Desalination and Water Treatment
www.deswater.com
doi:10.5004/dwt.2018.22843

125 (2018) 290–295
September

Methods and affecting factors of improving efficiency of biological removal of iron and manganese

Wen-Jing Ou Yang[a], Lei Wang[b,*], Hai-Feng Bao[a], Chu-Hong Lin[b], Yang-Lun Cai[b], Kai Ren[a]

[a]*College of Resources and Environment Engineering, Jilin University of Chemical Technology, Jilin City, 132022, China, Tel. 13804420505, email: oywj_jlict@163.com (W.-J.O. Yang), 81937936@qq.com (H.-F. Bao), 532465106@qq.com (K. Ren)*
[b]*Beijing Institute of Technology, Zhuhai, Zhuhai, 519000, China, email: wanglei_zhbit@163.com (L. Wang), 1204097969@qq.com (C.-H. Lin), 453094321@qq.com (Y.-L. Cai)*

Received 11 April 2018; Accepted19 July 2018

ABSTRACT

A pilot-scale trickling filter with lower layer support material was constructed. The dominant microorganisms of oxidize iron and manganese were immobilized on the surface of manganese sand with the improved circulation-stay type immobilization methods. The quantity of bacterium fluid was identified according to the solid-liquid ratio which was slightly greater than the porosity of support material. The performance of the trickling filter was tested for simultaneous biological removal of iron and manganese from groundwater. The results showed that mature time of bio-filter layer was 30 d. The adaptability to fluctuation of iron and manganese concentration was very strong and the biggest filtration rate could be increased to 12 m/h. The filter layer thickness could be reduced to 70 cm. The Fe^{2+} concentration declined to 0 mg/L at the filter layer depth of 10 cm. However, the Mn^{2+} concentration declined to less than 0.05 mg/L at the filter layer depth of 55 cm. The most efficient area of removing manganese was in the filter layer depth of 25~40 cm. As the concentration of Fe^{2+} was more than 1.5 mg/L, there was the phenomenon of manganese dissolution because of the chemical reaction between Fe^{2+} and manganese sand (MnO_2). In this research the actual consumption value of DO was less than the theoretical value. Strong aeration was not needed in the process of biological removal. In the progress of removing iron and manganese, ORP increased gradually. As ORP increased to 349 mV, iron was removed completely; while ORP increased to 422 mV, manganese was removed totally. Comparing the removal results of iron and manganese before and after inoculation, it was found that the iron and manganese removal of the filter layer mainly lied in bio-catalytic oxidation.

Keywords: Biological process; Iron and manganese removal; Efficiency; Method; Factor

1. Introduction

The high concentration of iron and manganese in the groundwater often results in serious chromaticity and iron taste, which has brought about much inconvenience to people's daily lives and even does harm to human health. The removal of iron and manganese from groundwater is a practical problem which must be solved. Therefore, the technology of removing iron and manganese from groundwater has been a hotspot for many experts and scholars. At present, various methods are used to remove iron and manganese from groundwater, such as chemical contact oxidation method, UF in conjunction with prechlorination, adsorption and filtration of Fe^{3+} impregnated or biological activated carbon, trickling filtration of biological quartz sand filter layer and so on [1–4]. The biological removal technology has been widely recognized and put into use with its simple process, no chemical agent, low energy consumption and high efficiency [5–8]. However, there are some disadvantages in the current process, such as

*Corresponding author.

Presented at the 4th Annual Science and Technology Conference (Tesseract'17) at the School of Petroleum Technology, Pandit Deen Dayal Petroleum University, 10–12 November 2017, Gandhinagar, India

1944-3994 / 1944-3986

inconvenient and inefficient operating method of biofilm colonization, long mature time of bio-filter layer and large equipment. And it is believed that there is still possibility to increase the efficiency [9,10].

