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Enhancing antibiotic degradation in a wastewater treatment process with (nZVI) activated potassium persulfate in Iraq

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Abstract

The objective of the study was to eliminate kanamycin and folic acid in wastewater using Nano Zerovalent Iron (NZVI) activated with potassium persulfate by means of sulfate radical (SO4 •-) generation. Effects of pH as well as concentrations of nano zerovalent iron, antibiotics and sulfate were analyzed to determine the optimal conditions for antibiotic degradation. Additionally, HPLC was applied to detect the concentration of antibiotics after treatment. The reaction time, K2S2O8 dosage, pH value, Fe0 dosage, initial concentration of kanamycin and folic acid were also investigated. The most efficient removal of kanamycin and folic acid were 98% and 93%, respectively

Keywords: nZVI; Persulfate activation; Radical oxidation

1. Introduction

Rapid urbanization and growing human activity have given rise to new toxic and hazardous chemical substances, the so-called "emerging pollutants", which may present unforeseen impact on ecosystems [1] Recently, antibiotics began to particularly draw peoples' attention as one of these emerging pollutants. [2] Wastewater from pharmaceutical industries, hospitals, livestock farms, landfill leachates and wastewater refineries is considered to be the major source of antibiotic discharge to the environment. And less than a 10% is modified by the body - an interesting fact when one considers that remainder amount is removed from human body without any alteration. [3] Wastes containing antibiotics are commonly released into the aquatic milieu due to water treatment plants accommodating failure in their removal [4] The unrestricted dispersion of antibiotics into the environment, that also contributes to the development of drug-resistant bacteria and their associated resistant determinants in aquatic environments. There are various ways for treating antibiotics in water sources. These procedures include advanced oxidation, ion exchange, ozonation and membrane technology processes, activated carbon adsorption, reverse osmosis (RO),and biological treatment. [3] The applications of high-efficiency processes in the industry is necessitated due to the escalating water source pollution.This combined advanced oxidation process was reported as efficient in the degradation of these antibiotics in aqueous solution, on a pilot scale experimental basis.This strategy also acknowledges the inherent complexities involved in antibiotic contamination, such as the generation of toxic by-products which are frequently overlooked when assessing only parent compounds [5]. (Svebrant et al., [6] This complete analysis will permit to better understand the overall treatment efficiency (Ewert et al., 2009) and potential ecological consequences [7]Many emerging pollutants such as antibiotics have complex chemical structures which make them recalcitrant in the environment, and thus are not completely removed through conventional wastewater treatment processes [8].Therefore this work examined the efficiency of new developed persulfate oxidation process in degradation of antibiotic i.e. folic acid and kanamycin existing within hospital wastewater.

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2. Materials and methods

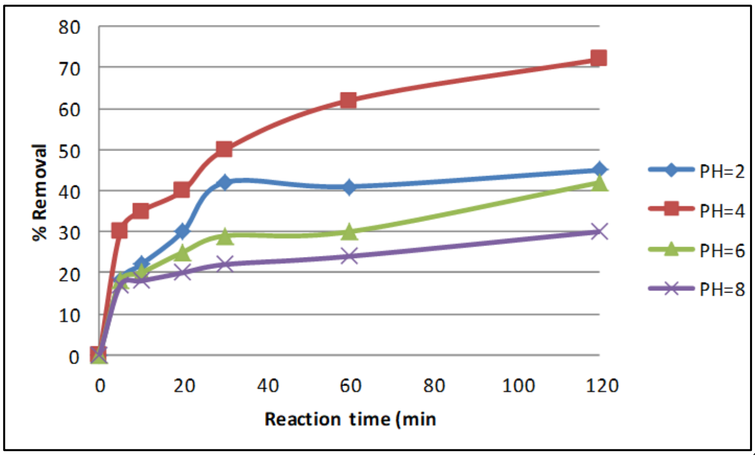
The removal of the studied antibiotics was compared under similar operational conditions by various oxidation approaches. The removal efficiencies of the persulfate/zero-valent iron (PS/Fe^0) system were the largest and smallest for the two antibiotics among the tested systems, suggesting its effectiveness was closely associated with operation conditions. Based on that, the PS/Fe^0 process was chosen for more detailed study as the best technology able to eliminate both folic acid and kanamycin. The optimization of the process was carried out and the effect of various experimental parameters on its efficiency were systematically studied.

