

Enhancement of electrocoagulation process efficiency using activated carbon

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ABSTRACT

The electrocoagulation (EC) process has been upgraded using powdered activated carbon (PAC). The conventional EC process has been supported by different concentration of PAC for wastewater treatment that has high concentrations of phosphate and ammonia, under different operational conditions. These operational conditions have included applied voltage (5, 10, and 15 V), retention time, initial total dissolved solids (TDS; 1,000, 2,000, 2,500, and 3,000 mg/L), initial pH value (4.5, 7.0, and 9.0), and PAC concentration (2.5, 5, and 10 g/L). The other operational conditions such as temperature, mixing power, and space between electrodes have been kept constant. It was found that EC supported with PAC has high removal efficiency of phosphate and ammonia reaching 75% and 73%, respectively. Further, the system has shown more stability for decreasing the TDS, pH, power consumption, and applied voltage during the experiment. The experimental results were analyzed and confirmed by Freundlich isotherm.

Keywords: Electrocoagulation; Phosphate; Ammonia; Powdered activated carbon; Energy consumption; Electrosorption

1. Introduction

Electrocoagulation (EC) is a wastewater treatment technique, which has many advantages over the conventional technologies such as low sludge production, less environmental pollution, and no difficulties in operation [1]. In additions, EC provides better removal efficiency for the same contaminants that chemical coagulation can remove [2–4]. Moreover, EC has better removal capabilities of some contaminants that are generally more difficult to be removed by other treatment systems [1]. Electricity-based EC has proved to be successful in the removal of oil [2,3], fluoride [4–6], arsenic [7–9], dyes [10–14], suspended solids [15], surfactants [16], chromium ions [17], and phosphate [18–20]. However, this technology increases the hydroxide ions and total dissolved solids (TDS) concentration of the treated effluent [22] due to the formation of some salts such as FeCl_2 and FeCl_3 [21,22], which is the main drawback of this process.

Recently, several hybrid versions of EC are approved such as EC with fluidized metal-impregnated granular activated carbon [23], EC with photocatalytic process [24], EC with air-breathing cathode [25], and EC with perfluorooctanoate on a zinc anode [26]. Most of the previous versions have concentrated on enhancing the removal efficiency of EC rather than reducing the effluent TDS concentration and pH of the treated effluent.

Recently, EC process can be used effectively to remove ammonia nitrogen from wastewater as a result of small bubbles as well [27,28]. In this process, much ammonia is converted to ammonia nitrogen gas, which is stripped with gases formed around the cathode [29]. Further, activated carbon can also enhance ammonia removal with successful results [30,31]. However, up to date, most of the previous researches haven't focused on combining the two techniques as a hybrid system in one reactor.

Newly, EC can be used also to remove phosphate compounds, which are degraded into PO_4 , and reacts with dissolved ions of iron and aluminum [32,33]. The formed iron

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hydroxide has a large surface area on which rapid adsorption of soluble phosphate takes place with a consequent removal of phosphates from wastewater [34]. Precipitation of phosphate involves the dissolved cations Fe when iron is present in the water, $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$, or mixed $\text{Fe}(\text{OH})_3\text{-FePO}_4$ [35]. Therefore, the main aim of this research is to add powdered activated carbon (PAC) dose to the EC process to remove high concentration of phosphate and ammonia from water under different operational conditions, and to investigate the effect of PAC in EC process on TDS concentration and pH value.

2. Materials and method

Fig. 1 shows a schematic diagram of the EC bench scale reactor that was used in this study. Further, Fig. 2 illustrates the real photo of this setup. The batch reactor consists of four iron electrodes with dimensions of 15 cm width, and 20 cm length connected in series to a positive, and negative electrical charged direct current (DC) supply. The distance between electrodes was kept 4 cm, and the electrodes were submerged in a glass basin with dimensions (40 cm long, 20 cm wide,

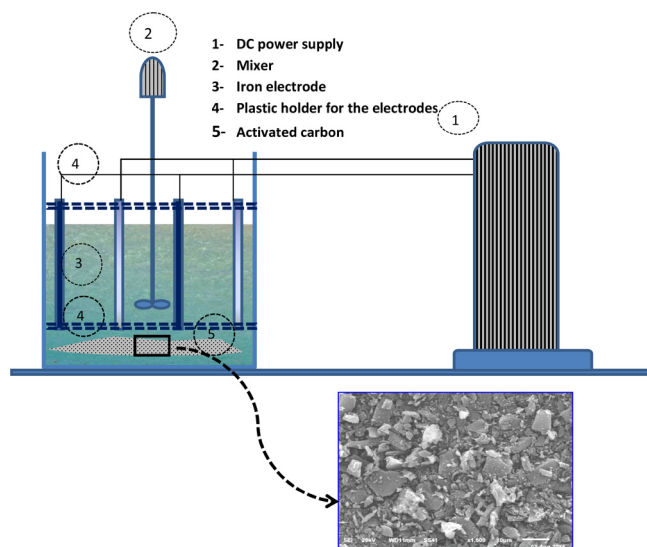


Fig. 1. Flow diagram of setup, the measurements, and SEM image of PAC.



