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RESEARCH ARTICLE

EVALUATING THE EFFICIENCY OF PHOSPHORUS REMOVAL IN AQUEOUS SOLUTIONS BY FENTON METHOD

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ABSTRACT

Background and Aim: A brief look at the history of wastewater treatment shows that wastewater treatment began in the twentieth century and with the removal of suspended and organic matters from the wastewater. However, with the passage of time and the emergence of inappropriate environmental conditions, experts decided to do to a fuller treatment of the wastewater. Phosphorus is essential for the growth of algae and other living organisms, but due to eutrophication in waters and various complications that it creates, it is known as a severe pollutant. Given the importance of the issue, this research is conducted to evaluate the efficiency of phosphorus removal from aqueous solutions by Fenton method.
Materials and Methods: This study is a descriptive study that the removal of phosphorus from aqueous solutions by the Fenton was carried out by preparation of the water solutions. The studied concentrations of phosphorus were in the range of 1-5 mg/l, pH 3-9, contact times 5 to 30 min, and the Fenton ratios in the range of 1:1 and 1:5 and 1:10.
The Results: The optimum obtained values of the studied parameters to maximum 80% removal, the phosphorus concentration 3 mg/l, pH = 4, Fenton ratio (H_2O_2: Fe) =1:5 and the contact time of 20 min.
Discussion and Conclusion: According to experiments conducted in this study, it was found that this method is effective in removing phosphorus from aqueous solutions.

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INTRODUCTION

A brief look at the history of wastewater treatment shows that wastewater treatment began in the twentieth century and with the removal of suspended and organic matters from the wastewater. However, with the passage of time and the emergence of inappropriate environmental conditions, experts decided to do to a fuller treatment of the wastewater (Ghanizade, Ghader). Due to the geographical situation of Iran and lack of accessible water resources, it is very essential to control pollutions entering such resources, especially by reducing the concentration of input pollutants into them. Receiving waters such as lakes and rivers are places for the sewage and wastewater disposal. Nutrients such as nitrogen and phosphorus are among the pollutants that can enter the receiving waters (McGhee Terence, 1991). Phosphorus compounds are among the potential pollutants of receiving waters that enter such resources through different wastewaters (Chapra, 1997). Phosphorus is essential for the growth of algae and other living organisms, but due to eutrophication in waters and various complications that it creates, it is known as a severe pollutant. In various methods of wastewater treatment in country different types of pollutants including phosphorus compounds are reduced, but in some cases this reduction is not sufficient or because of the importance of this factor, even low levels of it in discharged wastewater is of great significance. There are different forms of phosphorus in wastewater, which are divided into soluble and particulate forms (with regard to smoothing particles through 0.45 micron filters) according to the physical characteristics and into orthophosphate, polyphosphates and organic phosphate with respect to the chemical properties. Available statistics indicate the presence of 20 mg/l phosphorus compounds in Tehran wastewater in average which is a very important figure in the environment with regard to the high volume of produced wastewater

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(Chapra, 1997). Phosphate as a by-product in various medical applications resulting from human activities enters into water. Ultimately, due to the leakage of minerals or mines rocks in natural processes - degradation, detergents, industrial wastewater and as an important element of domestic wastewater, phosphate enters into groundwater or surface water. For the creation of algae blooms only the concentrations of 0.005 to 0.05 mg/l phosphate in terms of phosphorus are needed. The standard in drinking water is 0.2 mg/l and the standard for discharging wastewater into surface waters is 6 mg/l (Samadi *et al.*). Biological treatment processes alone cannot eliminate the resistant organic matters, so additional treatment is required to remove them. Chemical treatment methods based on the production of hydroxyl radicals (OH-) are known as the advanced oxidation process (AOP). Organic matter oxidation with Fenton solution which is known as Fenton reaction is also one of the advanced oxidation processes (United Nations Environment Program, 1996). Advanced oxidation is applied for the oxidation of both organic and inorganic matters. One of the chemical oxidation processes used in the treatment of water and wastewater is Fenton process which has become more practical in recent years due to the production and consumption of toxic and late biodegradable pollutants. This is a kind of oxidation-reduction reaction in which the metal ion of an electron is transferred. Wise and weber proposed the first reaction as follows:

