Original Article

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Effect of Iron Oxide Nanoparticles for the Removal of Coliform Bacteria from Contaminated Water

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ABSTRACT

BACKGROUND AND OBJECTIVE: Iron oxide nanoparticles at low concentrations can act as a source of iron ions required microorganisms but high concentrations can cause stress and cell damage in bacterial cells are reduced cell growth. The effect of iron oxide nanoparticles were studied remove coliform bacteria from contaminated water.

METHODS: In this experimental-laboratory study synthetic municipal wastewater by adding 5 mL to 60 liters of water in urban distribution network was built. Every time in half a liter of synthetic samples 100, 250, 500, 1000, 2000, 4000 and 6000 ppm of iron oxide nanoparticles were added at the time of zero, 20, 40, 60, 80 and 100 minute samples and with method 15-tube was tested.

FINDINGS: With increasing concentrations of iron oxide nanoparticles from 100 ppm to 6000 ppm the most likely number of coliforms per 100 ml (MPN) average removal rate from 70 percent to 82 percent synthesized samples. (Number of MPN) and removal efficiency at 0, 20, 40, 60, 80 and 100 minutes, was (660.52) 0% (330.42) 48.83, (317.00) 53.75, (200.14) 68.48, (161.66) 73.96 and (128.04) 80.16 respectively. Significant differences between time and removal of coliform bacteria (P=000.0) was observed. Maximum efficiency in the contact time of 100 minutes and 6000 mg/L concentration of iron oxide nanoparticles to 100 percent.

CONCLUSION: The results showed that with increasing contact time and increasing concentrations of nanoparticles, remove the MPN samples is increasing.

KEY WORDS: *Water, Disinfection, Iron Oxide Nanoparticles, Coliform Bacteria*.

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Introduction

The most common methods for water disinfection are chlorine, chloramine and chlorine dioxide. Other disinfectants include ultraviolet rays, chlorinated lime, bromine, iodine, and silver (1). Commonly used methods for drinking water treatment can effectively control microbial agents, there is a link between water disinfection methods such as chlorination and ozonation and the production of disinfection products (DBPs) (2).

Some physical treatments such as ultraviolet radiation, ultrasonic waves and membranes, although have many of the criteria for a good disinfection, they cannot provide sufficient residuals for systems with a wide distribution network or long shelf-life. Alternative chemical disinfectants including chloramine, chlorine dioxide, bromine, titanium dioxide and potassium permanganate have been limited due to the low efficiency, high prices and the production of poisonous disinfection products (3). Therefore, new technologies for the effective disinfection of water need to be explored and identified (2). The nanoparticles in the life cycle and the ecosystem exhibit the lowest toxicity. Therefore, the use of these agents to combat pathogenic microbes can be a good choice.

Metal oxide nanoparticles exhibit different antibacterial properties based on surface to volume ratio. Gram-positive bacteria exhibit more resistance than gram-negative bacteria against metal nanoparticles, which can be related to the cell wall structure (4). The difference between the negative charge of the microorganism and the positive charge of the nanoparticle acts as an absorbing electromagnet between the microbe and the nanoparticle, and causes the nanoparticle to be bonded to the cell surface, this connection often correlates with the interactions between the cell membrane and the nanoparticle, and thus can Causing cell death (5).

Iron oxide is found abundant in nature and synthesized in laboratories. Among these nanoparticles, hematite, magnetite and magnet have special properties for use in the biomedical field. Magnetic Resonance Imaging (MRI) nanoparticles have been widely used as a source of transparency, markup and isolation of animal cells, targeted drug delivery, and therapeutic methods for the treatment of cancer tumors (6-10).

Also, these nanoparticles have synergistic effects with anticancer drugs and can make them effective (11). Iron oxide nanoparticles have been identified as an effective absorbent for the removal of pollutants from the aquatic environment and in many cases have been used to remove organic pollutants and heavy metals from the aquatic environment (12-14). In addition to other uses of iron oxide nanoparticles, recent findings have opened another window in the application of iron oxide nanoparticles. These compounds have antimicrobial properties. There are numerous reports on the antimicrobial effects of these nanoparticles on gram-positive and gram-negative bacteria and even fungi (15-17).