Fig. 2. Experimental setup and the measurements.

and 30 cm height), to handle a volume of 10 L of water. The power supply produced a maximum current of 30 A and a series of volts as 5, 10, 15, 20, and 25 V with an ammeter, and voltmeter was connected to the power supply. A voltmeter was connected externally in parallel to measure the voltage and current induced on the plates, and multimeter (model Crison mm 40+) was used to measure TDS, conductivity, temperature, and pH. Commercial PAC, which passed methylene blue adsorption test, has been used as a sorbent material. A mixer was used for stirring the water contents during the treatment.

A series of laboratory experiments were conducted with and without the additions of PAC to investigate the effect of operational conditions (TDS, pH, carbon dosage, and voltage applied) on the EC effluent TDS, pH, and energy consumption. The experiments were conducted with different initial values of TDS concentration. First, TDS was gradually increased from 1,000 to 3,000 mg/L, at constant pH 7.0, and applied voltage (10 V). Second, various pH of the feed solution was investigated (4.5, 7.0, and 9.0), at initial TDS concentration of 2,000 mg/L, and applied voltage of 10 V. Finally, the applied voltage was gradually increased from 5 to 15 V, at pH of 7.0, and an initial TDS concentration of 2,000 mg/L. All of the above experiments have been conducted using a 10 g/L of PAC, and repeated without the addition of any PAC.

For nitrogen and phosphate removal, high concentration of potassium hydrogen phosphate (K_2HPO_4) and ammonia nitrogen ($\text{NH}_3\text{-N}$) was used as a source for phosphate and ammonia separately. The initial concentration of 20 mg/L of potassium hydrogen phosphate and 50 mg/L of ammonia nitrogen was used as the maximum values expected of these in wastewater [31,32,34]. The concentrations of ammonia nitrogen and phosphate in the effluent samples were estimated using spectrophotometer in accordance with the standard method [36].

The retention time for all runs was extended to 60 min. Samples were taken every 10 min, passed through a filter paper; then, TDS, conductivity, and pH were measured using an electrical multimeter (Crison mm 40 digital meter). In all runs, sodium chloride (NaCl) was used to prepare the feed solution with the required concentrations. The electrical current produced in the EC unit was recorded using an ammeter connected in the DC power supply, and energy consumed was calculated as the product of voltage and current applied. The electrosorption removal efficiency and the sorbent capacity of PAC were estimated as the following equations:

$$\text{Removal efficiency} = (\text{Influent} - \text{Effluent}) / \text{Influent} \times 100 \quad (1)$$

$$\text{Sorbent capacity} = \frac{\text{TDS removed (mg/L)} \times 10 \text{ L of water/}}{\text{Weight of PAC in g}} \quad (2)$$

Fig. 3 shows the peaks of Fourier-transform infrared spectroscopy (FTIR) spectrum of different functional groups according to their corresponding wavenumbers. Fig. 4 shows the isotherm for PAC between the relative pressure and volume. To reduce the ash content, the PAC was dried at 110°C for 12 h and then washed with a solution of $\text{HCl}:\text{H}_2\text{SO}_4$ (1:1)