Fe+2 + H2o →Fe+3 + O- + OH

In a study conducted by BijanBina (2003) in Sweden on examining the efficiency of a hydrogenic wastewater treatment system in the removal of nutrients it was shown that anoxic tanks were capable of removing 72% nitrogen and 42% phosphorus (Ina, 2005). Mousavi *et al*. (2006) studied on synthetic wastewater and the efficiency of Fenton oxidation process in reducing water pollution by detergents and showed that Fenton process was applied at high concentration of detergent in aqueous solution while in the past this process was mainly examined at low concentration of detergent (Mousavi *et al*., 2010) Mohammad Agha Nejad *et al* (2008) conducted a research on the efficiency of wastewater treatment plant of combined cycle power plant of Khoi and the optimization of phosphorus removal in it through aerobic-anaerobic method. The results showed that the system was not successful in the normal removal of phosphorus but it was possible to improve phosphorus removal efficiency by adjusting some parameters (Aghanezhad *et al*., 2009). The rate of oxidation particularly depends on the amount of H_2O_2 consumption. Since hydrogen peroxide is a main effective factor in Fenton process and hydroxyl radical production is the result of its presence and on the other hand hydroxyl radical has an unknown nature in the oxidation of organic matters, it seems necessary to determine the concentration of H_2O_2 and to obtain the optimal ratio of hydrogen peroxide to organic matter according to laboratory tests (Ina, 2005). This study aims to investigate the feasibility of phosphorus removal through Fenton process in the aqueous media.

MATERIALS AND METHODS

This study is an experimental study that was performed to remove phosphorus from aqueous solutions. This is an empirical study (Experimental) conducted to remove phosphorus from aqueous solutions. Water samples needed for the tests were prepared synthetically and the desired tests were carried out according to the standard methods reference for water tests in terms of work method and accuracy. In order to determine the sample size, full fact method was used.

Design of experiments

This study had 4 variables, the Fenton ratio, and phosphorus concentration of solutions, pH, and the contact times than the removal of each of the variables in the experiment was determined. All experimental tests were repeated 2 times and the mean of results is presented. The number of analyzed samples was 75. Phosphorus concentrations in the initial and final solutions were analyzed by using a direct reading spectrophotometer, Dr-5000 and Excel software is used to create a diagram. P concentration between 1-5 mg/l. pH between 3 to 9 and H_2O_2: Fe (1: 1 and 1: 3 and 1: 5 and 1: 7 and 1: 9 and 1:10) and contact time is from 5 to 30 minutes was considered. In the first part we average value of each of these variables were chosen to get the optimum amount means that the amount of phosphorus 3 mg, Ph value of 5, the ratio of H_2O_2: Fe = 1: 5 and 20 minute contact time was considered.

Solutions needed

Ammonium molybdate

25 g (MO_7O_{24} ($NH_4.4H_2O$) 6) in 175 ml of distilled water solution. The caution of 280 cc of concentrated sulfuric acid in 400 mL of distilled water and it's cold. Solution of ammonium molybdate added to the solution and its volume bring to a liter with distilled water.

Stannous Chloride

2.5 grams of H_2O_2 $Cacl_2$ in 100 cc of glycerol solve.

Phosphate standard solution

219 mg of anhydrous KH2PO4will be solve in distilled water in a one-liter Jorje lab flask to reach to the volume. It will be the solution of 50 mg/l in terms of P.

Method: 4 cc of ammonium molybidate solution is added to 100 cc of the sample and is shaken well. Then, 0.5 cc or ten drops of stannous chloride is added to it and is shaken well. After 10 minutes but before 15 minutes the rate of absorption or the sample flow percentage is obtained by spectrophotometer at a wavelength of 690 nm. Standard solutions of 0.1, 0.3, 0.5, 0.7, 0.9, and 1 mg/l phosphorus are provided and the rate of absorption or the flow percentage of each one is obtained according to the above method. The standard curve is drawn and the concentration of sample phosphorus is calculated in milligram per liter of phosphate ion.