Iron oxide nanoparticles in the field of microbiology have been used to stabilize microbial cells in industrial processes, wastewater treatment or removal of microbial contaminants (18-20). Nanoparticles of iron oxide have become more respected by the researchers (27-21) due to their proper magnetic properties, very low toxicity, high biocompatibility and relative ease of synthesis compared to other nanoparticles. Nasiri and colleagues showed that oxide nanoparticles have significant antibacterial properties against Escherichia coli and Staphylococcus aureus (28). Therefore, this study aimed to investigate the effects of iron oxide nanoparticles on the removal of coliforms from contaminated water.

Methods

This experimental study was carried out between October 2006 and December 2014 after obtaining permission from the Ethics Committee of Kerman University of Medical Sciences with the code 315/93 at the Environmental Health Engineering Research Center. US NANO branded iron oxide nanoparticles were purchased in the form of a powder of 20-30 nm in size. First, a synthetic solution contaminated with chlorophyll was made by adding 5 ml of municipal wastewater treatment plant to 60 liters of water distribution network that was chlorine free and chlorophyll-free.

Simultaneously, in 7 bottles containing half a liter of synthetic sample, different amounts of 100, 250, 500, 1000, 2000, 4000 and 6000 mg/L of iron oxide nanoparticles were added and from each container at different times 0, 20, 40 60, 80 and 100 minutes of sampling were taken and all of the above steps were repeated three times. In total, 126 microbial samples were prepared according to different sampling rates and times, including three replicates. The samples were tested in a 15-pipe method for water and wastewater testing. In accordance with the 15-tube method, 5 tubes were prepared for each dilution (10 mL, 1, 0.1, 5).

In the first 5 tubes, 10 ml of Lauryl Sulphate broth was cultured in two concentrations and in 10 subsequent tubes, 10 ml of LaRiL broth medium was placed in a concentration and then in the first 5 series, 10 ml of the sample, in the second 5 series, 1 ml Sample and in the third series, 0.1 ml of the sample was added. The mixture inside the tubes was mixed with a gentle shaking of the tube. Sample tubes and culture media were incubated for 48 hours at 35 ° C. Gas and acid production in tubes was considered as a positive result.

To confirm the presence of chlorophyram from all positive tubes of this stage, a sterile loop was cultured in tubes containing Berlyanet Green medium. The tubes were heated to 35°C for 48 hours. The formation of gas at any rate in the Durham pipes at any time of 48 hours of heating showed a positive response at this stage. The results were reported as the most likely number of coliforms in 100 ml (Most Probable Number=MPN).

According to the Iranian drinking water quality standard, the total water distribution in the distribution network should not exceed 3 (29). Sampling and testing were carried out according to the methods in the Standard Methods for Water and Sewage Testing (30). The results were first tested to ensure that the data were normal with Kolmogorova -Smirov test and then one way ANOVA test and analyzed by SPSS software. $P<0.05$ was considered significant.

Results

The results of the study showed that the amount of 6000 mg/L of iron oxide nanoparticles could decrease the MPN of the synthetic sample from 210 after 100 minutes of contact time to zero (table 1).

By increasing the contact time and increasing the concentration of nanoparticles in all the amounts, the MPN removal of the samples had a rising course so that the maximum removal efficiency of the coliforms at the contact time of 100 minutes and the concentration of 6000 mg/l of nanoparticles of iron oxide was obtained. By increasing the concentration of iron oxide nanoparticles from 100 mg/l to 6000 mg/l, the average removal percentage of MPN synthetic samples increased from 70% to 82%, as well as increased contact time from 20 minutes to 100 minutes led to increasing the average removal percentage of MPN of synthetic samples ranged from 49% to 80% (Fig. 1). There was a significant relationship between the contact time and removal of chlorophyll bacteria ($p=0.000$), as well as the concentration of nanoparticles and removal of chlorophyll bacteria ($p=0.000$). At a concentration of 6000 mg/L nanoparticles, the logarithm of the number of residual bacteria compared to the contact time is a straight line, which indicates that the elimination of coliform bacteria by the iron oxide nanoparticles suspension follows from the first-order reaction rates (Fig 2).

