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Nitrate removal from drinking water in laboratory-scale using iron and sand nanoparticles

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Abstract

In recent years, entering of wastewater of agricultural activities and rural and urban residential centers to the water supplies has caused significantly to increase nitrate of surface and groundwater as well as various diseases. In this regard, a major step has been taken in eliminating contaminated water by development of modern technologies and it has been attempt to make fresh water accessible for people. This research has been conducted to investigate nitrate removal from urban waters through elemental iron Fe^0 as a suitable, cheap and easy strategy to treat waters contaminated by nitrate. In this study, the effect of parameters pH, sand and iron particles on the amount of nitrate removal was tested. Also, mixture of iron and sand nanoparticles was used to increase efficiency and specific area. The results showed that the efficiency of iron nanoparticles was 65% in the first 20 min and it was 45% in the next times due to increase of pH resulted from reaction. Also the results showed that, the initial pH of solution is important to achieve the maximum efficiency of nitrate removal.

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Introduction

Surface water and groundwater pollution by nitrate is known as a serious problem in many points of the world and nitrate is the most common chemical contaminant of groundwater in the world. Chemical fertilizers, animal wastes and improper disposal of human and animal wastes are important sources of nitrate entering the groundwaters (Hoodak, 2000; Ward *et al.,* 2005). Nitrate is harmless by itself but, when nitrite is regenerated, public health of human is threatened in terms of gastrointestinal and liver cancers. In infants under six months, nitrate is regenerated to nitrite which is able to combine with hemoglobin to produce metaglobine and makes the blood dark due to lack of oxygen transport to the tissues (Kapour *et al.,* 1997). According to EPA and WHO guidelines, the maximum allowable concentration of nitrate is 10 mg/L by nitrogen and 50mg/L by nitrate have been accepted for public water system (teal *et al.,* 1998). Nowadays, use of nanotechnology is increasing to remove some important contaminants such as Nitrate ion from water resources. Nitrate reclamation using chemical denitrification by nanoparticles of zero-valent iron is expanding in decomposition of nitrate ions from water resources due to rapid reaction and high efficiency, being frequent and cheap (Noibaktep, 2010). When nanoparticles of iron Fe^o is applied on water containing nitrite and nitrate ions, divalent iron (Fe^{2+}) gas and ammonium (NH_4^+) or N_2 are created according to the following reactions. Produced gases are released in the environment and are removed from water resources using treatment processes (Lee *et al.*, 2006).

$$5Fe^0 + 2No_3 + 6H_2O \rightarrow 5Fe_2^+ + N_2 + 12OH$$

$$4Fe^0 + No_3 + 7H_2O \rightarrow 4Fe^{2+} + NH_4^+ + 10OH$$

More than 20 projects have been carried out in since 2000 in the U.S. Also, some other projects have are being done in Northern America, Europe and Asia (Environmental Protection Agency of America, 2008). The way of production and use of nanofilters have been the topic of many researches in the recent 10 years and this technique has been accepted as one of the newest methods to treat and elimination of water and wastewater (Chou *et al.*, 1998; Gianfa, 2010).

Huan *et al.* (2004) studied the Nitrate treatment by nanoparticle of iron in different states. In controlled pH of 2, 3, 4 about 95% of nitrate has been removed. Loui *et al.* (2005) conducted a research about use of iron nanoparticles, iron microparticles and iron nanoparticles coated by copper in reclamation of the solution containing non-buffering nitrate. Their results showed that the use of bimetallic nanoparticles has a higher potential in conversion of nitrate to nitrite or ammonium compared with iron nanoparticles and iron microarticles respectively.

Sucrou *et al.* (2006) investigated the nitrate removal and pesticides in polluted waters in Turkey using denitrification. A submerged biological reactor was used in this study to remove nitrate from drinking water and the initial amount of nitrate decreased by 48%. Fenali and Roberts (1998) showed that, bimetallic nanoparticles of Fe/Cu increases reclamation rate of the pollutant 1-TCA ،1 ،1 compared with bimetallic nanoparticles of Fe/Ni and NZVI. In this research, after producing bimetallic nanoparticles of Fe/Cu, the effect of various percentages of different percentages of copper coverage in iron nanoparticles on the trend of reclamation of nitrate in drinking water in the presence of other interfering ions has been investigated.