Experiment

In the first section, different initial concentrations ofphosphorus (1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5 and 5 mg/l) were contacted by a H_2O_2:$FeSO_4$ ratio= 1:5. Other study parameters include of pH=5, and the contact time =20 min.In tow stage, 7 pH values of 3 to 9 were tested. It should be noted that the optimum concentration of phosphorus (3 mg/l) was obtained from previous experiments was used as the phosphorus concentration of solution. Again, the contact time 20 min and the ratio of Fenton 1:5 were used.In the following, a series of experiments were implemented to determine of optimum Fenton ratio. The optimum ratio of Fenton in the range of 1:1 and 1:3 and: 5 and 1:7 and 1:9 and 10:1 were determined. In this way, the phosphorus concentrations of 3 mg/l and pH 4 were used. The contact time of these experiments was 20 min. In this section, The contact times of 5 to 30 min were tested.the previous obtained results of the experiments, p concentration=3 mg/l, pH=4, and the H_2O_2: $FeSO_4$=1:5 were applied for determination of the optimum contact time.

RESULTS

Analysis result of P provide by using PH, contact time and other factors. Fig. 1, illustrates the effect of initial phosphorus concentrations on the phosphorus removal efficiency by Fenton process and the most phosphorus removal was occurring in the phosphorus concentration of 3 mg/l.Fig. 2 displays the effect of the pH changes on the phosphorus removal from aqueous solutions. The maximum percentage of phosphorus ions were removed at pH = 4. This figure indicates that by pH increasing of the solutions the phosphorus removal efficiency was reduced. Fig. 3, demonstrates that the increase of the H_2O_2:$FeSO_4$ ratio, the decrease the phosphorus removal efficiency. In addition, the highest phosphorus removal efficiency was obtained in the ratio of 1:5. Fig.4, displays the impact of contact time on the phosphorus removal efficiency from aqueous solutions. As can be seen from this figure, it shows the phosphorus percentage removal was increased with increasing contact time and the removal efficiency was constant after 20 minutes.

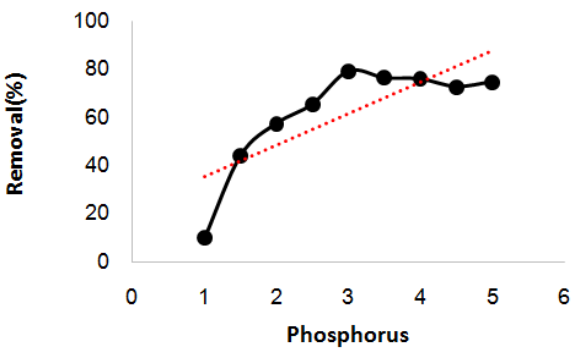

Fig.1. Effect of phosphorus concentration (Time: 20 min, ph: 5, H2O2:Fe=1:5)

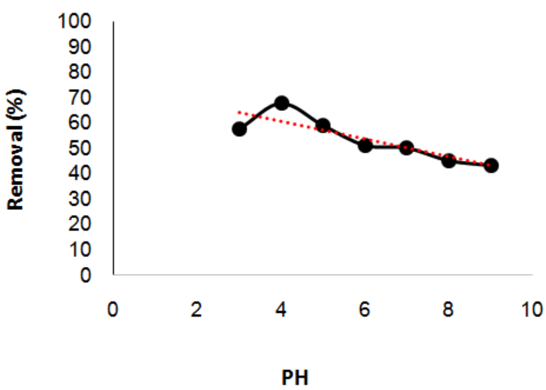

Fig.2. Effect of ph change on the phosphorus removal (Time= 20 min, Phosphorus Conc. = 3 mg/L, H_2O_2: Fe= 1:5)