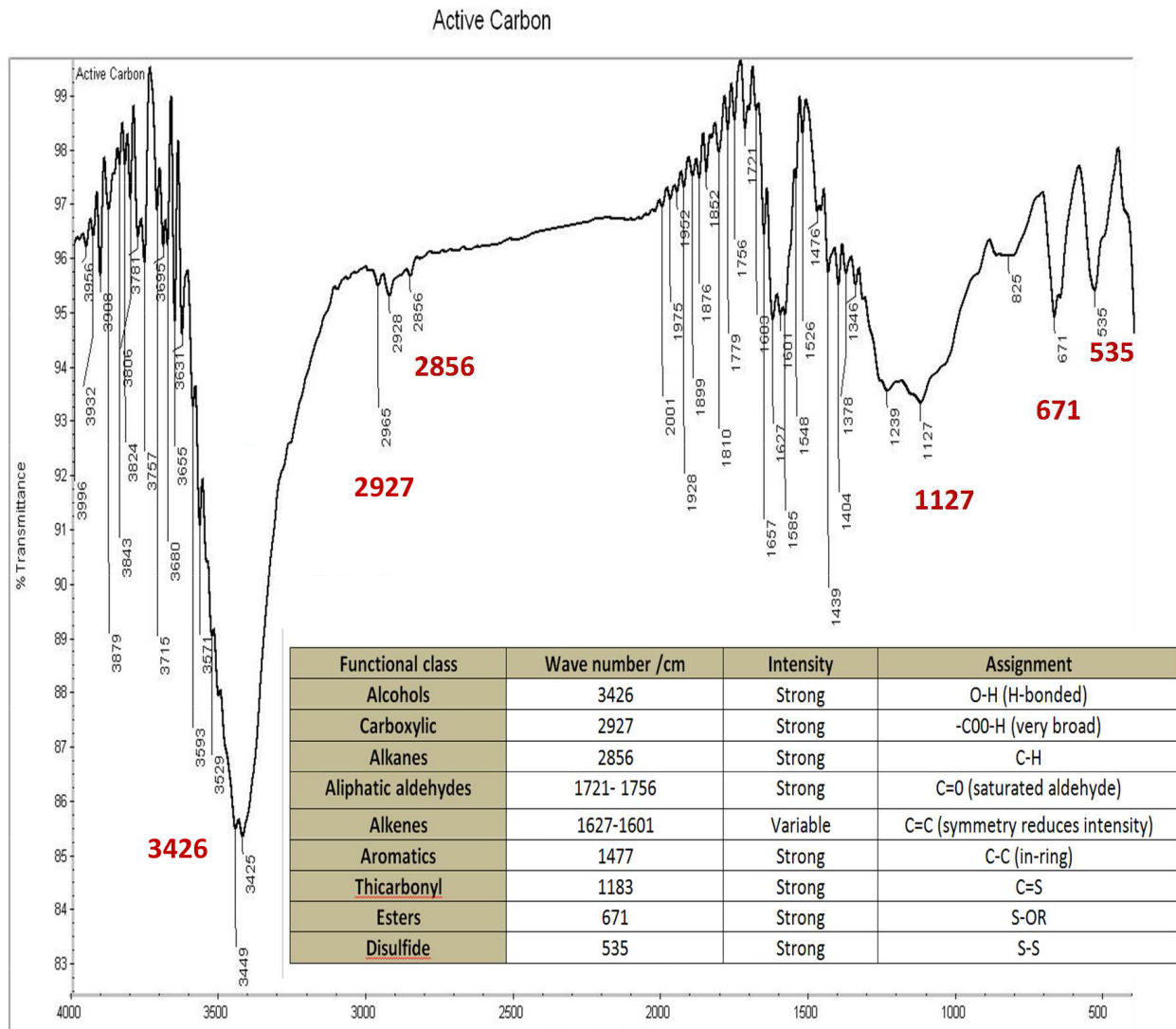


Fig. 3. FTIR spectrum of PAC and possible assignment of PAC spectra.

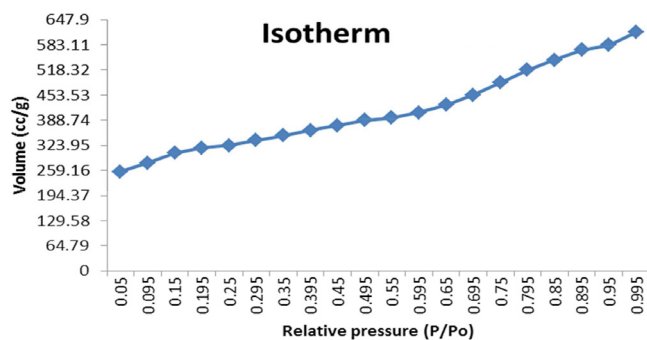


Fig. 4. The relation between relative pressure and volume for the isotherm of activated carbon.

for 24 h. The surface structure of PAC was also examined using scanning electron microscope (SEM) as shown in Fig. 1. FTIR analysis identified the different functional groups present in the PAC. The specific surface area of the PAC was

measured by Quantachrome Nova Automated Gas sorption system using nitrogen as an adsorbate at 77.35 K (the liquid nitrogen temperature). The surface area of the sorbent material was estimated as 1,038.23 m²/g.

3. Results and discussions

3.1. Effect of initial TDS on the process efficiency and energy consumption

Figs. 5 and 6 show the effect of PAC on TDS concentration. It is clear that the presence of PAC keeps the system at constant TDS, pH, and energy consumption irrespective of the retention time and initial concentrations. Conversely, without PAC, TDS concentration, pH value, and energy consumption increase with the retention time. This increase is related to the formation of some salts such as FeCl₂ and FeCl₃ during the system operation. Also, it has been noted that pH value has been raised from 7.0 to 11.0 in all experiments without PAC due to the formation of iron hydroxides. Further,

without PAC, the increase in TDS concentration was directly proportional to the initial TDS concentration.

