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Utilization of solid materials to remove ammonia from drinking water

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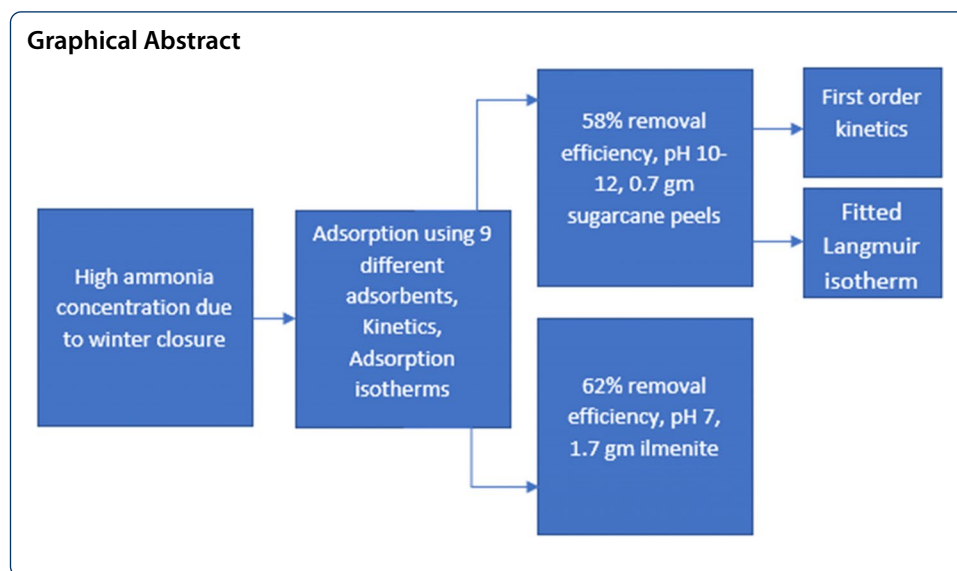
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Abstract

The winter closure is an annual action taken every year by the Egyptian authorities by closing water flows in series of channels for maintenance of water channels where levels in water channels are forced to reduce. However, Kafr El Sheikh and El Behaira, located in North Egypt, were affected by pollutant during winter closure due to the drainage of industrial wastes causing high pollution load of ammonia (mainly) and other pollutants. This paper focuses on testing agricultural wastes and natural materials to decrease ammonia in water at the inlet of water treatment stations that may reach 30 mg/l which happened during the winter closure. Nine adsorbents were investigated for ammonia removal: sugarcane peels, activated diatom, activated carbon, activated zeolite, rice straw, white foam, ilmenite, red brick, and a mixture of ilmenite with sugarcane. The sugarcane peels were the optimum treatment solution with a removal efficiency of 58% at an initial concentration of 38 mg/l, ~ 0.7 g of the adsorbent mass, and pH ranges from 10 to 12 after 1 h of contact time. At the same time, ilmenite reached an efficiency of 62% at an initial concentration of 21 mg/l, ~ 1.7 g of ilmenite, and pH 7 after 1 h of contact time. In addition, the reaction kinetics and adsorption isotherms were investigated for the selected adsorbent sugarcane peels, and the results showed that it matched the first-order kinetics with a regression coefficient (R^2) of 0.99 and Langmuir adsorption isotherm (R^2) of 0.96.

Keywords: Adsorption, Ammonia, Drinking water, Ilmenite, Sugarcane peels



Introduction

The Nile River's longest river globally passes through many countries in Africa and ends in Egypt. It is the primary source of drinking water for more than 100 million Egyptians; however, it could be polluted from the disposal of fertilizers and industrial wastes leading to raising the concentration of harmful contaminants, especially ammonia. Ammonia's high concentration would cause undesired odors, and many diseases such as loss of equilibrium, headache, insomnia, and coma sometimes may lead to death. Ammonia and ammonium ions are the most commonly encountered nitrogenous compounds in wastewater, existing in equilibrium; each species' quantity depends on pH and temperature. This ammonia disposal causes a winter closure in North Egypt, leading to a stoppage of treatment stations.

Since the Aswan high dam in Egypt was built in 1960, the water authorities are shutting water from targeted channels to make the annual maintenance of the water and drainage channels for 15 days in each of the five main provinces. The annual shutting of water is called "the winter closure," during that period, the water levels start to go down and potential pollution from industrial facilities may happen. This is because the water intake from the channels is directed to the drinking water treatment plants; in the case of this closure, the chlorine used in disinfection may react with the ammonia (high concentration due to industrial pollutants), forming chloramine that is toxic and causes many diseases. Therefore, an efficient and economical technique is required to remove ammonia from water intake during the winter closure.

Mohamed et al. [1] indicated that sewage disposal into fresh water and industrial effluents would increase ammonia concentration. Mohamed et al. [2] also cited some examples of disasters in the Nile river in the last few years. They also examined tap water quality [physico-chemical and biological quality] in Tanta city. Ammonia present in water would result in nitrite formation in the distribution systems leading to the failure of removal filters, taste, and odor problems.

Many researchers used different techniques for ammonia removal from drinking water depending on parameters such as initial concentration, pH, temperature, and contact time. For example, Zheng and Wang [3] investigated the ammonium ion removal using hydrogel (i.e., cross-linked polymeric networks) composite chitosan. The results indicate that the adsorption equilibrium can be achieved within 3–5 min using 10% rec-torite, adsorption capacity reached 123.8 mg/g, and the hydrogel composite has a higher adsorption capacity for NH_4^+ in a broad pH level ranging from 4.0 to 9.0. Furthermore, no significant changes in the adsorption capacity were found over the temperature range studied from 23 to 51°C. Additionally, regeneration of the hydrogel adsorbent was by 0.1 mol/l NaOH.

Zhao et al. [4] investigated ammonia removal using zeolite synthesized from red mud. Five models were used in their experiments to fit with the equilibrium isotherm data, and 17.5mg/g of ammonium adsorption capacity was obtained. The studied parameters were 2–11 pH, 5–500 mg/l initial concentration, and 0.5–25 adsorbent dosage.

Soetardji et al. [5] removed ammonia by modified zeolite mordenite using sodium hydroxide. Additionally, the modified adsorbent was used for the first time in reducing ammonia concentration compared to natural zeolite. The study proved that modified zeolite has a higher adsorption capacity than natural zeolites, reaching 53.91 mg/g adsorption capacity.

While Guaya et al. [6] removed ammonium using modified natural zeolite by incorporating hydrated aluminum oxide. Three different experiments were conducted: sorption capacity without pH adjustment, sorption capacity at a concentration of 25 mg/l at equilibrium pH from 2 to 11, and sorption capacity with the existence of other ions competing in the wastewater without pH adjustment. The sorption capacities were simultaneously 33, 30, and 26 mg/g.

