

## Iron removal efficiency in irrigation water by a zeolite added to sand media filters

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### ABSTRACT

Drip irrigation is characterized by a reduction of water use and energy consumption in agriculture. However, high levels of iron concentration in water have been a problem, due to emitter clogging. This work evaluated the efficiency of zeolite mixed with sand in media filters to remove bivalent iron ( $\text{Fe}^{2+}$ ) from irrigation water. Initially, an experiment was conducted to test mixtures with different combinations of sand-zeolite, as well as contact time between the mixture and the irrigation water, in static conditions. It was possible to observe a reduction of 95% in iron concentration in the water when a mixture of zeolite and sand was used in a proportion of 1:3 in volume. After filters manufactured with polyvinylchloride pipes were built, 10 cm diameter and 55 cm long. They were filled up with a mixture of zeolite and sand in proportion of 1:3 in volume, to evaluate iron removal efficiency by the mixture under dynamic conditions. An iron removal efficiency of 98% was verified. Finally, the bivalent iron solution was passed through the mixture (1:3) and the output concentration didn't reach 50% of the input concentration even after passing 800 pore volumes, equivalent to 800 times the volume of pores inside the column. These results allow us to conclude that the use of Control M.F. 574 zeolite can successfully remove bivalent iron from water at concentrations up to 5 ppm.

*Keywords:* Water treatment; Drip irrigation; Adsorptive filtration

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### 1. Introduction

Drip irrigation is used worldwide to save water and energy on irrigated crops. Despite its success, a frequent problem can be found on these systems related to drip clogging, caused either by poor water quality or wrong specification of filters [1]. Several physical, chemical, and biological factors have been identified as the major clogging causes. Clogging resulting from iron precipitation is especially

difficult to control, and the dissolved form in water can be associated with microbial activity [2].

Iron is one of the most abundant elements in the Earth's crust [3,4], and its compounds in water can be soluble as a result of rock dissolution in aquifers. Soluble forms occur naturally as the ferrous iron ( $\text{Fe(II)}$  or  $\text{Fe(OH)}^+$ ), or forms like ferric iron ( $\text{Fe(OH)}_3$ ) or bacterial forms [4]. Soluble iron would clog emitters only when oxidized, becoming insoluble in water. Even low concentrations such as 0.2 ppm can plug drippers [5] since iron oxidation

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promotes enzymatic proliferation of bacteria resulting in biofilms that interrupt water flow in drippers.

In tropical waters, iron concentration can reach high values and in groundwater it is common to find from 1 to 10 ppm of iron [6]. Use of such waters for drip irrigation can result in drip clogging, mainly due to changing of iron from reduced form ( $\text{Fe}^{2+}$ ) to oxidized form ( $\text{Fe}^{3+}$ ), which is insoluble and interrupts water passage in irrigation emitters [7,8]. Laboratory analysis of soluble iron in water is possible only if the water sample is acidified to pH values smaller than 3.0. The simple transport of water samples subject to shaking movements would cause oxidation transforming bivalent to trivalent iron [6].

To avoid any drip clogging and guarantee proper irrigation, systems must have water treatment units, mainly composed of filters to retain plugging particles. Several techniques can help to treat waters with high soluble iron content: ion exchange, activated carbon and other filtration materials, bioremediation, limestone treatment, as well as oxidation by aeration, chlorination, ozonation followed by filtration, by ashes, by the aerated granular filter and by adsorption [9–17]. Aeration and separation is the most widely method for the removal of iron from groundwater [4]. However, although aeration can precipitate iron, it does require building high-cost settling tanks that must be lined to avoid infiltration losses of water. On the other hand, biological iron removal processes are very sensitive to process conditions (e.g., temperature, filtration rate, oxygen concentration) and do not support large variation in flow and water quality [3]. Therefore, sorption techniques have attracted the attention of irrigation engineers, mainly if they are associated with filtration processes to obtain high-quality water [18,19].

Filter types used at irrigated farms are screen, disk or media filters, being sand the mostly used at irrigated farms. Among filtration systems used in drip irrigation, sand filters with grain sizes ranging from 0.8 to 1.2 mm are very efficient to remove suspended solids, fine sand, silt, algae and other organic materials [20,21]. Among sorption materials, zeolites can be very helpful to remove soluble metals [18–25]. The main mechanism to remove these metals can involve oxidation and/or adsorption. If added to sand in media filters, it is expected that a proper zeolite can decrease soluble iron concentration resulting in insoluble iron which can be removed in backwashing processes. Adding a zeolite to sand at media filters cost two or three times more than sand itself and the volume necessary is low and can last for 5–8 years, according to manufacturers.

