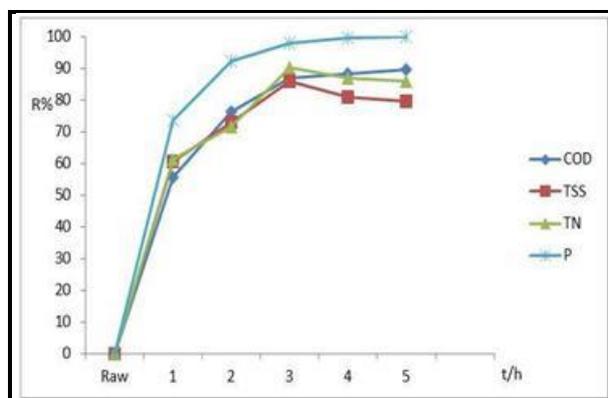


Figure 12: The breakthrough curves for COD, TSS, TKN, and T.P onto GACC

Table 3: The R % for COD, TSS, TKN, and T.P onto GACC

Time (h)	R % of Granular activated carbon corncob (GACC)			
	COD	TSS	TKN	T. phosphate
1	55.68	60.65	61.34	73.55
2	76.36	72.95	71.77	92.20
3	87	86.06	90.42	97.88
4	88.18	81.14	87.11	99.57
5	89.80	79.50	85.88	100

3.6 Case studies

Bench-scale experiments were carried out for the treatment of greywater collected from a residential household, located in Cairo, Egypt. The characterization of greywater was depicted in Table (1). Applying the optimum effects obtained from previous experiments such as contact time and adsorbent dose to obtain an optimal result for treating greywater, this was clarified in Table (4).

Table 4: Overall Treatment Processes

Pollutant	Before	After	* Limits/ EEAA (law: 48/1982)
pH	7.25	7.32	6.5-9.0
TDS	730	850	2000
COD	786	79	80
BOD ₅	456	38	40
TSS	442	35	40
TKN	28.8	0.39	15
T. phosphate	5.14	0.46	2.0
Oils & greases	76	0.025	105

* EEAA: Egyptian Environmental Association Affair

4. Conclusions

The above study proved the efficiency of the activated carbon corncob (GACC) in absorbing pollutants from greywater. This was done through the use of activated carbon prepared from the corncob to treat the use of greywater produced in homes. Through the application of many factors such as contact time, adsorbent dose, application of kinetic order equation, and also the application of a vertically packed activated carbon filter system capable of treating wastewater and reusing it for many purposes such as car washing, gardening, rinsing, and other indoor purposes. And through the results obtained, it was found that it can be said that by increasing the retention period, it is possible to generate wastewater that can be used again. From the previous discussion, grey water reuse can be seen as an effective way to reduce consumption from municipal points and water costs, without reducing the total water use in the home. The specifications of treated wastewater meet the requirements of government regulations for discharge into the sewage network. The removal efficiency of adsorbents depends on several factors such as contact time, the doses of adsorbents, and depth of column bed. The maximum percent removal attained at the 120 min is 88.40%, 88.37%, 97.05%, and 99.22%, for COD, TSS, TKN, and T.P respectively. The maximum percent removal at 0.8

g/L is 88.6%, 86.4%, 87%, and 99.2%, for COD, TSS, TKN, and T.P respectively. The results indicated that the second-order kinetic model gave a good fit of the data for adsorption of pollutants onto the GACC. The values of R^2 are very well in the pseudo-second-order kinetic model. The R^2 was 0.893, 0.986, 0.996, and 0.961 for COD, TSS, TKN, and phosphorus, respectively. Furthermore, the breakthrough study was conducted using GACC. The removal efficiency (R %) of GACC for COD, TSS, TKN, and T.P at 4 h were 88.18, 81.14, 87.11, and 99.57, respectively. The overall results in this study for greywater treatment using GACC were complying with the Egyptian Environmental Association Affair (EEAA) limits.

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