Bernardi et al. [7] evaluated ammonia removal efficiency using chitins, three commercial chitosan, and chitosan produced in the laboratory. The commercial chitosan entirely removed the minor initial concentration of 0.09 mg/l.

Cruz et al. [8] investigated polymer hydrogel with sorption capacities of $\text{NH}_4\text{-N}$ 8.8–32.2 mg/g, which corresponds to removal efficiencies ranging from 68 to 80% $\text{NH}_4\text{-N}$, unaffected by pH variations, as the sorption capacity remained constant from pH 5.0–8.0, while regeneration occurred at pH 4.

Huang et al. [9] reviewed the factors determining adsorbents' cost-effectiveness: source abundancy, low cost, handling process, removal capacity and time for removal, and easiness of regeneration or any other application. Many adsorbents were mentioned as natural and synthetic zeolite, polymers, carbon-based adsorbents, hydrogels, industrial and agricultural wastes, and nanoparticles. For example, natural Iranian zeolite reached 90% removal at 40 mg/l ammonia concentration, while natural Chinese clinoptilolite did not remove any ammonia at an ammonia concentration of 15 to 150 mg/l.

Yuan et al. [10] recovered ammonia using a rotating packed bed; the initial ammonia concentration was 1000 mg/l. Experiments were performed at temperatures ranging from 25 to 40°C, 0.05 l/min liquid flow rate, gas flow rate 80 l/min., and pH 11. The efficiency reached 81% within 13.3 s at 40°C. Cheng et al. [10] explained the simultaneous removal of ammonia, iron, and manganese from groundwater. Actual groundwater

samples were treated in a pilot-scale bio-filter with an ammonia removal efficiency of around 91% at 0.4 m.

While Shu et al. [11] studied the oxidation of ammonia by pulse electrolysis, this method removed ammonia and recovered manganese simultaneously. The investigated parameters and technological conditions were temperatures, initial pH, and concentration of added NaCl. Batool et al. [12] published a summary for membrane distillation removal for the dissolved ammonia. The membrane distillation technique was suitable for high-temperature wastewater treatment, with a low concentration of volatile compounds.

Van Nguyen et al. [13] tested cold plasma for treating surface water from microorganisms. After 30 min. of treatment, it was noticed that cold plasma increased nitrate and nitrite concentration and decreased *Escherichia coli* to 1.0×10^5 CFU/ml (colony-forming unit/ ml), ammonia decreased from 1.6 mg/l to 0.4 mg/l, but pretreatment steps before cold plasma were required.

Cold plasma operated at high voltage, weak acidity, and low initial concentration. Cold plasma could be used to remove ammonia from wastewater. Plasma was combined with zeolites to remove ammonia from wastewater. Fan et al. [14] reached around 70% ammonia removal in 30 min with the previous combination, which is better than the 53.7% removal with the zeolite model alone and 8.4% for cold plasma systems alone. Fan et al. [14] enhanced the oxidation of ammonia by the generation of ozone and hydrogen peroxide. The system also compared gases like argon, nitrogen, and air with oxygen as the working gases. Rashmei et al. [15] mentioned that the gliding arc technique generated hydrogen peroxide to enhance oxidation with ozone and decrease pH; this is mainly used to disinfect wastewater from bacteria.

Du et al. [16] stated that gliding arc also included high electric powers compared to corona discharge, and it had been used in a lot of industrial applications. So, the aim of this study is test agricultural wastes and natural materials to decrease ammonia in water at the inlet of water treatment stations a concentration of ammonia equals to 30 mg/l occur, simulating the ammonia concentration during the winter closure.

Nine adsorbents were investigated for ammonia removal: sugarcane peels, activated diatom, activated carbon, activated zeolite, rice straw, white foam, ilmenite, red brick, and a mixture of ilmenite with sugarcane. Reaction kinetics and adsorption isotherms were calculated to the best adsorbent. The novelty of this work is the use of agricultural wastes and natural materials that can be ideal economically and environmentally to developing countries similar to Egypt.

Experimental

The main objective of this work was to investigate agricultural wastes and natural materials as adsorbents for ammonia removal from the inlet of drinking water treatment stations. Safety precautions were taken during the experimental work.

Sample preparation

Standard solutions simulated samples of ammonia-contaminated water were prepared using different salts such as ammonium sulphate, ammonium chloride, and

diluted ammonia solutions from the standard concentration of 25% to the desired concentrations.

All chemicals used to activate some adsorbents or pH adjustment were of standard concentrations; HCL (37%), NaOH (1M) from ADWIC, and PIOCHEM companies for pure chemicals—Egypt. Properties and primary constituents of some adsorbents were performed using XRD (X-ray diffraction), while BET (Brunauer-Emmett-Teller) tests measured surface area and porosity.

Adsorbents Adsorbents used in the experiments were listed as follows:

- Agriculture wastes: activated carbon (produced from bagasse pyrolysis), dates kernel powder, sugarcane peels, pomegranate peels, and rice straw
- Natural materials: diatom earth from mountains in Egypt, zeolite
- Metal: ilmenite from black sand deposits in the Eastern desert of Egypt
- Others: red brick, white foam from electric devices packing boxes

All the above adsorbents (except the activated carbon) were used with simple preparation/activation to avoid the high cost, decrease the stages needed for treatment at the inlet of stations, and avoid the environmental effect. There is not much space available for many steps at existing water treatment stations in Egypt. The activation process of some of the adsorbents is described as follows.

Preparation/activation of some adsorbents Pyrolysis of activated carbon: the bagasse was separated from the peels, and 1.35 ml of sulphuric acid was added, washed, and carbonized at 500°C. Remove the ash with hydrochloric acid (10% concentration) and rewash it with distilled water to remove HCl. Dry at 110°C for 3 h to decrease moisture content.

Dates kernel: dried at 105°C for 2 h, then crushed into powder.

Sugarcane peels: separated from bagasse then dried at 105°C for 2 h, crushed, and sieved to different particle sizes.

Pomegranate peels: wash peels with distilled water, then dry at 130°C for 2 h.

Activation of diatom: grind into fine powders, add HCl (5%) in excess to get rid of CaCO_3 , effervescence for a certain amount of time wait until the reaction stops. Filter the mixture, get rid of filtrate, and wash the precipitate with distilled water until pH is neutralized. Filter the mixture again, get rid of filtrate, and then dry at 105°C until the powder turns white again.

Activated zeolite: wash with tap water and then dry at 105°C overnight.

To measure ammonia concentration before and after the experiments, T80 UV/Vis spectrophotometer with Nessler's reagent method was used at a wavelength of 425

nm. It should be noted that safety precautions were taken at the lab during this method because of the reagent toxicity.