The sorption capacity as well as its kinetics can be affected by temperature, zeolite surface area and initial soluble iron concentration. In fact, adsorption of iron by the same zeolite investigated at present research has been previously evaluated by [19] and the adsorption is related to concentration at pores by a linear function.

To predict how long a filter can operate to remove iron before backwashing, it is necessary to investigate the mixing of an input  $\text{Fe}^{2+}$  solution into the filtering media. This process is combined by molecular diffusion, mass transport and dispersion, being all affected by reactions that can slow down solution movement, mainly by adsorption to media particles. Physically, this convective movement

associated with dispersion and diffusion phenomenon is referred to as miscible displacement in porous media [26,27].

By use of miscible displacement techniques and measuring the output concentration, it is possible to graph the relative concentration (output/input) vs. time, or more appropriately, vs. pore volumes. These curves are referred as breakthrough curves (BTC). One pore volume refers to the total porosity inside the media filter.

Combining adsorption characteristics and effluent breakthrough curves, this research investigated the efficiency of different concentrations of a zeolite (Controll M.F. 574®) added in sand media filters to remove iron from irrigation water.

## 2. Methodology

The experiments were carried out at the Hydraulics Laboratory and at the Soil Chemistry Laboratory at Lavras Federal University in Brazil. The commercial zeolite (adsorbent material) analyzed in this study was the Controll M.F. 574® zeolite, which has been characterized as a catalytic media to remove iron, manganese, aluminum and sulfide from natural waters [19]. According to its manufacturer, it does not require regenerating agents and its effects can last up to 8 y. Its grains size varies from 0.35 to 0.85 mm, in dark brown color, with a bulk density of  $1.95 \text{ g cm}^{-3}$ . This zeolite has been detailed described in [19].

The sand grains used in this research presented grains 10% by weight smaller than 0.85 mm, 20% from 0.85 to 1.00 mm, 20% from 1.00 to 1.18 mm and 50% with a diameter varying from 1.18 to 1.70 mm.

### 2.1. Static conditions

The first experiment was carried out in static conditions to find out the best proportion of zeolite:sand to remove  $\text{Fe}^{2+}$ , as well as to determine the best contact time (residence time) to remove iron. Three proportion levels were investigated (zero, 20 and 50% zeolite, volume base) and five contact periods (15, 30, 45, 60 and 120 s).

The mixture of zeolite and sand ( $50 \text{ cm}^3$ ) was set in beakers and manually shaken to gain homogeneity. Then,  $50 \text{ cm}^3$  of 5 ppm iron water containing ammonium ferrous sulfate [ $\text{FeSO}_4(\text{NH}_4)_2\text{SO}_4 \cdot 6\text{H}_2\text{O}$ ] was poured into the beakers. This salt was used to guarantee a source of ferrous iron for the experiment. This concentration was chosen considering that the ferrous form of iron in surface waters are rarely larger than this value. Aliquots were taken after the different contact periods and iron concentration was measured by the colorimetric method [28] in a visible spectrophotometer.

The statistical analysis of this randomized design with three replications was carried using analysis of variance. Then the Scott and Knott test, at 5% probability, was used to compare means.

### 2.2. Dynamic conditions

#### 2.2.1. Media filters

Media filters were built with polyvinylchloride pipes 100 mm diameter and 550 mm long. They were partially

filled (80% volume) with the mixture zeolite and sand at the best proportion revealed by the static experiment. The remaining volume (20%) was left to allow expansion of the mixture during backwashing. The flow rate was set as 500 LH<sup>-1</sup> at a working pressure of 200 kPa. This flow rate resulted in a filtering rate of 63.7 MH<sup>-1</sup>, slightly higher than typical values found in drip irrigation filters (30–50 MH<sup>-1</sup>). The water passing through the filters were, in fact, an ammonium iron sulfate solution with 5 ppm of bivalent iron. At 5 min interval, 100 cm<sup>3</sup> samples were collected from the filter outlet to measure bivalent iron. This procedure was repeated 4 times (four replications).