3. Effect of Experimental Factors

3.1. Effect of Solution pH

The pH of the solution is considered as a key controlling factor in wastewater treatment processes, because it has great effect on chemical reactions and biological activities in the system [9]. The data suggests that adsorption of antibiotics gets higher as pH decreases, since shown in Figure 1.

The efficiency of antibiotic removal goes down in the alkaline pH compared to acidic conditions, and this can be justified as a result of simultaneous nano-iron particle precipitation and ferrous sulfate species having lower oxidation potential forms [10] The maximum removal percentage was found both at pH = 4 and it was equal to 72% and 87.9% for folic acid and kanamycin, respectively. The improved oxidation property is mainly attributed to the higher activity of persulfate and the production of sulfate radicals in acidic medium [11].



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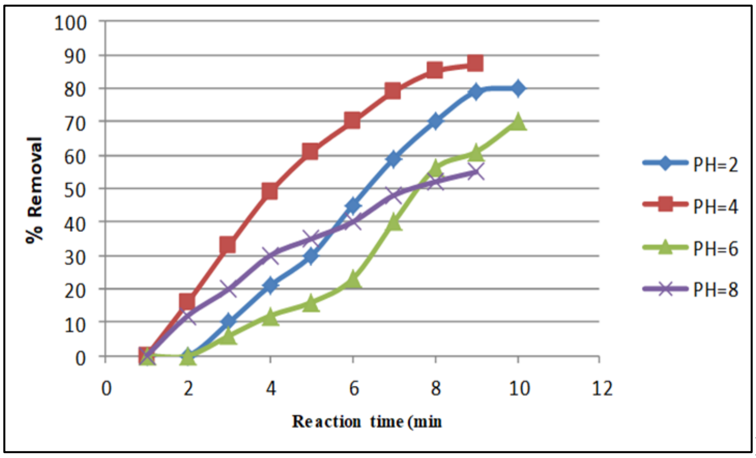

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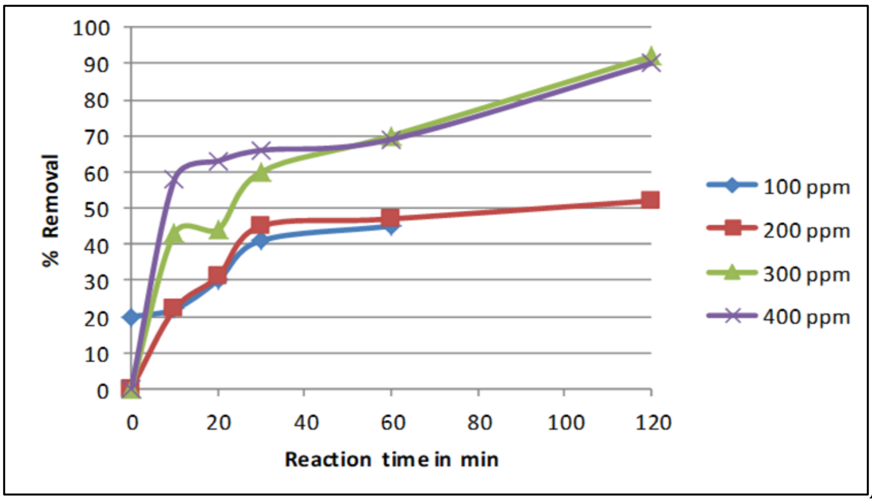
Figure 1 Effect of different pH on the removal efficiency of kanamycin A and folic acid B by Fe^0/PS

There is an even comparison of the removal efficiency performed by both antibiotics over acidic and alkaline pH ranges indicating that the pH is essential for the best adjustment of PS/Fe^0 oxidation process (Figure 1).