Preparation of Nessler's reagent [17]:

- Add 100 g mercuric iodide (HgI_2), 70 g potassium iodide (KI) in about 250 ml water.
- In another beaker, dissolve 160 g NaOH in 500 ml water.
- Mix the solutions from the two beakers and dilute them to 1000 ml.

Experimental setup The investigation was in a batch technique on a magnetic stirrer. More than 90 experiments were performed to study different parameters such as adsorbents type, pH, initial concentration, and mass of adsorbents. In a 250-ml beaker, add 100 ml of a prepared sample, then a certain amount of adsorbent was added.

Stir fast for a minute (400 rpm), let the sample settle for a specific time (i.e., around 10 min.), and filter the sample. Note that pH and temperature were measured before and after each experiment.

Results and discussion

All the adsorbents mentioned above were investigated under different operating conditions (i.e., initial concentrations, pH, and the mass of adsorbents), first activated carbon from bagasse pyrolysis. The initial concentrations of 28 and 36 mg/l were chosen to represent the problem occurring in the treatment stations. Masses of 0.01, 0.02, and 0.03 g were added to 100 ml of wastewater at a temperature of 23°C, and pH 11 as ammonia favors high pH. Three replicates were used for the analysis, and the results were as follows:

Activated carbon results

Increasing masses of activated carbon increased the removal of ammonia (slightly) as the number of active sites increased.

The maximum removal efficiency reached was 49% of ammonia at an initial concentration of 36 mg/l. The reason for this low efficiency may be due to the result of the BET analysis, which showed a small surface area of 343.7 m²/g. It was a potential candidate to remove ammonia ions. But those results did not comply with Ashour and Tony [18], who studied activated carbon from date pits with a surface area of 873 m²/g and reached a maximum of 37% removal at ambient temperature. Figure 1 shows the effect of activated carbon dosage.

White foam results

There was not much literature on this adsorbent, as the white foam is an undesired waste that originated mainly from packing electrical devices. As indicated in XRD analysis, white foam crystals are typical for polyurethane polymer shown in Fig. 2 with a BET surface area of 157.7 m²/g. The foam was added on 100 ml samples at pH 6, and

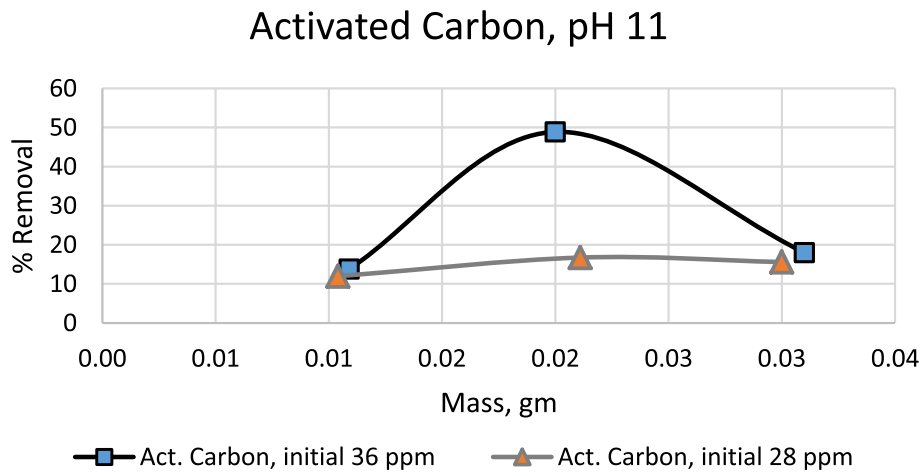


Fig. 1 Effect of activated carbon dosage

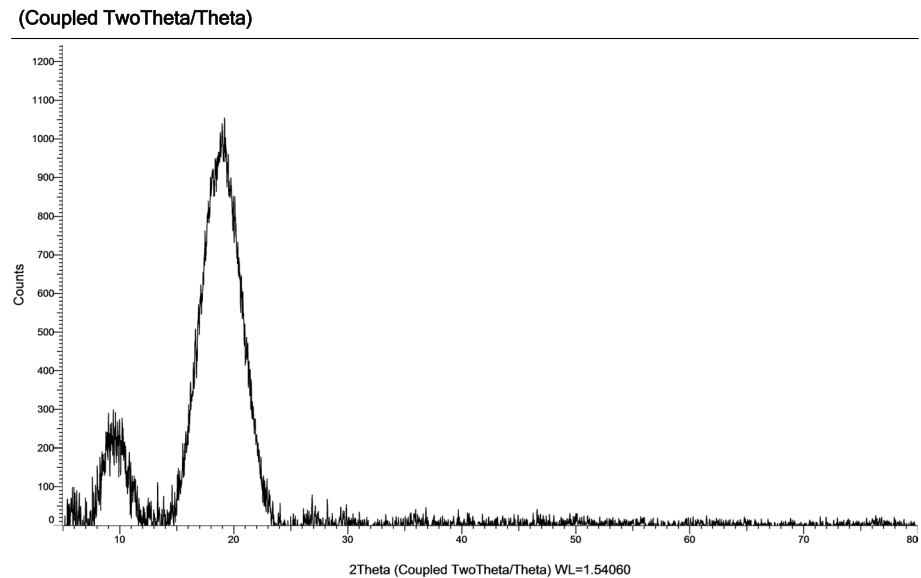


Fig. 2 XRD analysis of white foam

Table 1 Summary of white foam experimental results

Initial conc. (mg/l)	pH) before	pH) after	Mass, g	Final conc. (mg/l)	% Removal	Notes
39	6	6	0.3066	39	0	Stage 1 Foam
39	6	6	0.113	37	5	Stage 2 AC
42	12	12	0.216	40	5	Stage 1 Foam
40	12	12	0.228	37	8	Stage 2 Foam
39	6	6	0.3049	39	0	Stage 1 Foam
39	6	6	0.3184	41	0	Stage 2 Foam
41	6	6	0.295	43	0	Stage 3 Foam
65	6	6	0.3189	67	0	Stage 1 Foam
67	6	6	0.3041	67	0	Stage 2 Foam
67	6	6	0.2296	66	2	Stage 3 Foam

the contact time range from 12 to 30 min with different masses and initial wastewater concentrations.

A mixture of white foam and activated carbon was also investigated; the results were summarized in Table 1.

It was found that white foam did not affect ammonia removal; as indicated in the GCP applied technologies (global construction products) [19], online report that polyurethane foam has excellent chemical resistance to ammonia derivatives and other chemicals. So, another adsorbent was investigated, which was dates kernels powder.

Date kernel powder

Date kernels were separated from dates, dried, and crushed into fine powder. After addition to water, it showed color, and it had no effect on ammonia removal in the range of initial concentrations of 17 up to 25 mg/l, pH 7 and 9. Low porosity might be the reason for this phenomenon.