Fig. 6 shows the effect of PAC on TDS removal efficiency during 60 min. TDS was removed by 38%, 47%, 51%, and 55%, for initial TDS concentration of 1,000, 2,000, 2,500, and 3,000 mg/L, respectively. PAC has achieved adsorption of the produced salts and worked on the stabilization of TDS concentration, prevented the change in pH, and kept pH in the range of 7.0. It is clear that TDS removal efficiency was increased gradually with the increase of the initial feed TDS as shown in Fig. 7. The average TDS removal efficiency was 26%, 27%, 30%, and 37% at initial TDS of 1,000, 2,000, 2,500, and 3,000 mg/L, respectively. It was also noticed that the increase in initial TDS results to a similar increase in sorbent capacity of PAC as shown in Fig. 7. Therefore, PAC sorbent capacity was increased from 25.25 to 148.846 mg TDS removal/g of PAC by increasing the initial TDS concentration from 1,000 to 3,000 mg/L.

The diffuse double-layer capacity was one of the factors having an important role in enhancing and improving the electrosorptive capacity of PAC. The capacity of diffuse layer depends mainly on the electrolyte solution concentration as documented by widely accepted models such as the Gouy–Chapman–Stern theory [37]. The diffuse double-layer

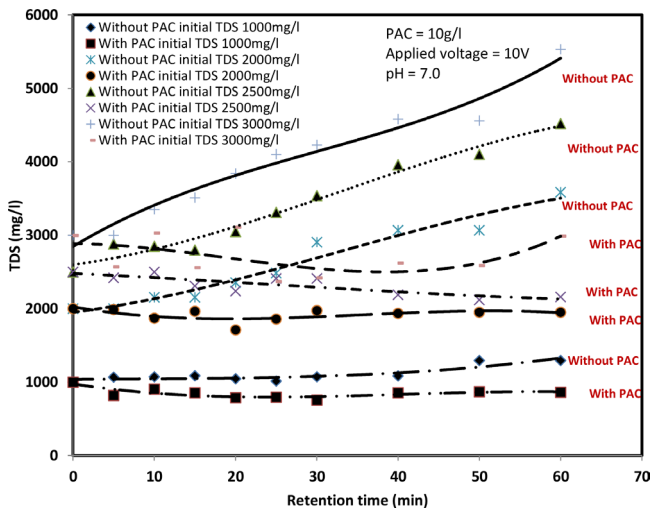


Fig. 5. Effect of PAC on TDS concentration at various initial feed TDS.

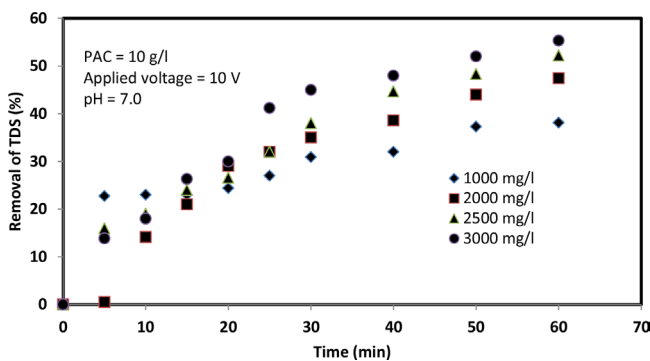


Fig. 6. The effect of PAC on TDS removal efficiency.

capacity increases as the electrolyte solution concentration rises. Consequently, salt storage capacity of PAC was increased directly with the concentration of electrolyte solution [38,39]. The present study confirms that increasing the feed TDS concentration could enhance the electrosorption of PAC through improving the diffuse double-layer capacity.

Fig. 8 shows the effect of initial feed TDS concentration on energy consumption in terms of KWh/m³ and KWh/g of TDS removed. The energy consumption was increased from 2.5 to 10 KWh for each 1.0 m³ water when the initial TDS was increased from 1,000 to 3,000 mg/L. The higher initial TDS concentration has increased the ionic conductivity of the electrolyte and reduced the resistance in electrolyte, so it has consumed more energy.

Conversely, the EC energy consumption in term of KWh/g of TDS removal was decreased gradually from 9.90 × 10⁻³ to 6.71 × 10⁻³, when the influent concentration was increased from 1,000 to 3,000 mg/L. This was related to the higher electrosorptive activity resulting from increasing the feed solution TDS concentration. Therefore, sorption rate rises as the concentration increases.

3.2. Effect of applied voltage on electrosorption efficiency and energy consumption of EC

Fig. 9 shows the effect of PAC on TDS concentration using constant initial TDS concentration of 2,000 mg/L at

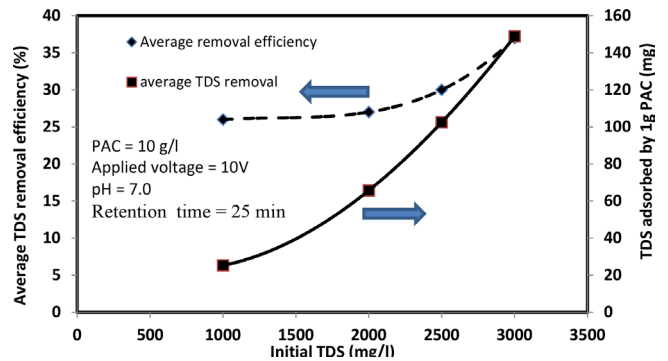


Fig. 7. The effect of PAC on average percentage TDS removal and TDS adsorbed by 1 g PAC (mg).