3.2. Influence of concentration of NZVI

displays the influence of varying Fe0 loadings (100, 200 and 400 ppm) on PS/Fe0 process in removing folic acid& (a), kanamycin&(b) by four times at optimum pH of 5–120 min. [12]



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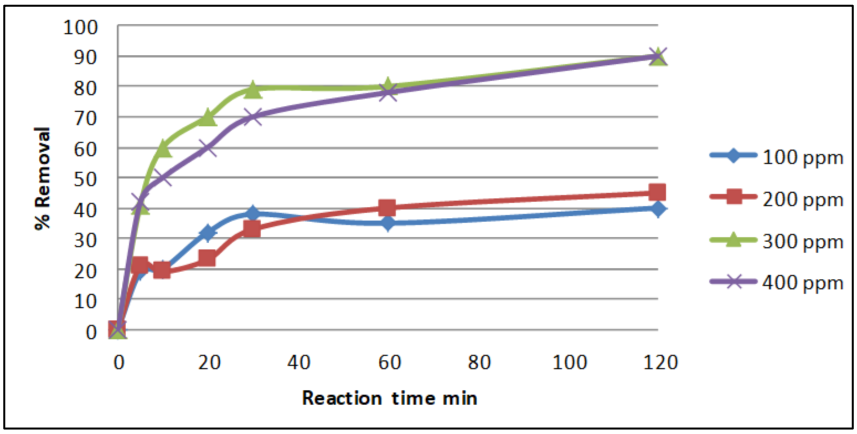

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Figure 2 Effect of Different Concentrations of NZVI on the Performance of the Removal Process of kanamycin (A) and folic acid (B) by PS/Fe0

The results are shown in Fig. 4, and they show that the removal efficiency of kanamycin or folic acid dose reached 90% at around 300 ppm. It exhibited a rising mode. The reason might be that the removal efficiency drops with the concentration increase (300,400 ppm), and the PS degradation increases obviously when more redox active sites are present.

. [13]. The results revealed that increasing Fe 0 doesn't have effect on process efficiency, where the increase of feces imitated increasement of antibiotic. This behavior is due to the interaction of SO4•- with additional iron cations and self-assembly of iron particles which occupies some of its reactive sites at high concentrations.

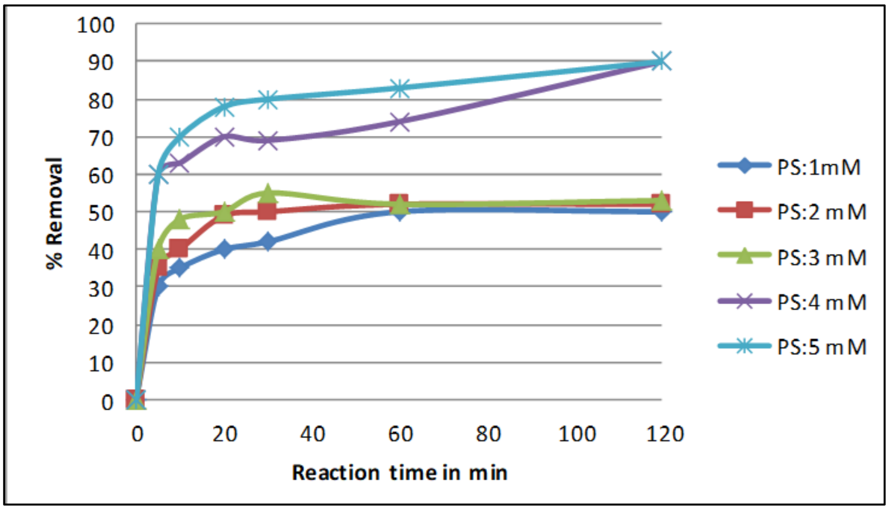
[14] The optima Fe(II)s concentration value was determined for the two tested antibiotics in order to achieve maximum removal efficiency during treatment by the Fe0/PS. For further oxidation investigation, concentration of 300 ppm Fe0 was selected. There were two common reasons mentioned for this observation, (1) the mechanism of scavenging free radicals by excess ferrous and inhibiting them from reacting with organic molecules.