Sugarcane peel results

Peels were separated from bagasse, dried at 105°C for 2 h, crushed, and sieved to different particle sizes. Chemical treatment was not recommended in this study to reduce cost as this treatment should be added to existing plants. The XRD analysis of sugarcane peels is shown in the following Fig. 3. The crystallographic planes (101) and (002) were typical for cellulose, lignocellulosic material, and BET surface area of 981.2 m²/g. Different masses, initial concentrations, pH, and particle sizes were evaluated, and the results are presented below.

Effect of adsorbent mass

Different weights of sugarcane peels were investigated within a range of 0.5–0.7 g at pH 11 and 710-micron particle size. It was found that increasing mass at higher

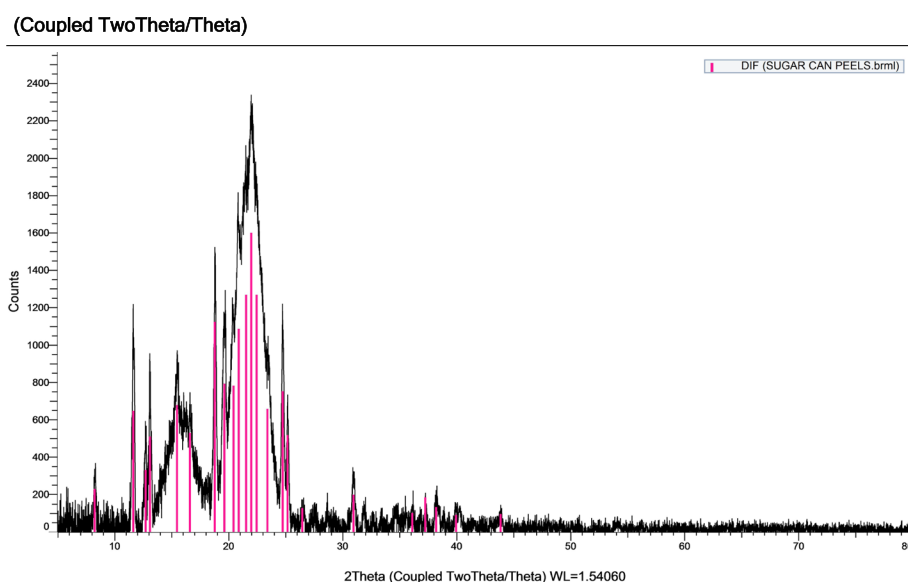


Fig. 3 XRD analysis of sugarcane peels

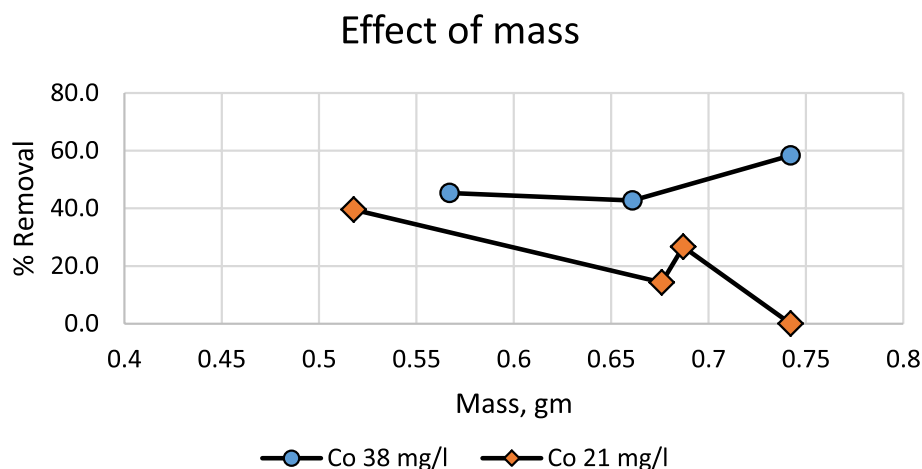


Fig. 4 Effect of adsorbent mass

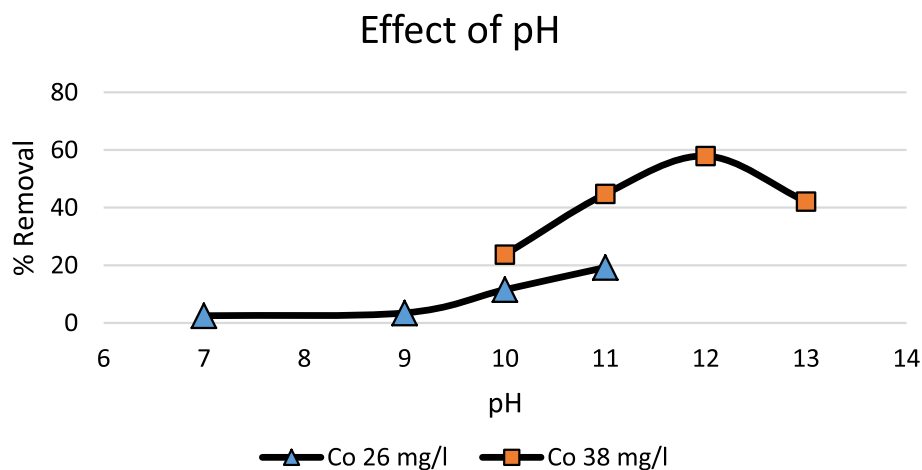


Fig. 5 Effect of pH on ammonia removal

concentration increased the removal efficiency up to 58% and did not increase any more as if it reached saturation, still, the higher the mass, the lower the removal for lower concentrations.

The mass with the most stable result at different concentrations was around 0.5 g. This mass was used in the following experiments to study the rest of the parameters and reach optimum efficiency. Figure 4 shows the effect of adsorbent mass.

Effect of pH

pH is an essential variable for ammonia as ammonium ions turn to ammonia at higher pH, and it becomes easily separated. The results showed that the removal percentage increased until pH 12. Two different initial concentrations were tested (i.e., 26, 38 mg/l). The lower the initial concentration at low pH, the lower the removal as ammonia favors high pH. Figure 5 shows the effect of pH on Ammonia removal.