The objective of this study was to explore a simple method of biofilm colonization and the appropriate quantity of bacterial suspension. This research also attempted to reduce ripening time and improve the effectiveness of bio-filter. The removal and variation rules were summarized during the process at the same time, for the purpose of providing technical service and theoretical guidance for engineering practice, and promoting the extensive application of the biological technology for removing iron and manganese.

2. Materials and methods

2.1. Testing device

The pilot-scale trickling filter (Fig. 1) consisted of a Plexiglastube, 120 cm high and 10 cm internal diameter. The filter material was manganese mineral from Mashan, Guangxi Province and its particle diameter was 0.6~1.6 mm. The filter layer thickness was 70 cm. At the bottom of the filter, there was cushion of cobble and its thickness was 15 cm. The particle diameter of cobble was 0.6~1.2 cm. The method of aeration was falling water aeration and the falling water height was 40 cm. Along the filter depth there were 5 sampling ports numbered as 1#, 2#, 3#, 4#, 5#, for measuring iron and manganese concentration at different filter depth in the bulk liquid. The interval between two sampling ports was 15 cm. It was therefore possible to have an experimental assessment of the iron and manganese concentration profiles along the filter depth. The distance from No. 1 sampling port to the top of manganese sand was 10 cm.

2.2. Raw water quality

The raw water is groundwater from a deep well in the suburb of Jilin city. The feeding concentrations ranged from 4.5 to 8.0 mg/L and 1.5 to 3.0 mg/L for ferrous iron and manganese, respectively. The temperature was 10~14° and pH was 6.50~6.80. Before and after aeration, the concentration of dissolved oxygen in water was 0.8~1.0 mg/L and 6.5~7.4 mg/L, respectively.

2.3. Biofilm immobilization

The microorganisms used in the test were separated from active membrane on the mature manganese sand. The bacterial suspension with high concentration and activity was made by enlarging cultivation in the laboratory and was immobilized on the surface of manganese sand in a cir-