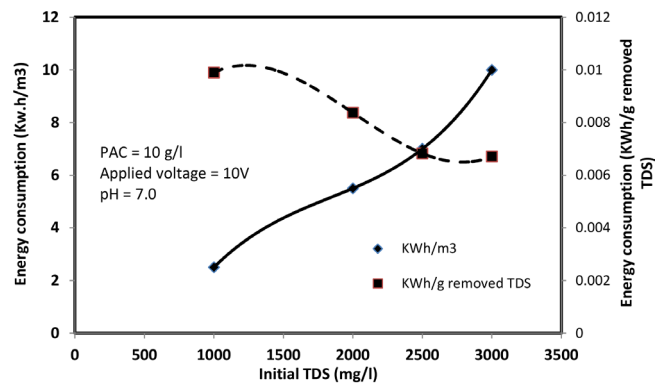


Fig. 8. The effect of initial feed TDS concentration on energy consumed and specific energy consumed per removal TDS concentration.

various applied voltage of 5, 10, and 15 V in two cases. First, operating the EC system without the addition of PAC sorbent media and, second, operating with the addition of 10 g/L of PAC.

In the first run without PAC, it was found that TDS concentration has increased with time depending on the applied voltage. This increase was due to the amount of iron ions that released from the respective electrodes, which has led to the formation of more salts. It was found that 5 V corresponds to the lowest rise in TDS concentration, while 15 V achieved the highest increase. This result indicates that the increase in TDS concentration is directly proportional to the applied voltage. PAC had the same effect on the solution as described before. It was clear that the TDS removal efficiency increased gradually with the increase in applied voltage as shown in Fig. 10. The average TDS removal efficiency was 7%, 26%, and 27%, when using the applied voltage of 5, 10, and 15 V, respectively. It was noticed that the increase in applied voltage corresponds to a linear increase in sorbent capacity of PAC as shown in Fig. 10. The PAC sorbent capacity was increased from 15.1 to 87.22 mg TDS removal/g of PAC by increasing applied voltage from 5 to 15 V.

Fig. 11 shows the effect of voltage applied on the EC unit energy consumption in terms of KWh/m³ and KWh/g of TDS removed. The EC energy consumption was increased

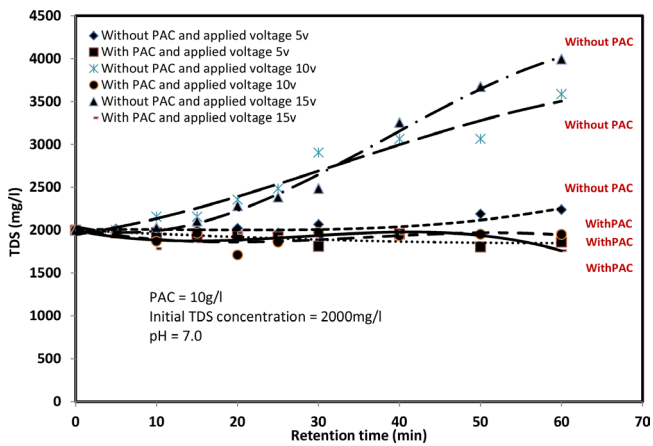


Fig. 9. The effect of PAC on TDS concentration at various applied voltage.

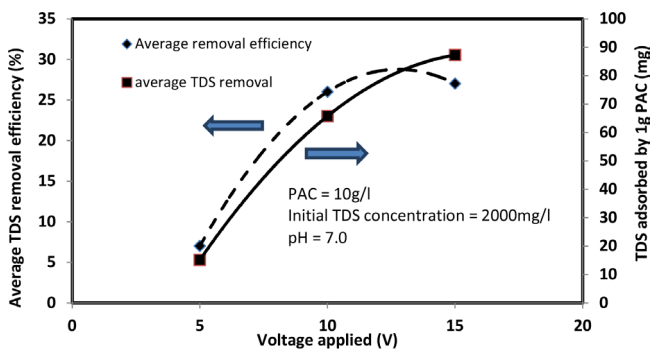


Fig. 10. The effect of PAC on average percentage removal TDS and TDS adsorbed by 1 g of PAC (mg).