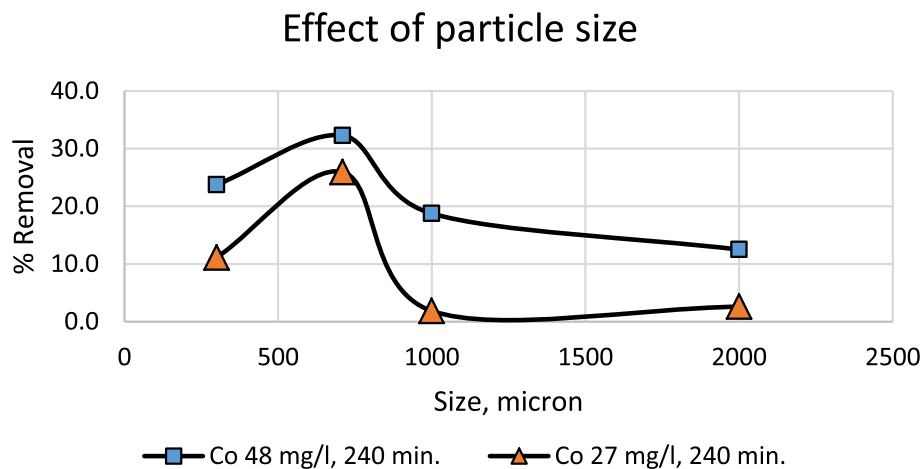


Fig. 6 Effect of particle size on ammonia removal

Effect of particle size

The investigated particle size range was 300, 710, 1000, and 2000 micron for sugarcane peels. The smaller sizes, 300 and 710 microns, had higher removal percentages at higher initial concentrations. It was observed that sugarcane absorbs water during the experiment and became swelled, so this low efficiency may be due to reaching saturation. Figure 6 shows the effect of particle size on ammonia removal.

Effect of initial concentration

Different initial concentrations for wastewater were investigated with a constant mass of 0.5 g of adsorbent at pH 11- and 300-micron particle size. The higher efficiencies were at lower concentrations until 40 mg/l. The low mass could not remove more than 60%. Figure 7 shows the effect of initial concentration on the removal of ammonia.

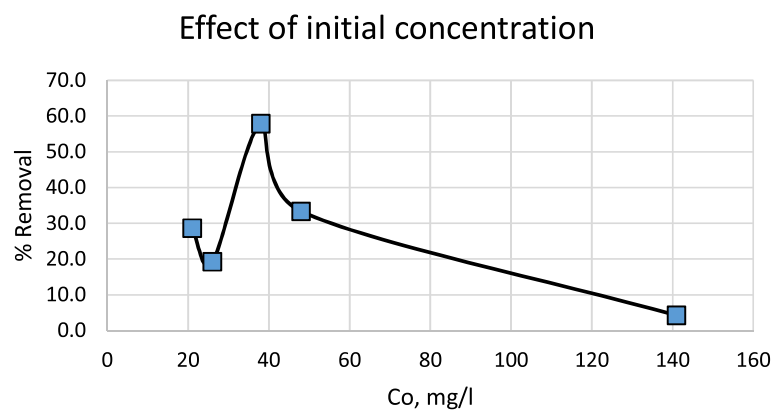


Fig. 7 Effect of initial concentration on ammonia removal

Table 2 Values of kinetic constant parameters

First-order parameters			Second-order parameters		
C_o	K_1	R^2	C_o	K_2	R^2
27	0.0014	0.9932	27	0.00006	0.9983

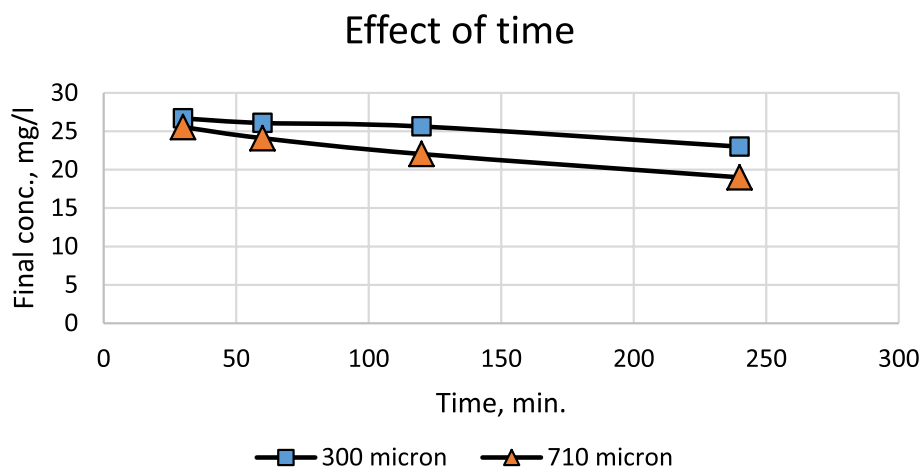


Fig. 8 Effect of contact time

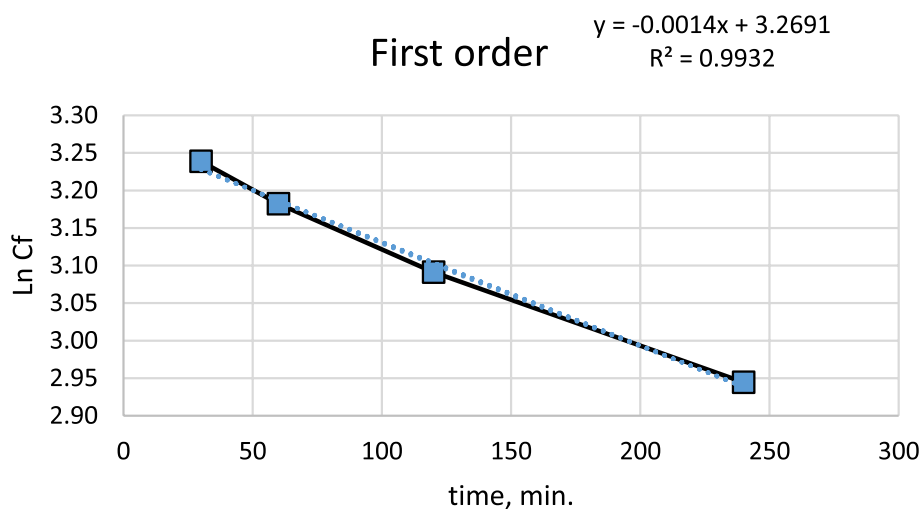


Fig. 9 The first-order kinetics

Kinetics

Experiments were performed to identify the reaction order and mechanism. The results validated the pseudo-first-order, as the pseudo-first-order had a higher determination coefficient of 0.9932. Table 2 shows the values of the constant parameters for this order.

Kinetics model

Kinetics of ammonia adsorption were investigated using the first- and second-order reactions and drawn after linearization to $\ln C_f = \ln C_o - k_1 t$ for 1st order and $\frac{1}{C_f} = \frac{1}{C_o} + k_2 t$ for 2nd order.

where C_o is the initial concentration of reactant C , K_1 is the rate constant of the first order, K_2 is the rate constant of the second order, and t in the time (s).

It can be concluded from the values of R^2 for both orders that the first-order explains the adsorption of ammonia to sugarcane peels pores, as shown in Figs. 8, 9, and 10.