(2). By feedback mechanism, the more generate of free radicals in reaction solution will produce (S2 O8)2- continuously. [15,16] Effect of Sulfate Dose Per The effect of per sulfate dose on performance of the PS/Fe0 process for folic acid and kanamycin removal was shown in Fig.

(3). At low [PS], Sulfate free radical amounts are not enough to degrade the antibiotics [17]. The DOF's removal efficiency is even enhanced with the increasing molar ratio of PS over pollutant, as much more oxidant molecules are generated at hand to attack the organics. [18] This is consistent with our results. A concentration of 5 mM per sulfate was the optimum value for PS/Fe0 process to achieve an excellent removal efficiency of antibiotics, so we chose this concentration as the optimum dosage for further study optimization.

3.3. Effect of Amount of Per sulfate

Figure 3 shows the effect of per sulfate content on tetracycline, amoxicillin during PS/Fe0 process. Free radical systems at very low PS concentrations are unable to destroy antibiotics as SO4·– free radicals become weak. Moreover, the degradation ratio enhances with the increase of the molar ratio of PS/pollutant since more oxidant molecules are accessible to attack the organic matter. [19] This is consistent with what we observed. The corresponding concentration of 5 mM per sulfate was obtained in which the largest removal efficiency in adsorption was achieved, and it was desired to be applied as an appropriate dose for further PS/Fe0 process optimization investigations.



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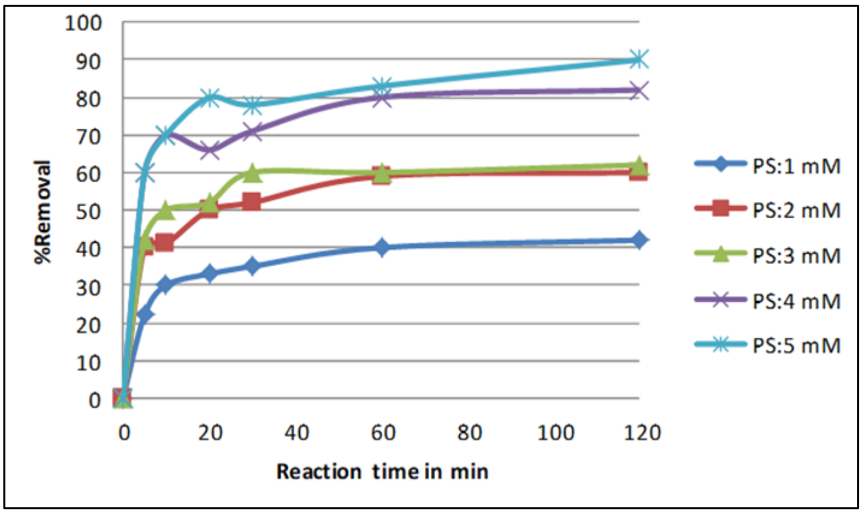