Table 1. Mean MPN of contaminated synthetic water samples at different times and different amounts of iron oxide nanoparticles (mg/L)

MPN concentration / Duration of effect (min)	100 mg/L Mean±SD	250 mg/L Mean±SD	500 mg/L Mean±SD	1000 mg/L Mean±SD	2000 mg/L Mean±SD	4000 mg/L Mean±SD	6000 mg/L Mean±SD	MPN Standard of Drinking	P-value[b]
Synthetic sample	1143.66±392.59	313.33±63.5	376.66±151.76	1373.33±392.59	793.33±219.39	793.33±219.39	210±36.05	#3	-
20	376.66±151.76	216.66±40.41	193.33±40.41	793.33±219.39	150±17.32	476.66±109.69	106.33±25.69	#3	0.003
40	453.33±150.11	216.66±40.41	180±55.67	793.33±219.39	150±17.32	370±160.93	55.66±8.73	#3	0.002
60	326.66±40.41	193.33±46.18	146.66±20.81	413.33±109.69	116.66±11.54	183.33±63.5	21±4	#3	0.000
80	326.66±4041	153.33±15.27	146.66±20.81	276.66±63.5	120±17.32	99.66±17.89	8.66±7.57	#3	0.000
100	226.66±11.54	120±17.32	109.66±30.5	276.66±63.5	94.33±15.5	69±17.32	0	*3	0.000
P-valuea	-	0.019	0.013	0.316	0.006	0.578	0.000		

*:Significant, #non-significant, P-value: concentration compared to the initial concentration: b P-value: time compared to synthetic sample

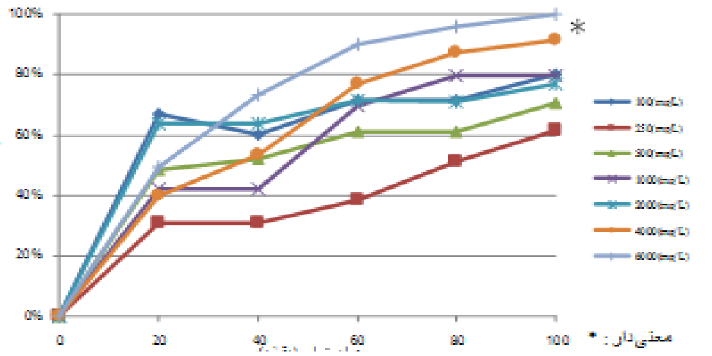

Figure 1. Process for removing MPN in different amounts of iron oxide nanoparticles at different times

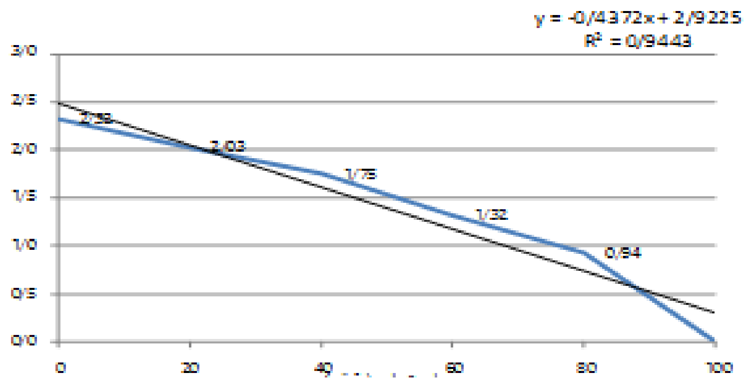

Figure 2. Changes in the logarithm of the number of remaining coliforms in 100 ml of the sample at 6000 mg/l of iron oxide nanoparticles

