

Physico-chemical and biological iron removal from potable water

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Abstract

A pilot-scale trickling filter was constructed and tested for iron removal from potable water. Iron removal was found to be caused by both biological and physico-chemical iron oxidation. The extent of each oxidation type was assessed. The system was inoculated with mixed culture and its performance was tested under continuous operation at 14 °C. Feed iron concentrations and volumetric flow rates (VFR), ranged between 1–4 mg/l and 1000–3000 ml/min (225–680 m³/m²day), respectively. First order kinetics was used to describe the physico-chemical iron oxidation while Monod-type kinetics was used to describe the net biological iron oxidation. An increase of VFR from 1000 to 3000 ml/min reduced the filter's physico-chemical removal efficiency from 93 to 80%, while it remained constant for all the iron feed concentrations (1–4 mg/l) that were tested at each VFR. Bio-oxidation improved filter efficiency by about 5–6%. In all cases there was very good agreement between model predictions and experimental data. A series of experiments were also performed in order to investigate the simultaneous ammonia and iron oxidation. It was found that ammonia influence on iron removal becomes substantial only for high iron and ammonia concentrations. The simplicity of the pilot-scale design, the lack of the need for an external mechanical aeration source and the ease for the design and prediction of the system operation offers a very attractive solution for iron removal from potable water.

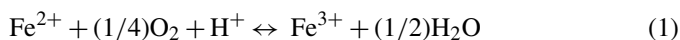
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1. Introduction

Iron is found in surface and ground waters at varying concentration levels, usually up to 3–4 mg/l, but in some cases up to 15 mg/l [1]. When present, even at low concentrations it can be linked to aesthetic and operational problems such as bad taste and color, staining, as well as deposition in the water distribution system leading to incidence of high turbidity [2]. Also, iron promotes the growth of certain types of chlorine-tolerant micro-organisms in water distribution systems, thus causing increased costs for cleaning and sterilizing systems in addition to odor and taste problems. The highest permitted limit of iron concentration for drinking water is 0.2 mg/l [3].

Ferrous iron is oxidized in air according to the following reaction:



The state of iron in water depends above all on the pH and the redox potential. By increasing the pH, dissolved iron (Fe²⁺ or

Fe³⁺) hydrolyzes to form precipitates [4]. The ferrous ion hydrolyzes to produce the array of mononuclear species FeOH⁺ to Fe(OH)₄⁻² between pH 7 and 14. The ferric ion (Fe³⁺) hydrolyzes much more readily than the ferrous ion, beginning at about pH 1. Baes and Mesmer [5] presented diagrams, which show that iron at the range of pH 7–8 is a precipitate.

The rates of ferrous iron oxidation by air increase with pH and about 90% conversion may be achieved in a few minutes at a pH of 7 [6–8]. Tamura et al. [8] suggested that the oxidation product, ferric hydroxide, causes an autocatalytic effect, which becomes negligible at initial ferrous iron concentrations of <3 mg/l. Sung and Morgan [9] studied the kinetics and the products of the reaction of ferrous iron with oxygen in aqueous systems and concluded that for pH greater than ~7, the kinetic data indicate autocatalytic disappearance of aqueous Fe(II).

The rates may be increased by the presence and the action of certain micro-organisms [10–12]. There are several genera of bacteria that oxidize dissolved iron with different mechanisms, including *Gallionella* sp., *Leptothrix ochracea*, *Crenothrix polyspora*, *Leptothrix* sp., *Crenothrix* sp., *Clonothrix* sp., *Sphaerotilus* sp., *Toxothrix thrichogenes* and *Siderocapsaceae* [13]. The experimentally observed growth rates of bacteria have been modeled [14] and bioreactors have been designed

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and constructed with a high efficiency of ferrous iron bio-oxidation [15].

A new approach for the biofilm structure of iron-oxidizing bacteria has been suggested by Karamanev [16]. According to Karamanev's theory, bacterial cells themselves, do not contribute to the mechanical strength of the biofilm. They adsorb on the frame of the biofilm forming a monolayer. Since bacteria cover completely the porous surfaces, their further reproduction results in one daughter cell remaining on the surface, with the other one becoming freely suspended in the liquid medium. Nemati and Webb [17] proved experimentally this theory for the early stages of a bioreactor operation. However, they concluded that the ultimate structure of the biofilm is a combination of growing bacteria and ferric iron precipitates.

Many researchers have studied extensively the biological oxidation of ferrous iron in mine water treatment [14,17–23]. However, in mine water ferrous iron concentration ranged in the order of kg/m^3 (and not of g/m^3 as is the case of potable water), while the solutions' pH was about 2.