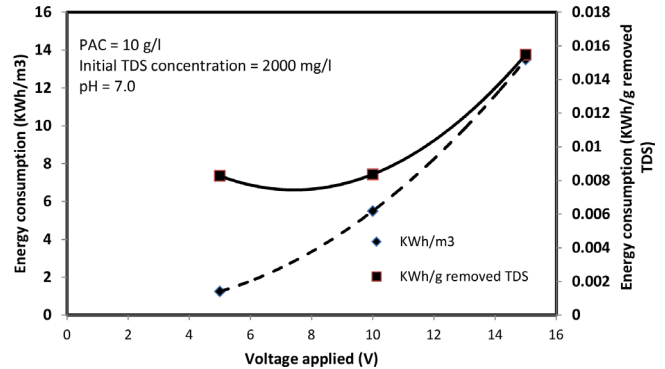


Fig. 11. The relation between energy consumed/removal TDS concentration at different applied voltage.

gradually from 1.25 to 13.5 KWh/m³ of purified water when the applied voltage increased from 5 to 15 V. This result occurred due to the increase in voltage causes more current to be produced, which means power is consumed.

Conversely, the EC energy consumption in term of KWh/g of TDS removed increased from 8.27×10^{-3} to 15.5×10^{-3} when the voltage was increased from 5 to 15 V be cost-effective when the voltage applied, and energy consumption are within the optimal range.

The electrical current density was also monitored and was increased gradually from 3.5 to 10 A after increasing the TDS concentration from 1,000 to 3,000 mg/L. Further, the current density was increased gradually from 2.5 to 9.0 A after increasing the applied voltage from 5 to 15 V. PAC has achieved the same effect on the solution as described before in terms of TDS removal efficiency, which kept the current density constant during the operational time of EC process. The current density was related to the applied voltage value and the concentration of TDS as follows:

$$\text{Current density (A)} \approx 0.0003 \times \text{Applied voltage (V)} \times \text{TDS (mg/L)} \pm 0.5 \quad (3)$$

3.3. Effect of feed solution pH on electrosorption efficiency and energy consumption

Generally, pH of the effluent is changed during the EC process. The variations of final pH depend upon the electrode material and the initial pH of the effluent.

Fig. 12 shows the effect of PAC on TDS concentration at different pH values using initial TDS concentration of 2,000 mg/L and applied voltage of 10 V with and without PAC. It is clear that addition of 10 g/L of PAC has reduced the increase of TDS. Conversely, without PAC, TDS concentration was increased with time. This increase was related to the initial pH of the solution as the highest increase in TDS occurs at alkaline pH and the lowest at acidic pH during the system operation. This result also indicates that the increase in TDS was directly proportional to the initial pH, as it was related to the EC cell reactions at different pH values.

Fig. 13 shows the effect of pH values on the removal of TDS and TDS adsorbed by each gram of PAC. It is clear that TDS removal efficiency increased gradually with the increase in initial pH value. The average TDS removal efficiency was 17%, 30%, and 34% when using the initial feed pH values of 4.5, 7.0, and 9.0, respectively. It was noticed that the increase in pH corresponds to an increase in sorbent capacity of PAC as shown in Fig. 13. The PAC sorbent capacity was increased from 43.16 to 155.03 mg TDS removal/g of PAC by increasing pH from 4.5 to 9.0.

3.4. Effect of applied voltage and EC retention time on phosphate removal

Fig. 14 shows the removal efficiency of phosphate with the initial concentration of potassium hydrogen phosphate 20 mg/L, initial TDS concentration of 2,000 mg/L, pH 7.0 at various applied voltage of 10 and 15 V, with the addition of 10 g/L of PAC. Fig. 14 shows the removal efficiency of ammonia nitrogen with the initial concentration of NH₃-N 50 mg/L, constant initial TDS concentration of 2,000 mg/L, with pH 7.0, and applied voltage of 10 V, at various concentrations of PAC of 2.5, 5, and 10 g/L.

It was found that phosphate concentration decreases with time depending on the applied voltage. The decrease is due to the amount of iron ions, which release from the

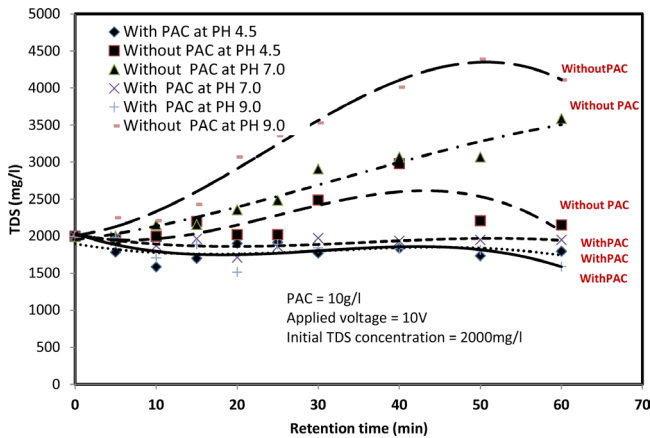


Fig. 12. Effect of PAC on TDS concentration at different pH acidic, neutral, and alkaline.