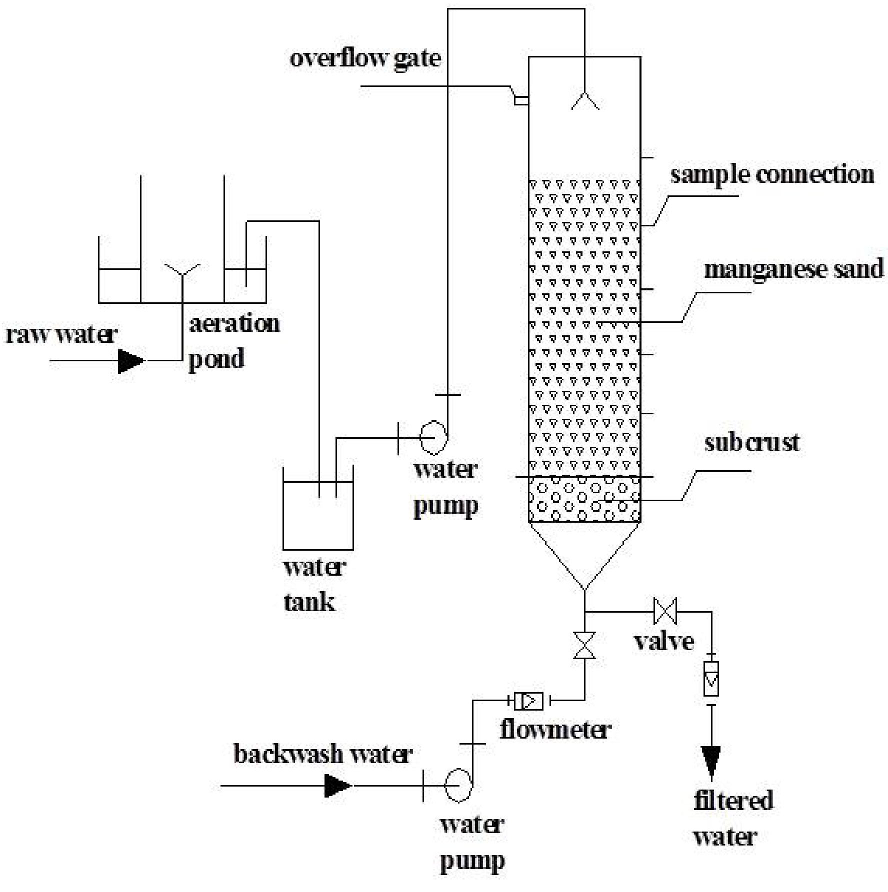

Fig. 1. Schematic drawing of the pilot-scale trickling filter.

culating and staying way. The quantity of bacterial suspension was determined by the liquid-solid ratio (1:3) which was slightly greater than the porines of filter material. The bacterial suspension was circulated 2 times per day and the residence time was 12 h. The groundwater was poured into the filter after 7 d because strains grew and multiplied fastest during this period.

2.4. Trial progress

To control cultivating and operating parameters, the groundwater was initially poured into the filter in the filtration rate of 1 m/h, and then the rate was gradually improved in a smaller range. Before the biological filter layer was mature, the filtration rate was improved no more than 3 m/h and the intensity of backwashing was 10 L/(m^2·s). The time of backwashing was 2 min and the cycle of operation was 5 d. After the biological filter layer was mature, the intensity of backwashing was 12 L/(m^2·s), the time was 3 min and the cycle of operation was 3~4 d. And then the concentration of iron and manganese in water were measured every 24 h in different operating conditions of 6, 8, 10, 12 m/h, for the purpose of inspecting the factors that affected the filtration performance such as iron and manganese content, filter velocity, filtration bed thickness, dissolved oxygen (DO) and oxidation-reduction potential (ORP). The various indexes were measured in accordance with the national standards.

3. Results and discussion

3.1. Startup and running of biofilter

To improve the aging characteristics of the biofilter, the key was to shorten the mature time of biofilter layer. After raw water injection, the color of filter layer was different in the depth of 10 cm. The upper part of the filter material gradually became yellow, while the lower part turned into black. This was because the filter removed iron first and then manganese. Through the bacteria cultivation count, the number of bacteria which were in the filter material surface increased exponentially. The iron and manganese concentration in filtered water was trace and stable after inoculation for 30 d. At this time the biofilter layer was mature. Compared with the mature time of 60~90 d reported in [11], the mature time of biofilter layer was shortened and time-effectiveness was improved signally, after adopting the above mentioned immobilization method, suitable quantity of bacterial suspension and appropriate culturing parameters.

3.2. The influence of iron and manganese content on removal effect

In the conditions of raw water pH 6.68~6.78, temperature 12~14° and filtration rate 6 m/h, the influence of iron and manganese content on removing effect was examined. The results were shown in Figs. 2 and 3.

As shown in Figs. 2 and 3, when Fe^{2+} concentration in the raw water changed in the range of 4.5~8.0 mg/L, the content of iron in filtered water was nearly 0 mg/L and the removal rate was close to 100%. When Mn^{2+}concentration in raw water changed in the range of 1.5~3.0 mg/L, the content of manganese in filtered water was all less than 0.05 mg/L and the removal rate was 96–100%. If the concentration of iron and manganese in raw water changed within a certain range, there was almost no influence on the biological removal of iron and manganese. The quality of filtered water was better than the quality index regulated in Sanitary Standard for Drinking Water (GB5749-2006).

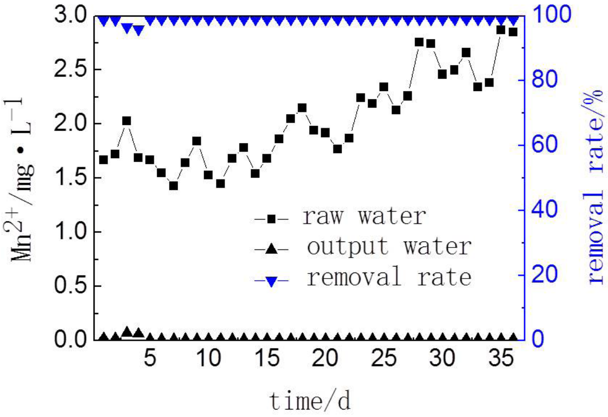

Fig. 2. Removal of iron in the condition ofdifferent iron content.