Although there are numerous references for ferrous iron removal from mine discharges, and hydrometallurgical processes, there are only limited references concerning ferrous iron removal from groundwater [1,2,9–13,24]. Physico-chemical iron oxidation has been studied extensively and kinetic models have been developed [9] to describe ferrous iron oxidation rate. Combined physico-chemical and biological iron oxidation has also been studied by many researchers [10–13,24] and a first order kinetic model has also been developed [10]. However, only few studies refer to the contribution of biological oxidation on total iron oxidation rate [11–13], while there are no kinetic data available for the bacteria that are responsible for iron removal from drinking water. The main reason for this is the fact that groundwater contains low iron concentrations in comparison with industrial processes, which can be removed by simple aeration followed by precipitation.

Biological iron removal has been postulated to lead to high removal rates [13]. However, no protocol has been developed for biological system start-up. Several reactor designs have been studied in the effort to improve the rate of ferrous iron oxidation, e.g. the rotating biological contactor, the fluidized-bed reactor and the packed-bed reactor with different packing materials [15]. For potable water treatment, pressurized filters and gravity plants are commonly used [13]. The pressurized filters are designed to operate at high rates while gravity plants use fine sand for filtration and only low rates can be achieved. An aeration system is absolutely necessary for these systems.

Trickling filters may be an alternative solution to pressurized filters and gravity plants. The filter media usually consist of either gravel or a variety of plastic packing materials. An adequate flow of air is of fundamental importance for the successful operation of a trickling filter. The principal factors responsible for air flow, is natural aeration, the driving force for airflow being the temperature difference between the ambient air and the air in the void. Moreover, if the size of the filter media is quite small, e.g. a few millimeters, then complete natural aeration and very good filtration may be effected at the same time [11,24]. Jarvis and Younger [25] have used

trickling filters for iron and manganese removal from mine waters.

Iron oxidation, usually is affected by the presence of other elements and especially by the presence of ammonia and manganese. Gouzinis et al. [24] studied the interactions between ammonia, iron and manganese when present simultaneously in a biological filter and the influences they have on filter performance.

The aim of the present paper is: (a) to investigate the contribution of each type of iron oxidation (physico-chemical and biological) on the performance of a pilot-scale trickling filter for iron removal from potable water, (b) to develop and validate a mathematical model able to predict the discrete contribution of each type of iron oxidation in a trickling filter, and (c) to study the simultaneous ammonia and iron removal.

2. Materials and methods

The pilot-scale trickling filter (Fig. 1) consisted of a Plexiglas tube, 160 cm high and 9 cm internal diameter. This pilot-filter height is typical of a full-scale industrial filter. Since it is the loadings (hydraulic and iron) per unit cross-sectional area that matter, no scale-up study is necessary. The support material was silicic gravel with a mean diameter of 3.9 mm, and specific surface area (A_S) of $13.85 \text{ cm}^2/\text{cm}^3$, while the depth of the support media was 143 cm and the filter porosity 0.38. The silicic gravel originated from a quarry near the city of Larissa, Greece. The filter media were loaded to the filter and were washed with water from the supply network for 48 h. The support material was not flooded for flow rates up to 3000 ml/min or hydraulic loadings up to $680 \text{ m}^3/\text{m}^2\text{day}$. All flow rates used in this work were within this limit. At the top of the filter, there was a fixed nozzle, which distributed the incoming water evenly to the whole filter surface. An underdrain system collected the treated water and any biological solids that would detach from the media. Along the filter depth there were 10 sampling ports for iron concentration measurements in the bulk liquid. Thus, it was possible to have an experimental assessment of the ferrous iron concentration profiles along the filter depth. The pH and dissolved oxygen measurements were made using the Hanna pH211 pH meter,

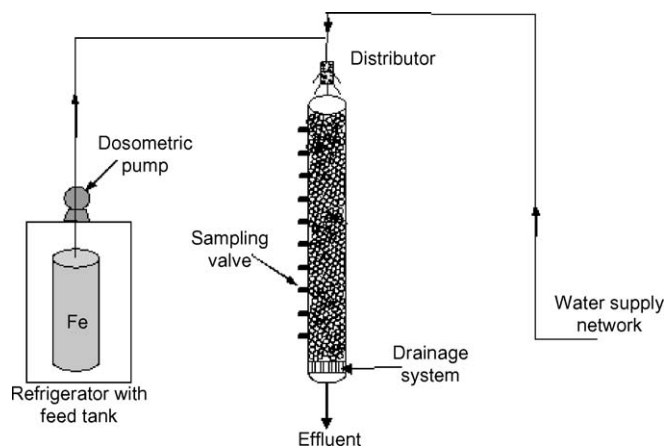


Fig. 1. Schematic drawing of the pilot-scale trickling filters arrangement.

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