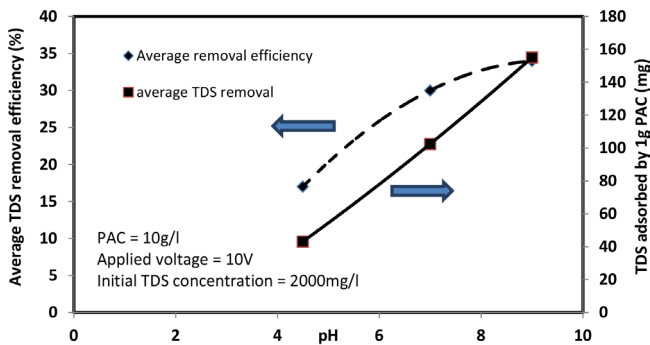


Fig. 13. The effect of pH values on TDS removal and TDS adsorbed by each gram of PAC.

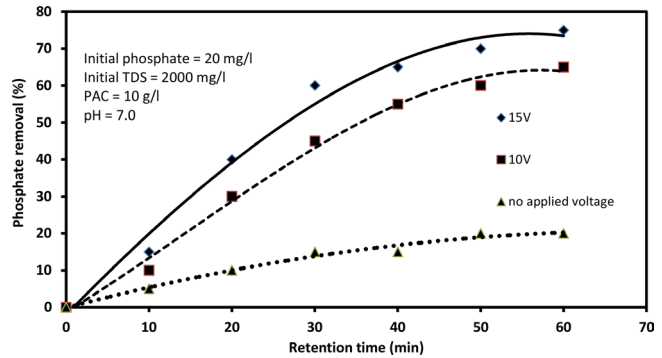


Fig. 14. The relation between removal efficiency of phosphate with retention time and applied voltage.

respective electrodes that leads to the formation of iron hydroxide, which adsorbs a large amount of phosphate on its surface. It was clear that the removal efficiency of phosphate increased gradually with the increase in applied voltage. The removal efficiency of phosphate after 60 min was 65% and 75% at volt values of 10 and 15 V, respectively. Also, longer duration time increases the generation of iron ions resulting in an increase in the formation of iron hydroxide as shown in Fig. 14. Most of the phosphate concentration is removed under optimal values of EC duration time of 50 min, while any further increase of the retention time has a small influence on the phosphate removal.

This result indicates that the increase in percentage removal efficiency of phosphate is directly proportional to the applied voltage efficiency in accordance with earlier studies [40,41]. The adsorption of PAC for phosphate was a physical process rather than an electrostatic process [42], with a removal efficiency of 20%. It was noticed that the PAC had the same effect on the solution in terms of TDS concentration and pH as described before.

It was found that the removal efficiency of NH₃-N increases with time depending on the concentration of PAC. This increase is due to the increase in total carbon surface area and availability of more adsorption sites with increase in PAC dosage, which adsorbs a large amount of NH₃-N on its surface. It was found that 2.5 g/L of sorbent material corresponds to the lowest percentage removal efficiency of NH₃-N, while 10 g/L of sorbent material achieved the highest percentage removal efficiency. Also, longer duration time increases generation of iron ions resulting in an increase in the formation of iron hydroxide as shown in Fig. 15 most of the NH₃-N concentration is removed under optimal values of retention time of 40 min, while any further increase in the retention time has a small influence on the degree of NH₃-N removal.

It is clear that removal efficiency of NH₃-N is directly proportional to the concentration of PAC. The removal efficiency of NH₃-N without the addition of PAC was 36%. It was clear that the removal efficiency of NH₃-N increased gradually with the increase in the amount of sorbent material. The removal efficiency of NH₃-N after 60 min was 58%, 64%, and 73%, when using concentrations of PAC 2.5, 5, and 10 g/L, respectively.

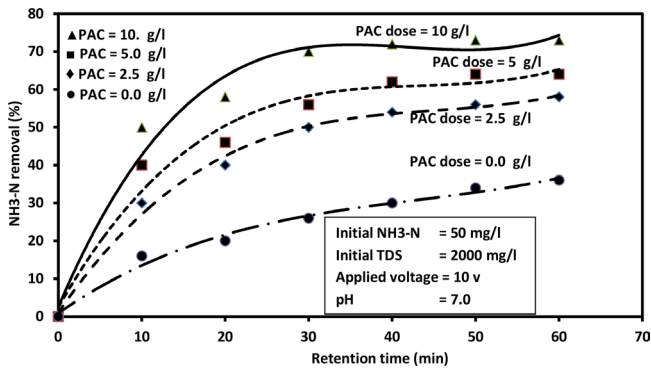


Fig. 15. The relation between the removal efficiency of NH₃-N with retention time under various concentrations of PAC.