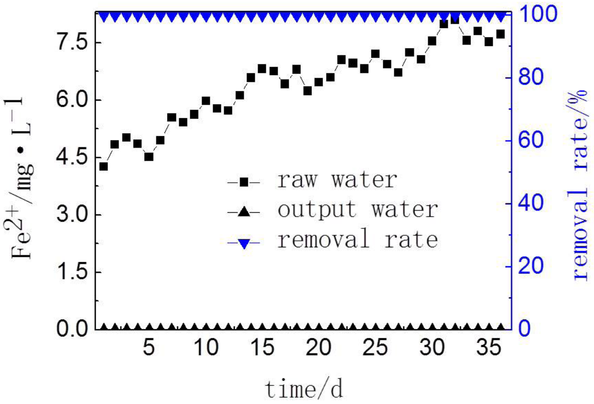

Fig. 3. Removal of manganese in the condition of different manganese content.

3.3. The influence of Fe^{2+} concentration on manganese removal

The content of manganese in output water was higher than that of inflow at the filter depth of 10 cm. It might be the chemical reaction between Fe^{2+} and manganese sand (MnO_2) which caused the phenomenon of manganese dissolution. The chemical reaction might be related to the concentration of Fe^{2+}. The concentration of manganese in output water was measured in the conditions of different Fe^{2+} concentration of 0, 0.5, 1.0, 1.5, 2.0 mg/L in inflow water. Other conditions, like the value of pH and DO, were as uniform as possible. The results were shown in Table 1.

As shown in Table 1, with high concentration of Fe^{2+} (more than 1.5 mg/L), the content of manganese in output water was higher than that of inflow at the filter depth

Table1
The removal of manganese at different filter depth in the conditions of different Fe^{2+} concentration

Fe^{2+}	Mn^{2+} (mg/L)					
(mg/L)	0 cm	10 cm	25 cm	40 cm	55 cm	70 cm
0	2.40	2.34	1.98	0.33	0	0
0.5	2.80	2.72	2.52	1.03	0.06	0
1.0	2.56	2.46	1.93	0.27	0	0
1.5	2.24	2.54	1.89	0.29	0	0
2.0	2.22	2.98	1.68	0.81	0	0

of 10 cm and the phenomenon of manganese dissolution occurred. As the concentration of Fe^{2+} was less than 1.0 mg/L, no manganese was dissolved. It was concluded that the manganese dissolution was related to the concentration of Fe^{2+}. As the concentration of Fe^{2+} was low, Fe^{2+} was oxidized rapidly by dissolved oxygen with bio-analytical oxidation and there was no time for the reaction between Fe^{2+} and MnO_2. Therefore, the phenomenon of manganese dissolution would not appear. Only with high concentration of Fe^{2+}, some Fe^{2+} was oxidized and the other Fe^{2+} reacted with MnO_2. Such the reaction between Fe^{2+} and MnO_2 was the reason for the phenomenon of manganese.

3.3. The influence of filtering velocity on removal effect

In the conditions of raw water pH 6.65~6.80, temperature 12~14°, Fe^{2+} 5.86 mg/L, Mn^{2+} 1.57~2.99 mg/L, the content of iron and manganese in output water was measured at different filter velocity of 6, 8, 10, 12 m/h . The results were shown in Figs. 4 and 5.