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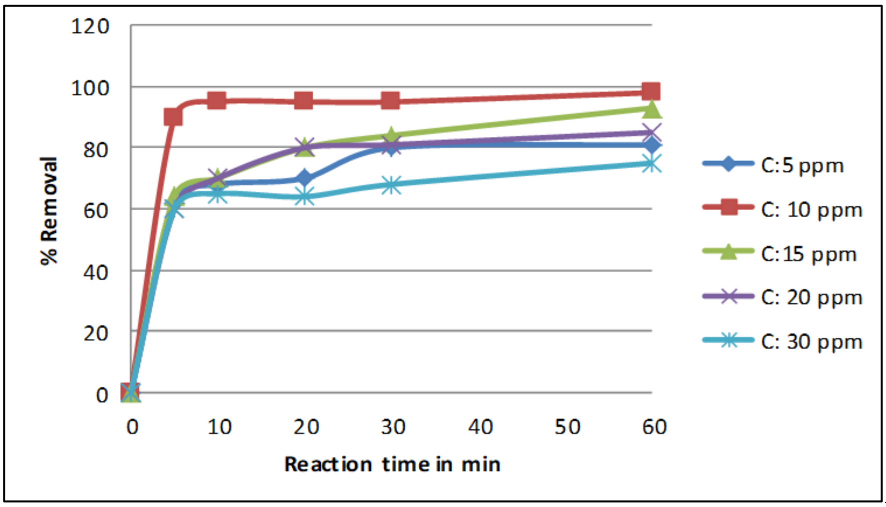
Figure 3 Effect of Different Concentrations of PS on the Performance of the Removal Process of kanamycin (A) and folic acid (B) by PS/Fe^0

3.4. Effect of Initial concentration Antibiotics

Effects of the Starting Concentration Antibiotic Concentrations The effect of the starting concentration (5, 10, 15 and 20.30 mg/L) of kanamycin and folic acid on PS/Fe0 performance is shown in Figure 4. Results indicate that in the optimized conditions, D = 5.23 was achieved using antibiotic concentrations of 30 ppm, 20 ppm, 10 ppm andбC6.C8=0м4ppSingle Group)5 respectively [20][21].



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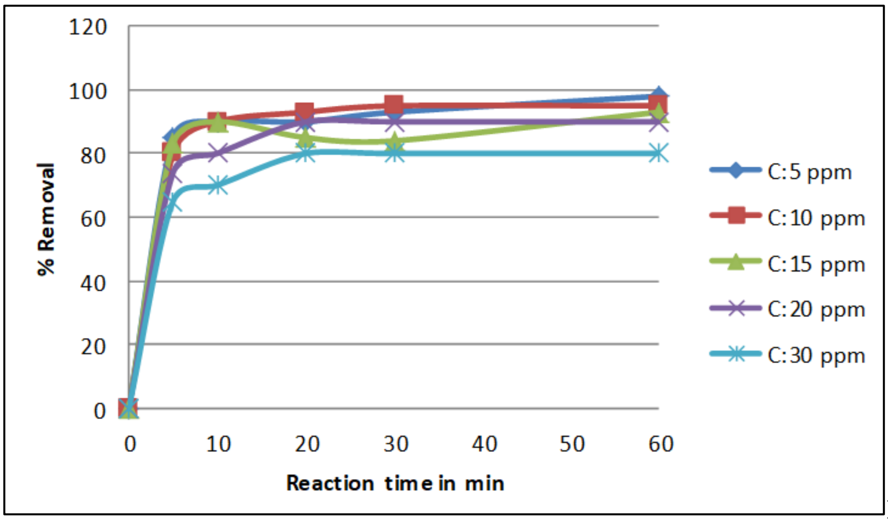

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Figure 4 Effect of Initial Concentrations of kanamycine (A) and folic acid (B) Antibiotics on PS/Fe0

4. Conclusion

The results indicated the pH sensitivity of the method. A greater percentage of the antibiotics was removed when 300 ppm of iron were supplied. As the concentrations of the above antibiotics rose, their ratio became significantly lower. The extraction efficiency was improved with the increase of PS concentration in solution. In addition, the removal of antibiotics from a practical water sample was well done for PS/Fe0 system. The effective PS/Fe0 method of antibiotic removal can also treat waste water with folic acid and kanamycin. Moreover, recycling of the process to some extent is inexpensive until just before the low-cost chemical oxidation treatment.

Compliance with ethical standards

Disclosure of conflict of interest

All authors declare no conflict of interest of regarding the publication of this paper.

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