Iron removal efficiency was calculated as:

$$E = 100 \times \frac{(C_0 - C)}{C_0} \tag{1}$$

where *E* is the iron removal efficiency (%); *C*<sub>0</sub> – input concentration of bivalent iron (ppm); *C* – output concentration of bivalent iron (ppm).

2.2.2. Miscible displacement experiment

This experiment was carried out to find out for how long the zeolite could be effective for the large concentration chosen (5 ppm). In order to obtain the BTC for iron, the stainless steel column, 6.94 cm internal diameter and 8.23 cm high, was filled with a mixture of zeolite (25%) and sand (75%) and the resulting bulk density was 1.46 g cm<sup>-3</sup>. The particle density of zeolite and sand mix was measured using a pycnometer method [29] and its value was 3.046 g cm<sup>-3</sup>, such that the total porosity was 0.5204 cm<sup>3</sup> cm<sup>-3</sup> with a pore volume of 162 cm<sup>3</sup>.

Through the column passed distilled water until reaching permanent flow conditions and then, a 5 ppm Fe<sup>2+</sup> solution was passed, and the effluent concentration measured in 80 cm<sup>3</sup> aliquots. The scheme shown in Fig. 1 represents the experimental apparatus used. The pore velocity measured was 13.59 cm min<sup>-1</sup> (8.15 mh<sup>-1</sup>), equivalent to a filtration rate of 15.6 mh<sup>-1</sup>. Despite the

difference in velocity to the experiment described in 2.2.1, it does not invalidate the purpose of the miscible displacement experiment. Dynamic details of such technique can be seen in [26].

3. Results and discussion

3.1. Static conditions

Regarding results obtained from the static conditions, the remaining iron concentration is presented in Table 1 for different contact periods and mixture rates. Results presented are means of three replications.

Zeolite’s potential to remove bivalent iron is clearly shown when compared to control (0% zeolite), demonstrating the technical viability to remove iron from waters. Considering sand as an inert material, iron removal was not expected to occur at control samples. Since the initial concentration was 5 ppm, the iron removal by control (no zeolite), around 40%, can be assigned to oxidation processes, possibly caused by flow velocity or turbulence during the initial 30 s of contact time or simply by exposing the water to atmosphere where it could have been oxidized.

For 25% and 50% mixture proportion, 15 s of contact were long enough to remove 95% of bivalent iron. It can be noticed that despite the contact time, the iron removal efficiency was approximately 95%.

The mixture proportion is an important factor considering that zeolite costs more than sand. In fact, the greater the zeolite concentration, the better would be the iron removal [30]. However, this was not observed at this experiment, considering that no difference was observed comparing 25%–50% mixture rates. In other words, the larger the zeolite quantity at the mixture didn’t guarantee larger bivalent iron removal.

Therefore, considering economic reasons, this research suggests that 25% zeolite proportion in relation to sand is enough to be used in media filters.

3.2. Dynamic conditions

For this reason, the dynamic experiment was carried out with this 25% proportion and the bivalent iron concentration can be observed in Table 2 for input and output solutions. It can be noticed that the input bivalent iron concentration varied from 4.15 to 4.41 ppm while the output varied from 0.02 to 0.14 ppm, resulting in removal efficiency of 98%.

Table 1  
Bivalent iron concentration (ppm) after contact to a mixture of zeolite and sand for different contact periods

Zeolite proportion (volume based)	Contact time (s)				
	15	30	45	60	120
0% zeolite	4.00 a	3.07 a	3.13 a	3.17 a	3.00 a
25% zeolite	0.17 b	0.20 b	0.00 b	0.03 b	0.20 b
50% zeolite	0.37 b	0.13 b	0.10 b	0.13 b	0.20 b

Means followed by the same letter at columns do not differ statistically by Scott and Knott (*p* < 0.05).