As shown in Figs. 4 and 5, the filtration rate which was in the range of 6~12 m/h had almost no influence on iron and manganese removal. The quality of filtered water was up to Sanitary Standard for Drinking Water. With the increase of filtration rate, the filter layer thickness for entirely removing manganese increased rapidly. As a result, the manganese removal was the main factor to limit filtration rate. In addition, during the operation it was found that the high filtration rate could lead to excessive growth of head loss and increase of backwashing frequency, which would bring negative impact on the removal stability of iron and manganese. In consideration of above mentioned reasons it was proposed that the best filtration rate was in the range of 6~8 m/h. For water with higher content of iron and manganese, to achieve high filtration rate it was suggested that a two-stage filtration system was designed to avoid the excessive growth of head loss and the increase of backwashing frequency.

3.5. The influence of filter layer thickness on removal effect

In the conditions of raw water pH 6.72, temperature 12°, Fe^{2+} 3.86 mg/L, Mn^{2+} 2.43 mg/L and filtration rate 6 m/h, iron and manganese concentration in filtered water at different filter depth was measured, respectively. The results are shown in Fig. 6.

As shown in Fig. 6, Fe^{2+} content of output water reduced to 0 mg/L at the filter depth of 10 cm. In the process, Fe^{2+}

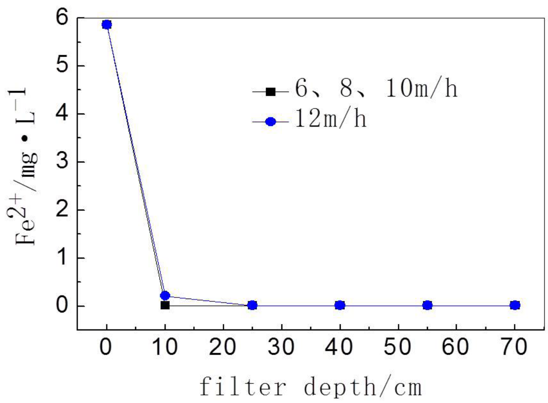

Fig. 4. Removal of iron in the condition of different filter velocity.

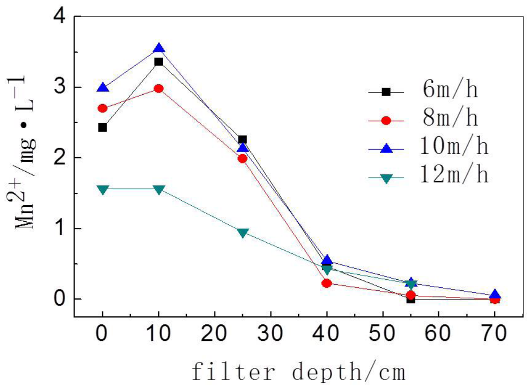

Fig. 5. Removal of manganese in the condition of different filter velocity.

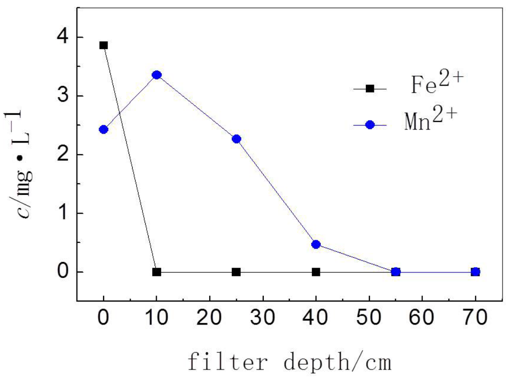

Fig. 6. Removal of iron and manganese at different filter depth.

was biological catalytic oxidized and physical chemical oxidized and converted to $Fe(OH)_3$, which was trapped by the filter layer. The content of manganese in effluent was higher than that of inflow at the filter depth of 10 cm. The content of manganese in filtered water decreased rapidly below the filter depth of 10 cm and the position of the highest removal velocity was in the filter depth of 25~40 cm. The content of manganese in filtered water was trace at the filter depth of 55 cm. Mn^{2+} was biological catalytic oxidized and turned into MnO_2, which was trapped by the filter layer and adopted by

