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Simultaneous removal of nitrate and phosphate using aluminum electrode coated by ZnO nanoparticles in electrocoagulation process

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ABSTRACT: The excessive usage of nitrogen fertilizers leads to phosphate and nitrate become as a common inorganic pollutant in groundwater resources. Therefore, an alternative process performance seems to be examined to remove these anions from water resources. This study aims to investigate the effect of two variables, water hardness and electrode distance, on the electrocoagulation process for simultaneous phosphate and nitrate removal. For this purpose, initial concentration=100 mg/l, current density=22.5 A/m2, pH=6 and retention time=40 min were considered. Under these conditions, the optimum removal efficiency of phosphate and nitrate were achieved at 2 cm electrode distance with 93.8% and 78%, respectively. While, the water hardness of 65 mg/l CaCO3 was shown 99.3% and 86% removal efficiency, respectively. Then, in order to improve the electrocoagulation process and reduce the passivation rate of the aluminum anode, the electrodes were modified with ZnO nanoparticles by the sol-gel method. The accuracy of electrode coating with zinc oxide nanoparticles was determined by SEM, EDX and XRD tests. The effect of coating was investigated by cyclic voltammetry, showing that current intensity at the modified electrode has improved 202.44%.

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1- Introduction

One of the most valuable water resources for urban, industrial and agricultural uses is groundwater. The intensive use of nitrogen fertilizers in agriculture activities may cause increasing nitrate and phosphate concentration. Excessive anions concentration in water causes overgrowth of plants and the algal bloom phenomenon [1].

Electrocoagulation (EC) process is found an effective water and wastewater treatment method, as it can able to remove a variety of pollutants [2]. In an electrocoagulation system, several electrochemical reactions occur simultaneously at the cathode and anode. Coagulants are produced by dissolving the anode metal. Cation species released by anode cause the hydrolysis and neutralization of the negative charge of contaminants and destabilize suspended particles, led to attracting particles of opposite charge and form flocculants. The formation of a layer on the electrode surface over time is one of the problems of the electrocoagulation process. The combination of electrocoagulation with other technologies has improved efficiency. Several parameters such as type of electrodes, electrodes distance, pH, current density and electrolyte affect the electrocoagulation. The effect of these factors varies depending on the type of pollutant wastewater. Finding the optimal range of effective parameters to achieve proper efficiency is important [3-5].

In 2020, Hashemzadeh et al. Studied the electrocoagulation process in the simultaneous removal of nitrate and phosphate. In this study, the removal efficiency of nitrate and phosphate under conditions of initial concentration 10 mg/l, 60 min, pH= 7 and initial voltage 30 v, was reported 95 and 45%, respectively [6]. In 2020, Ghazouani et al. investigated the combination of electrocoagulation and electrochemical treatment for simultaneous removal of nitrates and phosphates from synthetic wastewater. Electrochemical treatment process using boron-doped diamond (BDD), which was evaluated as one of the most promising methods for the removal of nitrogenous compounds. The results showed at high current density, the removal efficiency of contaminants is more than 70% [7].

For this purpose, the factors such as hardness, electrode distance, type of electrode coating with nanoparticles and the effect of cyclic voltammetry on the anions removal simultaneously were investigated, as well as the characteristics of the sludge produced during this process. One of the novelties in the electrocoagulation process is coating the electrodes with nanoparticles to improve the process efficiency. There are different methods for coating, but the sol-gel process is very suitable because of its low reaction temperature, low cost, and the production of high-quality and pure nanoparticles (producing particles at the same size)

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[8]. To prevent the passivation rate of the aluminum anode, the electrodes were modified with ZnO nanoparticles by the sol-gel method. The effect of coating was investigated by cyclic voltammetry; accordingly, the coating improves the performance and provides more current to the system.

2- Methodology

A batch reactor with an effective volume of 1.5 L was made from Plexiglas. Sodium nitrate and potassium dihydrogen phosphate (Merck) were used to prepare the samples. The effective area of each applied aluminum electrode was 80 cm^2. The electric current used was imposed by means of a DC power supply (ps-605D). At the end of the experiment time, samples were taken from the cell and were analyzed the effect of hardness on removal efficiency. Nitrate and phosphate were measured by spectrophotometer (Hach). The cadmium reduction method was applied for nitrate concentration and the molybdovanadate rapid liquid method was used for phosphate concentration.