Discussion

According to the results of this study, the percentage removal of MPN from synthetic samples has increased with increasing iron oxide nanoparticle concentration and contact time. Also, at the time of contact with 100 minutes, the highest percentage was related to a concentration of 6000 mg/L of iron oxide nanoparticles, which resulted in the removal of 100% of the MPN of the synthesized sample. In studying the effect of CuO, TiO_2, and ZnO nanoparticles on the removal of gram positive and negative bacteria from urban wastewater, Malakootian and colleagues concluded that with increasing nanoparticle concentrations, the percentage of removal of bacteria increased and the concentration of 6000 mg /L has the greatest percentage of removal of the bacteria including Staphylococcus aureus, Bacillus subtilis, Pseudomonas aeruginosa and Escherichia coli (31). In the study of Miranzadeh et al., to investigate the effect of nanosilver on the elimination of coliform bacteria from contaminated water concluded that increasing the contact time with nanosilver causes further elimination of coliforms (32). Ebrahiminezhad and colleagues also found that in low concentrations (less than 20 µg/ml), Listeria monocytogenes could benefit from magnetite nanoparticles for further growth, probably as an iron source, but appears to be gradual with an increase in the concentration of antibacterial effects, and in 40 µg/ml magnetic nanoparticles have a significant antibacterial effect (33), which is consistent with the results of this study. The reason is that iron oxide nanoparticles at low concentrations can act as a source of iron ions needed for microorganisms and thus will be eliminated. However, high concentrations of these nanoparticles in microbial cells can cause cellular degradation by inducing tension and cellular damage (34). Huang and colleagues concluded that amine-functional magnetic nanoparticles were fatal to a wide range of bacteria (19).

In the study of the effect of iron oxide nanoparticles on the effect of antibiotics, Cotar et al. Concluded that iron oxide nanoparticles containing penicillin, streptomycin, erythromycin, kanamycium and cefotaxime had a lower inhibitory growth potential than any of antibiotics alone (35). The results are consistent with the emphasis on antimicrobial activity in this study. It seems that the binding of iron oxide nanoparticles to the cell wall of bacteria also disrupts cell membrane function and increases its permeability. This will increase the transfer of materials and improve the efficiency of industrial processes(34). At a concentration of 6000 mg/L nanoparticles, the number of remaining bacteria in comparison with the contact time is a straight line, which indicates that removal of coliforms by the suspension of iron oxide nanoparticles follows the first-order reaction rates. In the study of Miranzadeh et al., to investigate the effect of silver nanoparticles on the elimination of contaminated coliform bacteria, it was concluded that the removal of coliform bacteria by a nanosilver suspension follows a first-rate reaction (32), which is consistent with the results of this study. Therefore, with the help of the in vitro and the line equation for the concentration of 6000 mg/L nanoparticles (y= -1.9681x +165.3492), it can be concluded that in this indicated amount to reduce the number of bacteria to less than 3 of the 100 milliards in Iran's 3-116 Standard Code (29) about 49.82 minutes as contact time is essential that should be greater than the time of contact with water and chlorine, which is about 30 minutes (1), which requires a larger amount of the structures of the dock. The use of chemical disinfectants has been limited due to the production of toxic products (chlorine production of trihalomethanes and ozone in bromide and bromate production). In addition to having antimicrobial properties, iron oxide nanoparticles have been identified as an effective adsorbent for the removal of organic pollutants and heavy metals from the aquatic environment (12-14).

Therefore, iron oxide nanoparticles can be considered as an effective ingredient in the treatment of drinking water and sewage. Considering the other properties of these nanoparticles, such as its synergistic property, suitable magnetic properties, very low toxicity, high biocompatibility and relative ease of synthesis, the application of iron oxide nanoparticles in the removal of pollutants can be considered as a reliable and efficient method.

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