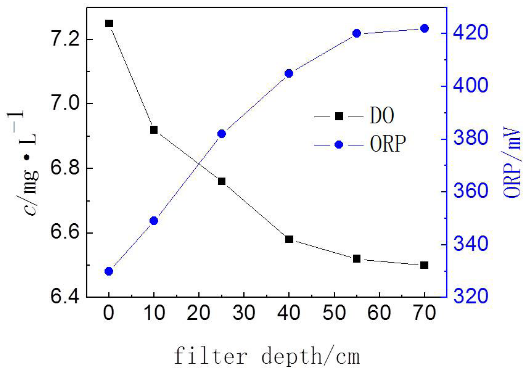

Fig. 7. Variation of Fe^{2+} and Mn^{2+} content in output water.

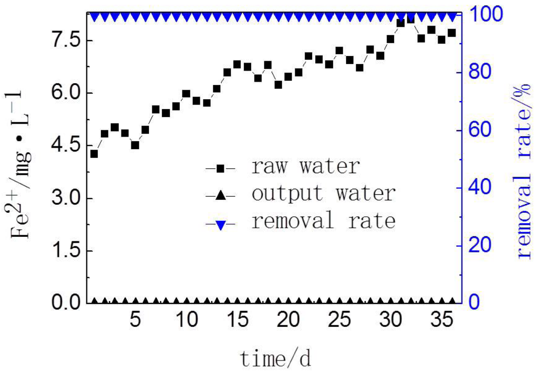

Fig. 8. Variation of DO and ORP content in output water.

the bacteria. Compared to the filter thickness of 120~150 cm reported in [12 and [13], the lower filter thickness was needed when removing the same amount of iron and manganese. As a result, the smaller equipment size, and lower investment and energy consumption were followed which was of great significance to the engineering design.

3.6. The changes of DO and ORP in the process of iron and manganese removal

In the conditions of raw water pH 6.70, temperature12°, Fe^{2+} 3.02 mg/L, Mn^{2+} 2.22 mg/L and filtration rate 6 m/h, the content of iron and manganese, DO and ORP of output water were measured at different filter depth, respectively. The results are given in Figs. 7 and 8.

The biological removal of iron and manganese was the process that enzymes of the cell surface catalyzed-oxidized low-valent and reductive iron-manganese. No matter how complex the process was, no matter how energy and electron were transferred, the final electron acceptor was the dissolved oxygen in water. The theoretical value of oxygen consumption could be calculated according to the theory of electron gain-loss conservation. It met the following equation.

$$c(O_2) = 0.143\ c(Fe^{2+}) + 0.29\ c(Mn^{2+})$$

When the content of Fe^{2+} and Mn^{2+} in raw water was 3.02 mg/L and 2.22 mg/L, respectively, the theoretical value of oxygen consumption that oxidized iron-manganese completely was 1.08 mg/L according to the equation. From Figs. 7 and 8, the actual value of dissolved oxygen consumption was 0.75 mg/L. That was to say that the actual value was less than the theoretical value in the progress of biological removal of iron and manganese. It might be the reason that some of Mn^{2+} was adsorbed on the bacterial surface and formed dynamic process of adsorption and desorption. The amount of adsorption was greater than that of desorption, so the actual oxygen consumption was less than the theoretical value. As shown in Fig. 8, when Fe^{2+} and Mn^{2+} were oxidized, the value of $[Fe^{3+}]/[Fe^{2+}]$ and $[Mn^{4+}]/[Mn^{2+}]$ rose gradually. Therefore, ORP increased gradually in the progress of removing iron and manganese. When ORP increased from 278 mV to 349 mV, Fe^{2+} was removed; when ORP increased from 349 mV to 422 mV, Mn^{2+} was removed.