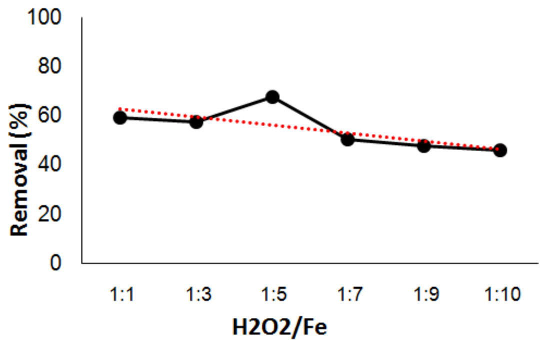

Fig.3. Impact of Fenton ratio on the phosphorus removal (Time= 20 min, pH= 4, phosphorus Conc. = 3 mg/L)

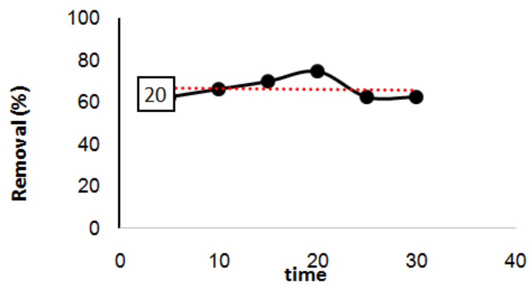

Fig.4. Contact time effect on the phosphorus removal, (phsphorus Conc. = 3 mg/l, pH=4, H2O2: Fe= 1:5)

DISCUSSION

Effect of initial phosphorus concentrations

In this study it was observed that the percentage of phosphorus removal (78%) with increasing concentration of phosphorus removal rate increases and the level of 3 mg / l, and then the removal rate increases. However, totally the removal trend of the phosphorus from the aqueous solutions was increased by increasing of the initial phosphorus concentrations.

Effect of pH

The pH of the solution is an important variable, which affect the removal of phosphorus. Hence, the effects of the solution pH were studied in the range of 3 to 9. Results that are shown in Fig.2 indicates that with the increasing the solution pH, the phosphorus removal efficiency was decreased. The maximum phosphorus removal (69%) was occurring at the initial pH 4. Therefore, it is well established that the phosphorus was highly removed in acidic conditions. The highest efficiency of phosphorus removal at acidic condition may be related to the presence of the highest value of dissolved iron in water at such conditions. In this situation, the highest hydroxyl radicals are formed (Poinern *et al.*, 2011). In this study the pH 4 was chosen as the optimal pH in the Fenton reactions. In the Fenton reaction, the hydroxyl radicals are the main oxidation agents, which are produced by the decomposition of hydrogen peroxide catalyzed by metal ions.

This lower removal efficiency may be due to the exchange interactions between p− and OH− groups in the solutions (Tomar *et al.*, 2013). Meijuan *et al.* (2007) observed similar results (Xu *et al.*, 2007).

Effect of Fenton ratios

Fig.3, in all studied ranges, except the 1:5 ratios, the trend line of the phosphorus removal is decreasing by the increasing of the Fenton ratios. Removal of phosphorus in the ratio of 1:10 and 1: 9 of H_2O_2: Fe is negligible low because hydroxyl radicals produced was not sufficient for fluoride removal from the aqueous solution and the iron hydroxide precipitation was observed.

Effect of contact times

Phosphorus removal efficiency as a function of contact time is shown in Fig. 4. This Figure shows that by the increasing of the contact times, the phosphorus removal efficiency was increased up to 75% at the contact time of 20 min. This may be due to the diffusion of hydroxyl radical into the solution (Rafique *et al*., 2012). Argun *et al*., observed similar results in a heavy metal removal study (Argun *et al*., 2008).

Conclusion

The proposed method has successfully been applied to the removal of phosphorus in different aqueous solutions containing a wide concentration range of the phosphorus. The simplicity of this method, short time and the use of safer chemicals and high removal efficiency, demonstrate the high potential for routine phosphorus removal from water samples. These values were then optimized individually double-tested and showed a maximum 80% phosphorus removal. The obtained results showed that Fenton process could be an effective method for the removal of phosphorus from aqueous solutions.

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