3.5. Effect of PAC dosage on TDS concentration and electro-sorption isotherm

Fig. 16 shows the change of TDS concentration and electro-sorption efficiency with the retention time using PAC dosage (2.5, 5, and 10 g/L), initial TDS of 2,000 mg/L, at initial pH 7.0, and applied voltage 10 V.

It is clear that the TDS removal efficiency decreased gradually with the decrease in PAC dosage. The average TDS removal efficiency was 24.5%, 25.9%, and 30% when using the initial feed TDS value of 2,000 mg/L. This implies that when choosing the PAC dosage of an EC unit, it is important to take both the removal efficiency and PAC dosage into consideration to minimize the cost.

It was noticed that the increase in removal efficiency of feed TDS corresponds to a linear increase in PAC dosage as shown in Fig. 16. Although, the PAC sorbent capacity decreased from 400 to 102.5 mg TDS removal/g of PAC by increasing PAC dosage from 2.5 to 10 g/L.

To obtain the electro-sorption isotherm, several experiments were carried out using NaCl solutions with different initial concentrations at constant pH and temperature. The initial concentrations of NaCl feed solutions were 1,000, 2,000, 2,500, and 3,000 mg/L as shown in Fig. 17. The Langmuir isotherm Eq. (4) and Freundlich isotherm Eq. (5) were used to fit the experimental data for electro-sorption.

$$q = \frac{q_m K_L C}{1 + K_L C} \tag{4}$$

$$q = K_F C^{1/n} \tag{5}$$

where C is the equilibrium concentration (m mol/L); q is the amount of adsorbed NaCl (in mg/g of activated carbon); q_m is the maximum adsorption capacity corresponding to complete monolayer coverage with recommended value for Langmuir model is 1,664.03, K_L is a constant related to the heat of adsorption with recommended value for Langmuir model is 0.0000271, K_F and n are constants related to the system characteristics with recommended value for Freundlich model are 0.00123 and 0.68976, respectively.

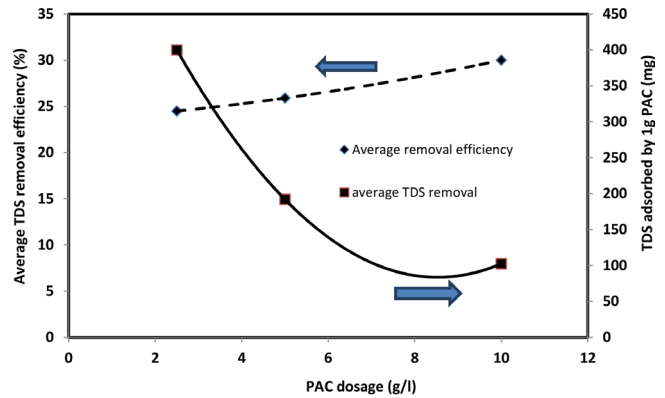


Fig. 16. The relation between different dosage of PAC and average percentage removals of TDS concentration and adsorbed TDS by 1 g of PAC.

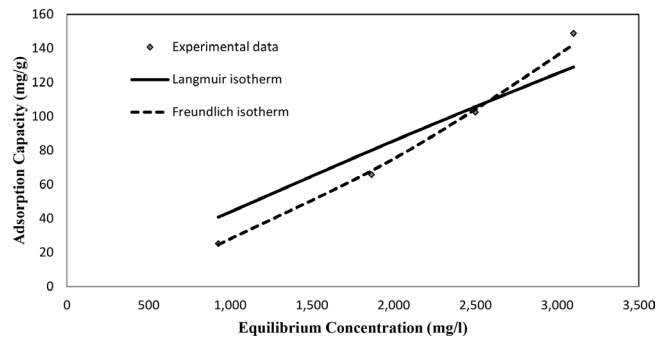


Fig. 17. Electro-sorption isotherm.

It was found that the electro-sorption isotherm follows both Langmuir and Freundlich models. However, the Freundlich isotherm correlates better with the experimental data according to the R² values with recommended value for Langmuir and Freundlich models are 0.84697 and 0.9966, respectively. This suggests that the monolayer adsorption is predominant during the adsorption process. The obtained electro-sorptive results were seen to follow the Freundlich isotherm well, that is, the electro-sorption can be described by this adsorption isotherm.

4. Conclusion

Combination of EC and PAC (EC-PAC) in one reactor was proved to be an effective method to remove phosphate and ammonia with removal efficiency of 75% and 73%, respectively, without increase in TDS concentration and pH values of the treated water. The removal efficiency, sorbent capacity, and energy consumption (in kWh/m³) of EC-PAC are directly related to the initial TDS concentration and applied voltage. PAC concentration needed for EC-PAC is directly related to the initial TDS and inversely related to PAC sorbent capacity. Combination of EC and PAC achieves higher TDS removal efficiency and sorbent capacity at alkaline pH. Combination of EC and PAC has isotherms that follows Freundlich for NaCl removal.

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