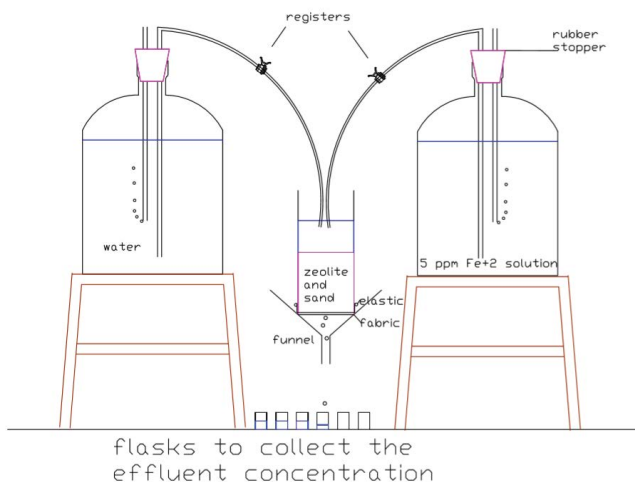


Fig. 1. Miscible displacement apparatus.

Table 2  
Bivalent iron removal efficiency (%) by zeolite and sand mixture at 25% proportion zeolite: sand in media filters

<i>t</i> (min)	Input	Output	<i>E</i> (%)
	Bivalent iron concentration (ppm)		
0	4.15	0.09	97.88
5	4.15	0.02	99.44
10	4.15	0.08	98.10
15	4.22	0.06	98.68
20	4.25	0.08	98.14
25	4.32	0.06	98.71
30	4.38	0.08	98.19
35	4.38	0.06	98.73
40	4.41	0.14	96.75
Mean	4.27	0.07	98.29
Std. dev.	0.11	0.03	0.75
CV (%)	2.48	44.94	0.76

Although the iron stock solution had 5 ppm bivalent iron, the input concentration did not reach this value. Such fact can be attributed to oxidation processes that happened before passing the water through the filters.

Considering 0.3 ppm as a target concentration of bivalent iron in waters for drip irrigation and also for consumption [31], it can be seen that the removal efficiency was high enough to low bivalent concentration to values smaller than this reference. Previous research [19] evaluating iron and manganese removal from natural water, also presented this zeolite as a material with a good adsorption capacity for both metals in natural water, reducing 74% and 66% respectively, generating consumable water with concentrations below the values permitted by the World Health Organization [32].

It should also be pointed out that flow rate can affect iron removal efficiency considering that it decreases the contact time [33]. Researchers [33–35] have noticed that zeolites can be saturated earlier if submitted to high flow rates. We expect it not to be a problem if backwashing in filters can happen more frequently, typically every 2 h or

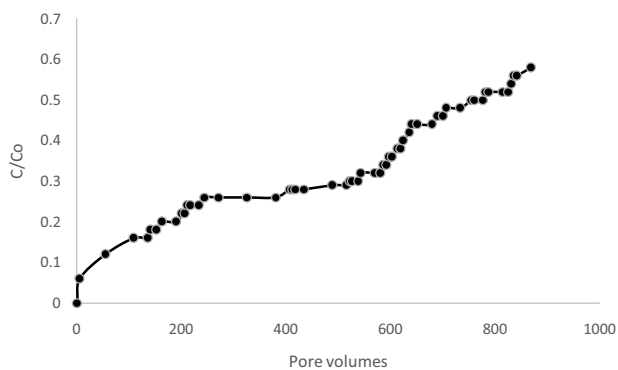


Fig. 2. Bivalent iron breakthrough curve in a mix zeolite (25%) and sand (75%).

less in drip irrigation systems as recommended by filter manufacturers.

### 3.3. Miscible displacement of bivalent iron

Considering the effluent curve obtained at miscible displacement apparatus, shown in Fig. 2, it can be seen that the relative concentration (output/input concentration) didn't reach 1.0 (maximum value) despite passing through the mix of zeolite and sand more than 800 pore volumes that took 8.3 h to pass through the column. Neither for 870 pore volumes, the relative concentration reached 1.0 indicating extremely high interaction of influent with filtering media. Considering that a media filter can be back washed every 2 h (equivalent to 200 pore volumes), the relative concentration wouldn't reach 0.25 meaning that 75% of iron can be easily removed.

## 4. Conclusions

Considering the static conditions, the MF 574 Control MF zeolite can be mixed to sand at 1:3 proportion (25%), volume base, to be technically viable to remove bivalent iron from irrigation waters, reaching removal efficiency as high as 95%.

Bivalent iron movement in a mix of zeolite and sand (1:3) will come out of filters with high retardation and the relative concentration (output/input) didn't reach 50% even after filtering 800 pore volumes.

Adding zeolite to sand media filters is promising for water treatment for drip irrigation, considering that the filtration rates are such that the contact time will be long enough to remove iron up to 98%.

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