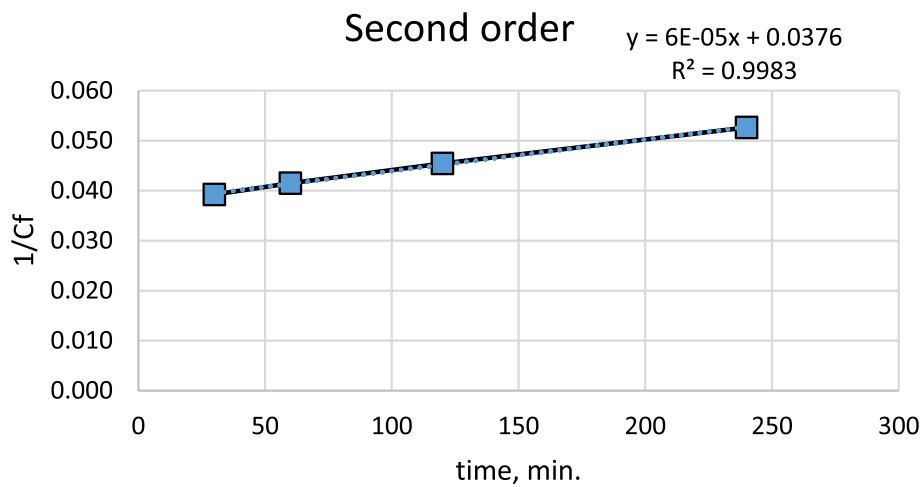


Fig. 10 The second-order kinetics

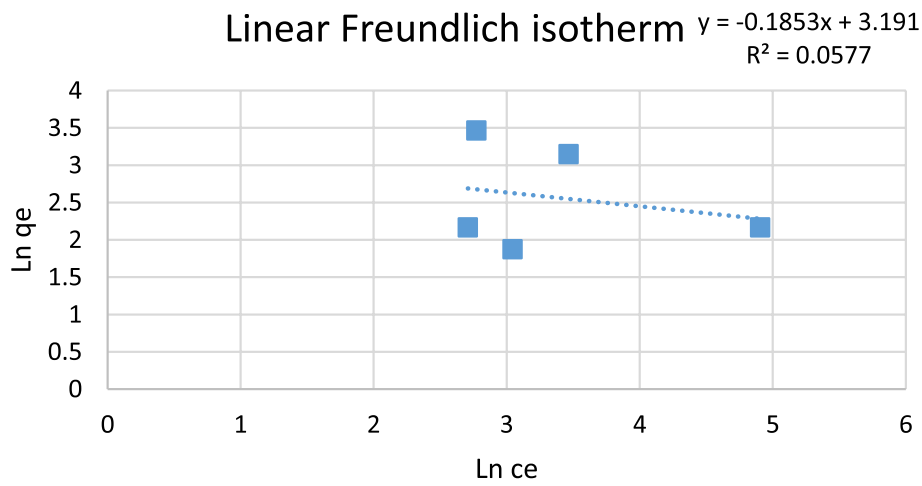


Fig. 11 Linear Freundlich isotherm

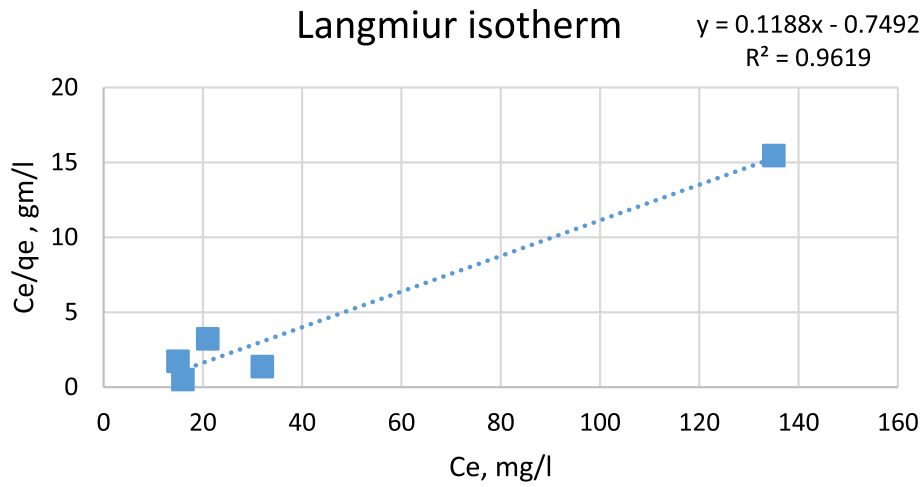


Fig. 12 Linear Langmuir isotherm

Adsorption isotherm models

The results were validated using two different models, Freundlich and Langmuir isotherms. The results were presented in the following Figs. 11 and 12 for 100-ml sample volume and pH 10 at room temperature for five samples.

First: linearized Freundlich isotherm

$$\ln q_e = \ln K + n \ln C_e$$

Second: linearized Langmuir isotherm

Since the regression coefficient of Freundlich did not fit the data, the Langmuir isotherm was investigated

$$C_e/q_e = (1/q_{\max}) * C_e + 1/q_{\max} * K_L$$

where q_e = adsorbent conc. at equilibrium, C_e = concentration of adsorbate at equilibrium, and Q_{\max} and k_L are the maximum adsorption capacities.

The data fitted Langmuir adsorption (monolayer adsorption).

Ilmenite powder results

Ilmenite is an iron titanium mineral; it was suggested as most zeolites' (one of the effective adsorbents) compositions containing titanium dioxide that removed ammonia with high efficiency. Ilmenite is found mainly in sedimentary rocks, and it contains iron that should be removed in another stage. The XRD analysis of ilmenite was shown in Fig. 13, showing iron titanium oxide typical for pure material and with a BET surface area of 65.5909 m²/g.

Formula: FeO₃Ti (pure metal)

Effect of pH

The pH range was tested from 4 to 12; the optimum pH was 7 at an initial concentration of 25 mg/l with 0.6 g of adsorbent after 60 min; it was noted that ilmenite did not change the pH after the test and a good potential for ammonium removal. Figure 14 shows the effect of pH.