Mohammadi *et al.* (2011) by investigating the effect of pH and sulfate in nitrate removal from zero iron nanoparticles showed that, in concentration of 200 mg/l of nitrate with pH 5, 6, 7 the amounts of nitrate in 60 min are 88, 72 and 62 % respectively. The amount of nitrate removal in the concentration of 100 and 300 mg/l and pH=6 has reached from 60 to 83%. In concentrations of 200 and 300 mg/l of nitrate along with the same concentration of sulfate in pH=7, the efficiency of removal in the situation without sulfate is important so that, in lower pH there is a higher amount of removal.

With regard to the high importance of controlling the nitrate content of drinking water in preservation of the human health, so, the objective of this research is to investigate the possibility of nitrate removal from drinking water and water solutions by iron particles under different temporal conditions, pH, amount of nano material and nitrate concentration.

Materials and methods

Study area

This experiment was conducted in nanotechnology laboratory of Tabriz to remove nitrate from drinking water in 2012 through following approaches.

Iron nanoparticles

The size of studied particles in laboratory was 20 to 30 nanometer with a specific area by 40-60 m^2/g and purity higher than 99% , black color, spherical shape and actual density of 5.1 g/cm. the studied iron nanoparticles was provided according to the presented method by Marotto *et al.* (2009) as below: At first, sodium borohydride ferric chloride with a ratio of 1 by 3 were weighted and then, the created solution was cluttered in a 2000 CC beaker and after 20 minutes having contact, the solution was passed through a Whatman filter paper, then it was placed in the environment space and dried. Meanwhile, for sedimentation of the black particles of zero valent iron nanoparticle, starch adhesive was used.

Sand

Sand is used to increase specific area of iron nanoparticles. The experimented sand had an approximate diameter by 2 mm.

Synthetic solution

Synthetic solution containing Potassium nitrate (KNO_3) with certain concentrations of 50, 25, 10 and 5 mg/l was poured into a 500 ml jar glass. Then, nano materials with the certain amount of 5 g/l (in this experiment, 2.5 gram in 500 ml) were added to the contaminated water by nitrate. Then, jar beakers were placed in the jar test device and was adjusted on 250 rpm. After that, sampling was done at time stages of 10, 20, 30, 40, 50 and 60 min and pH= 6, 7, 8.

Results

Generally, pH of surface waters and drinking waters has been reported between 5.5 and 8.3 and accordingly, pH in this experiment has been considered between 6 and 8, and considering that, nitrate reclamation under alkaline conditions takes place hardly since, in high pHs sediments and considering Hung's study in which it has been reported that, reclamation takes place easily in pHs less than 4 (Ward, 2005). In this experiment, pHs less than 8 were studied. In order to measure nitrate DR 5000 device was used and to measure pH, desk pH meter was used. All experiments were done in temperature of 24 °C.

The results of neutral pH variations demonstrated that, nitrate removal has showed its maximum efficiency in the last 10 minutes (Table 1).

Table 1. Nitrate amount variations in different times and neutral pH.

Nitrate concentration (mg/l)	Time
50	0
47	10
27	20
20	30
10	40
1	50
0	60

The results showed that, after 60 minutes nitrate was completely removed by nanoparticles and removes almost 99.6% of the pollutions.

Since, in this experiment the water pH was prior and its goal was to remove nitrate in urban and drinking waters, so, the samples were assessed by measuring the pH variations. The results of measurements showed that, by decreasing pH over the time, nitrate removal increases (Table 2).

Table 2. Nitrate variations in various times and pHs in the presence of iron.

Time	Nitrate concentration in pH=6	Nitrate concentration in pH=7	Nitrate concentration in pH=8
0	50	50	50
10	41	42	42
20	23	26	28
30	16	23	24
40	3	12	13
50	2	6	6
60	1	5	6

The results showed that, after 60 minutes and in pH=6, 7, 8 nitrate was not completely removed by iron nanoparticles and removes almost 98.6% of the pollutions (Figure 2).

Generally, in pH<6 nitrate removal is done easily (teal *et al.,* 1998). According to Figure (2), nitrate removal by zero valent iron in pH=6 is higher and acceptable so that, in the first 20 minutes nitrate removal reached 60%. But, in high pHs due to iron hydroxide sedimentation, the removal efficiency decreases and in the final time of sampling, the removal remains incomplete and removes almost 95% of the pollutions.