To coat the electrodes with Zno nanoparticles by sol-gel method, 0.87 g of zinc acetate (Merck), 50 ml of methanol and 4 drops (16 μl) of ethanolamine were applied to prepare a solution of 0.08 M zinc acetate. Ethanolamine was added to increase the solubility of zinc acetate in methanol. 50 ml of methanol was poured into a beaker and then zinc acetate was added and stirred for 20 minutes with a stirrer at 60 °C. At the time of mixing, the beaker's door was closed and the liquid-vapor was returned into it by placing ice. 4 drops of ethanolamine were added to the solution and finally, the sample was aged for 24 hours.

Then, samples were prepared at 4000 rpm for 20 seconds by spin-coating. The spin coating process was repeated 4 times and after each step, the substrates were dried by the heater at 110 °C. Heat treatment was performed after coating at 450 °C in the oven at 5 degrees per minute thermal profile until the desired temperature and staying at this temperature for 1 hour.

3- Results and Discussion

Under optimal conditions (initial concentration=100 mg/l, current density=22.5 A/m^2, pH=6 and retention time=40 min), the effect of hardness and electrode distance on the simultaneous removal efficiency of phosphate and nitrate were investigated. By changing the distance from 3 to 1 cm, the phosphate removal efficiency is reduced by about 20%. The optimal distance between the electrodes was 2 cm and the removal efficiency of phosphate and nitrate was 93.8% and 78%, respectively.

To evaluate the effect of hardness, well water with 260 mg / l $CaCO_3$ hardness was diluted with distilled water in certain proportions. Experiments showed that by reducing the hardness, the removal efficiency of nitrate and phosphate increased by about 10%. Phosphate and nitrate removal efficiencies at 65 mg / l $CaCO_3$ were 99.3% and 86%, respectively. Therefore, at lower hardness, the removal efficiency of contaminants was improved.

The produced sludge during the electrocoagulation was evaluated under the initial concentration of 100 mg / l of nitrate and phosphate, the current density of 22.5 A/m^2, pH = 6 and time = 40 minutes. The produced sludge was formed floating on the surface during the process. The gray color of the sludge was observed due to the dissolution of the aluminum metal. To determine the Sludge ingredients, EDX analysis was applied.

The presence of ZnO nanoparticles on the electrode, the morphology of the electrodes and the accuracy of coating with the sol-gel method were investigated by SEM. XRD analysis determines the position of aluminum and ZnO peaks and proves their existence.

The effect of coating was considered by cyclic voltammetry. Accordingly, the starting arc of the descending arc in the voltammeter output diagram is related to hydrogen reduction. Aluminum at -1.14 v and modified aluminum with the sol-gel method at -1.1 v have begun to regenerate the hydrogen peak. The lower the absolute value of these numbers, the sooner reduction occurs.

Sol-gel coating was used to improve the performance of the electrodes and also to prevent their passivation. For this purpose, ZnO nanoparticles were used to coat the electrode. Voltammetry was performed to evaluate the effect of nanoparticle coating. The current intensity at an equal voltage for a coated electrode is about twice more than a simple electrode, which means that a higher current intensity can be obtained at an equal voltage. This shows the importance of nanoparticle coating.

4- Conclusion

In this study, the effect of electrode distance and hardness on the simultaneous removal of anions and also the effect of electrode coating on the improvement of the electrocoagulation process were considered. Energy consumption increased as the space between the electrodes increased. On the other hand, reducing the distance increased the electrical resistance, reduced the efficiency of the electrode and prevented coagulation. The best removal efficiency was obtained at a distance of 2 cm.

The increased hardness had a negative effect on the process, one of the reasons being the formation of by-products and stable compounds in the presence of calcium and magnesium cations. In general, the application of nanoparticles in the process of electrocoagulation reduced energy consumption, increased electrode surface and received more energy at lower current intensities. Therefore, the coating can be mentioned as a suitable innovation to improve the performance of the electrical coagulation process.

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