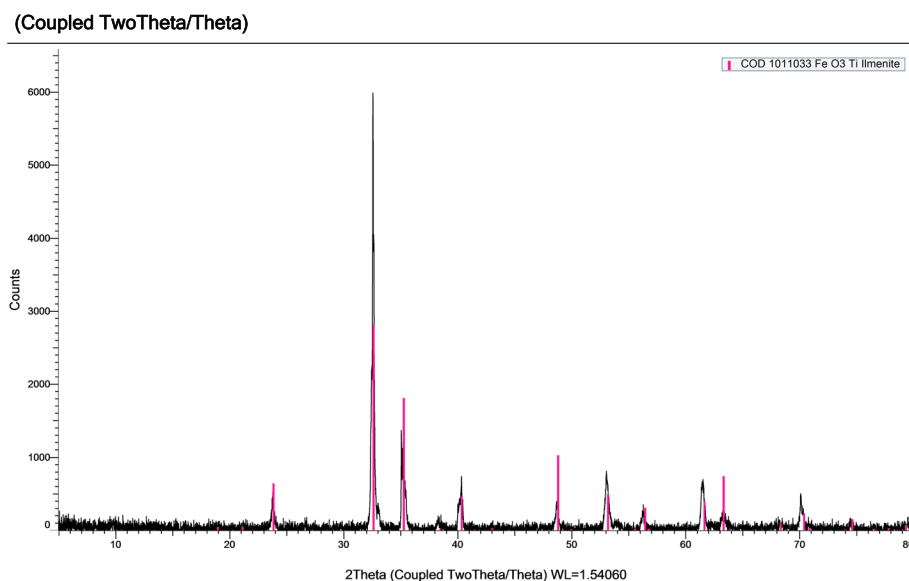


Fig. 13 XRD analysis of ilmenite powder

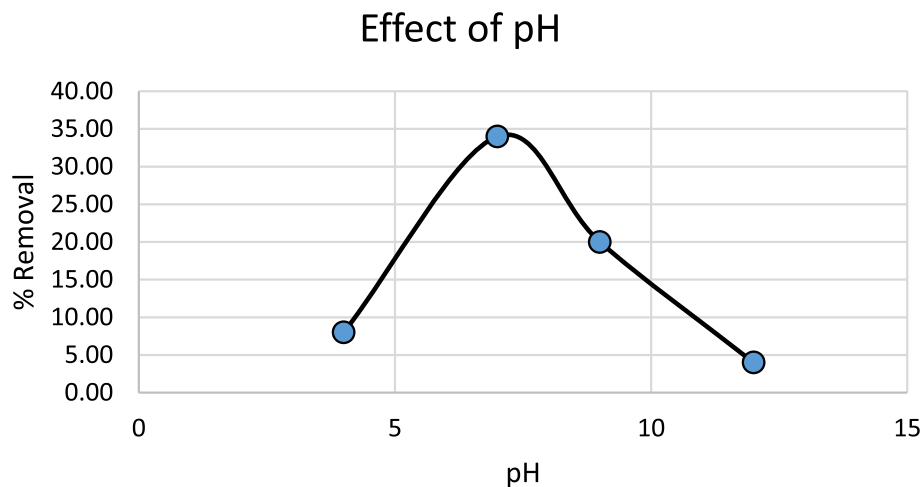


Fig. 14 Effect of pH at an initial concentration of 25 mg/l

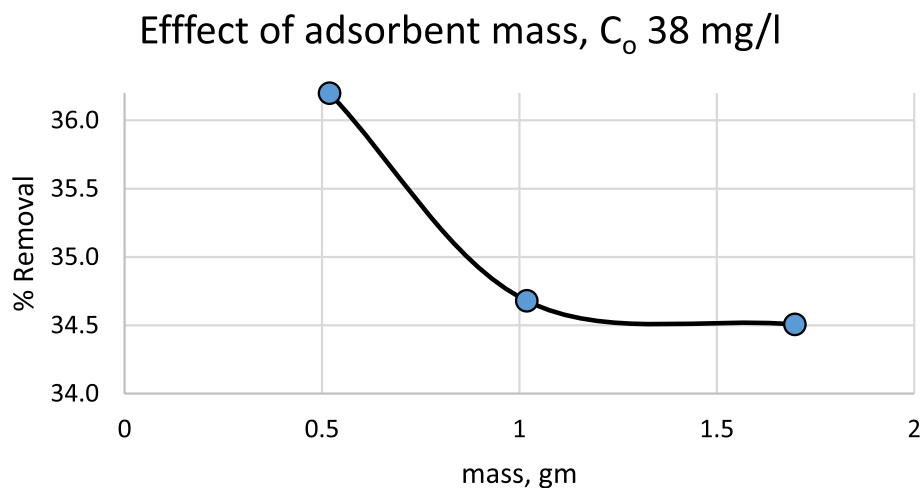


Fig. 15 Effect of adsorbent mass at high concentration

Effect of adsorbent mass

Different weights for ilmenite were investigated with a tolerance ranging from 0.5 to 1.7 g; it was found that the most suitable mass at different concentrations was around 0.6 g at a pH 7. This mass was used to study the rest of the parameters and reach optimum efficiency. Figure 15 shows the effect of adsorbent mass at a high concentration.

The higher mass at low concentration reached the maximum removal efficiency of 60%, then whatever higher mass or time, the removal efficiency did not exceed 60% as the wastewater became saturated. Figure 16 shows the effect of adsorbent mass at a low concentration.

Although it had higher efficiency than other adsorbents, the water was associated with a change of color and traces of ilmenite particles at the outlet. This is because iron ions were dispersed in the solution which is unfavorable and needs further treatment.