At the next stage, Adding sand to nanomaterials caused to increase specific area and increase of nitrate removal efficiency. Sand particles were added to the nano materials with a ratio of 1 by 1 at the next stage. When iron nanoparticles are added as 2.5 per 500 ml in polluted water, 2.5 g of sand also is added to the solution and then, the mixture is riled in jar test device with 250 rpm for 60 minutes. This action accelerates the reclamation. According to Figure (3) it is observed that, sand particles accelerate reclamation and make up the defect of nanoparticles in their efficiency in high pHs and the removal has been carried out with the maximum removal (Table 3).

Table 3. Nitrate variations in different times and pH in the presence of iron and sand particles.

Time	Nitrate concentration in pH=6	Nitrate concentration in pH=7	Nitrate concentration in pH=8
0	50	50	50
10	38	38	39
20	21	23	24
30	15	17	17
40	4	6	8
50	2	2	3
60	0	0	0

Achieved results showed that, after 60 minutes and in pH=6, 7, 8 the nitrate was completely removed by iron nanoparticles in the presence of sand and removes almost 100% of the pollutions (Figure 3).

Discussion

Achieved results showed that, iron nanoparticles in pH=7 (neutral) remove nitrate from polluted water but, in alkaline pHs the removal remains incomplete and cannot remove nitrate completely and iron nanoparticles have the highest efficiency in acidic Ph (Figure 1). In all conducted tests, nitrate removal reached its maximum efficiency in the first 20 minutes and nitrate removal was carried out slowly which was due to increase of pH in the samples after the first 20 minutes of the reaction (Loui *et al.,* 2005). Meanwhile, decrease of nitrate concentration of the sample after this period affects the reaction negatively. One disadvantage of this experiment is creation of ammonium gas caused by the reaction of iron with nitrate. This gas is volatile and is removed from the environment immediately and appropriate use of health facilities plays an important role in controlling these gases. About the advantages of this research also we can mention to being cheap, ease of the work, high accuracy and high efficiency.

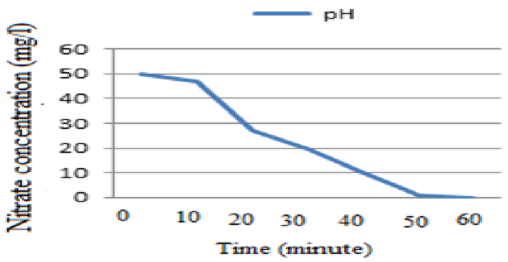

Fig. 1. The effect of iron nanoparticles with certain amount after 60 minutes.

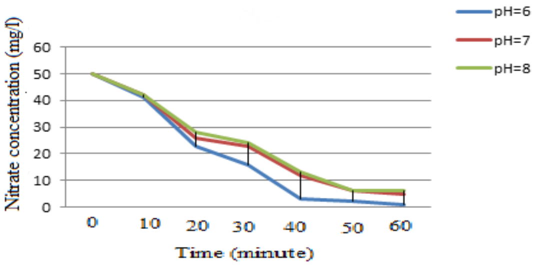

Fig. 2. Assessment of the effect of iron nanoparticles in different pH on nitrate removal.

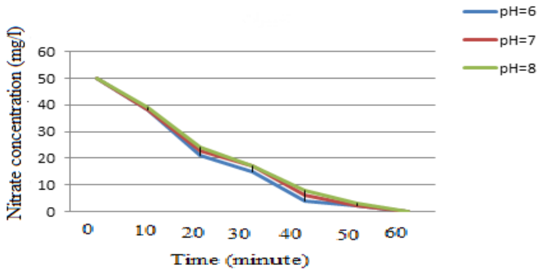

Fig. 3. Assessment of nitrate removal efficiency by nanoparticles in the presence of sand in different pH.

Conclusion

An important pollutant of drinking water is nitrate and its removal using nanotechnology is one of the most appropriate and cheapest methods. Water nitrate removal using modern technology can prevent blood and parasitic diseases. In this study, the trend of water nitrate removal by iron and sand was investigated based on which iron nanoparticles in alkaline pHs showed the lowest reclamation while, removal action was completely done in acidic pHs. Due to replacement of acidic state with alkaline state over the time, so, reclamation remains incomplete before determined time. It is recommended that, sand particles to be added to the iron particles with the ratio of 1 by 1 to make up this defect and the reclamation takes place completely.

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