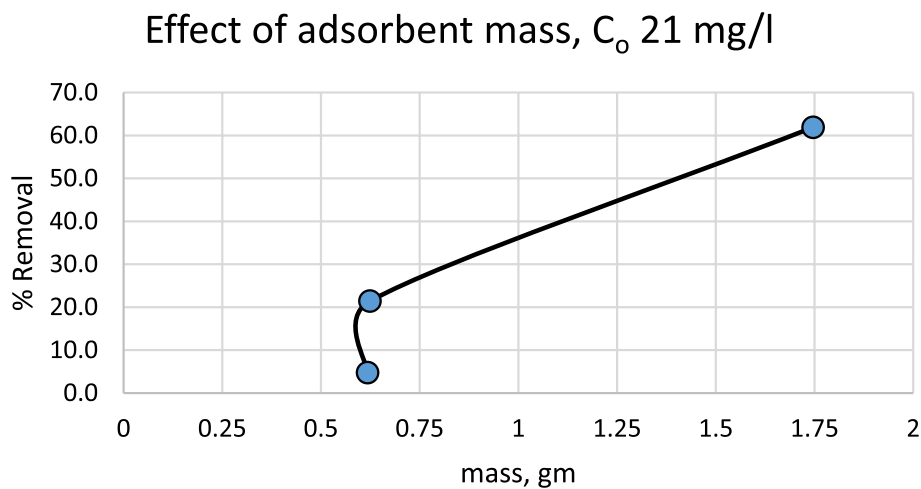


Fig. 16 Effect of adsorbent mass at low concentration

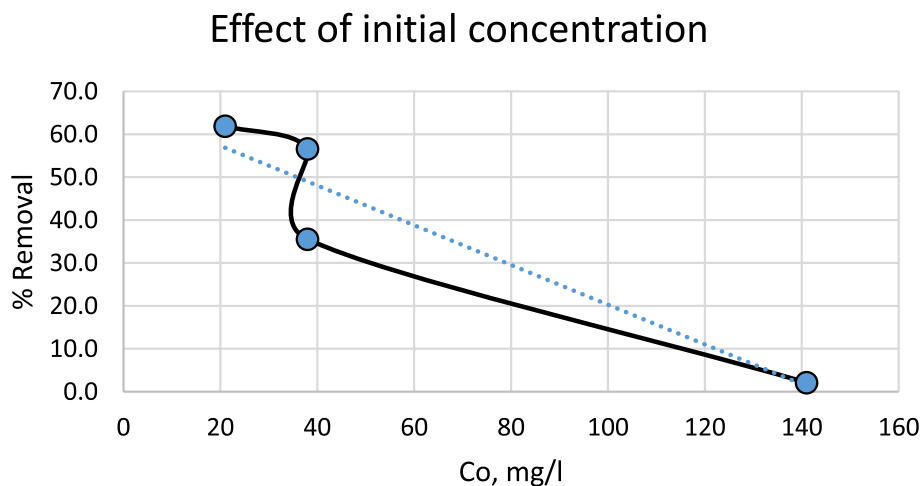


Fig. 17 Effect of initial concentrations

Effect of initial concentration

As the most efficient adsorbents were sugarcane and ilmenite, a mixed ratio of the two adsorbents was tested to see their effect together. As shown in Fig. 17, the result showed that at a low initial concentration of 15 mg/l at pH around 7, the highest removal ratio was 21.6%, using a ratio of 1:1 of both adsorbents after 90 min.

This low removal efficiency was unexpected, and the reason was not found, so it is recommended to study the effect of mixing sugarcane and ilmenite by other researchers and further testing on a molecular level. Figure 18 shows the effect of mixing sugarcane peels and ilmenite.

Activated zeolite

The activated zeolite results did not exceed 15% ammonia removal at an initial concentration of 21 mg/l, pH 7, and masses ranging from 0.2 to 1.5 g in 100 ml samples. Unlike

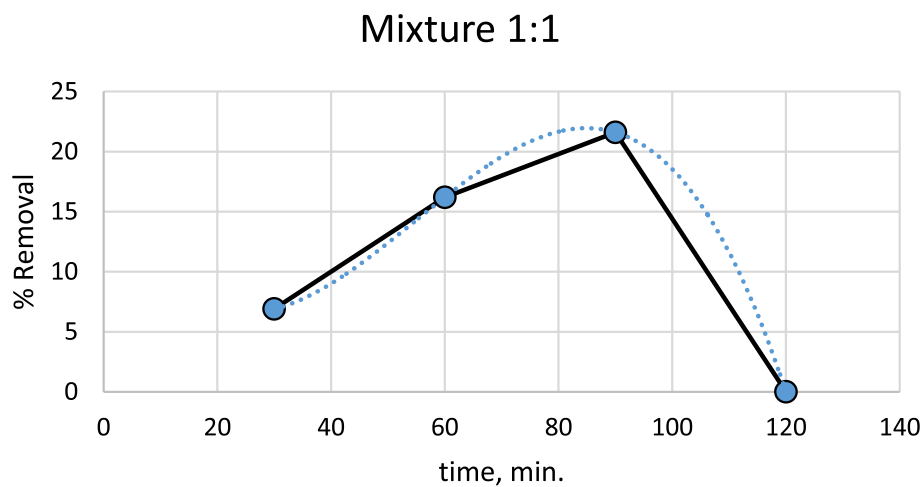


Fig. 18 Effect of mixing sugarcane peels and ilmenite

Table 3 Comparison table shows the difference in BET

No	Adsorbent	S. area, m ² /g	Max. removal %	Pore radius, nm
1	Sugarcane peels	981.213	58	1.3897
2	Activated diatom	826.374	9	1.6501
3	Activated carbon	343.742	49	3.967
4	Activated zeolite	225.478	20	6.0478
5	Rice straw	184.669	17	7.3842
6	White foam	157.669	24	8.6487
7	Ilmenite	65.5909	62	20.79
8	Redbrick	9.38497	0	145.3
9	Ilmenite and sugarcane	-----	22	-----

the results in literature, it was not chemically activated in this study to minimize cost and avoid the environmental effect.

Finally, the BET test determines the surface area of m²/g and the pore radius in the adsorption technique. A comparison of BET analysis for the whole adsorbents was made in Table 3, showing the sugarcane peels had the highest surface area but lowest pore radius. There is a comparison between other adsorbents at the optimum conditions for these experiments: at the initial concentration of 38 mg/l, temperature 25°C, the mass of adsorbent 0.67 g, and 30 min. The maximum removal efficiency of each of them was as shown in Table 3.

Conclusions

- The usage of agricultural wastes without further chemical treatment reduces emissions and makes the environment cleaner and healthy, which is ideal for developing countries where most of the agricultural wastes are disposed of by burning.
- The winter closure requires an action in the developing countries that is economically and environmentally friendly as suggested by using agricultural wastes as adsorbents.

- Sugarcane peels and ilmenite were the best adsorbents available, low cost, and easy to operate, reaching removal efficiencies of around 60%, and after regeneration, sugarcane peel removal was about 20%.
- Sugarcane peels were the optimum adsorbent as they did not leave traces of iron in the waste as the ilmenite did in the experiments.
- Reaction kinetics were investigated, and the first-order kinetics was chosen based on R^2 of value equals to 0.99 which was similar to the second-order kinetics; however, the first-order was chosen for the simplicity.
- Adsorption isotherms were investigated, and the Langmuir isotherm was chosen based on R^2 of value equals to 0.96.
- In future research, a column study using the sugarcane peels should be tested to check if ammonia removal's efficiency will increase in a pilot plant behavior. Additionally, the solid waste adsorbents can be tested in future research for the selectivity with respect to other ions present in the drinking water with similar characteristics such as potassium, sodium, calcium, and magnesium.

Abbreviations

ADWIC	El Nasr Pharmaceutical Co
BET	Brunauer–Emmett–Teller
GCP	Global construction products
PIOCHEM	Pioneer For Chemical Company
R^2	Regression coefficient
XRD	X-ray diffraction

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Authors' contributions

R.S made the experimental work, correlated the results with the authors' discussion, and wrote the manuscript, Dr. I. I proposed the research point and assisted in the experimental work, Dr. N. A aided in the whole research discussion of the experiments and results. At the same time, Dr. A. A assisted in writing and revising the paper and follow-up of the submissions. The authors read and approved the final manuscript.

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Availability of data and materials

The datasets supporting the conclusions of this manuscript are included within the manuscript.

Declarations

Competing interests

There are no conflicts to declare.

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