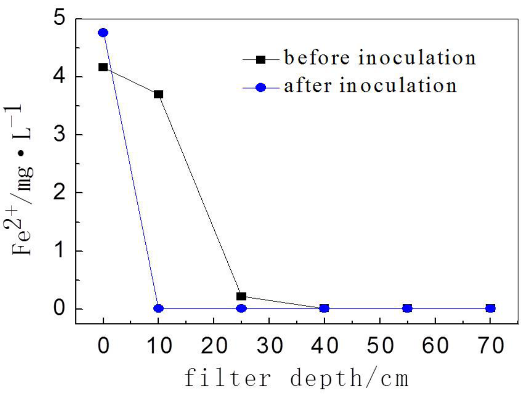

Fig. 9. Removal of iron before and after bacteria inoculation.

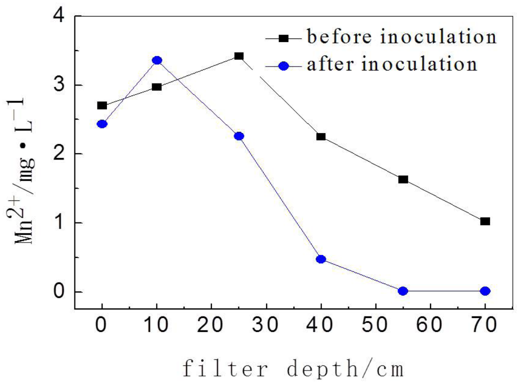

Fig. 10. Removal of manganese before and after bacteria inoculation.

As a result, the removal of iron and manganese could be predicted based on the change of ORP.

The removal effects of iron and manganese before and after inoculation.

In order to determine whether the bio-catalytically oxidation played a major role in the removal of iron and manganese, the removal effect of iron and manganese before and after inoculation and mature was studied. Other conditions were as uniform as possible. The results are shown in Figs. 9 and 10.

As shown in Fig. 9, after inoculation and mature the iron content in filtered water was decreased from 4.76 mg/L to trace at the filter depth of 10 cm and the removal efficiency for iron was nearly 100%. But before inoculation the removal of iron was only 11% at the same depth, and Fe^{2+} was still detected at the filter depth of 25 cm. From Fig. 10, the content of manganese in filtered water was decreased from 2.43 mg/L to trace at the filter depth of 55 cm after inoculation, and the removal efficiency could be achieved 100%. Before inoculation, the content of manganese still remained 1.63 mg/L at the filter depth of 55 cm and the removal efficiency was only 39.6%. By comparison it was found that the effect of removing iron and manganese after inoculation was better than before. The results showed that oxidation of iron and manganese was mainly bio-catalytic oxidation.

4. Conclusions

(1) With improved bio-film method, suitable amount of bacteria, and optimized culturing and operating parameters, the mature time of bio-filter layer was reduced to 30 d. The filter layer thickness could be reduced to 70 cm with strong adaptability to high iron and manganese concentration and high filtration rate.

(2) The removal rules of iron and manganese were identified. Along the filter layer depth, iron was removed between 0~10 cm, while manganese was removed between 10~55 cm. The most efficient area of removing manganese was in the filter depth of 25~40 cm. When Fe^{2+} concentration was more than 1.5 mg/L in inflow, there was oxidation-reduction reaction between iron and manganese which led to the phenomenon of manganese dissolution.

(3) In this research the actual consumption value of DO was less than the theoretical value. Strong aeration was not needed in the progress of biological removal of iron and manganese from groundwater. During the process of removing iron and manganese, ORP increased continuously. As ORP increased to 349 mV, iron was removed completely, when ORP increased to 422 mV, manganese was removed totally.

(4) By comparing removal effects before and after inoculation, it was found that the iron and manganese removal of the filter layer mainly lied in bio-catalytic oxidation.

References

[1] K.H. Choo, H. Lee, S.J. Choi, Iron and manganese removal and membrane fouling during UF in conjunction with prechlorination for drinking water treatment, J. Membr. Sci., 267 (2005) 18–26.

[2] P. Mondal, C.B. Majumder, B. Mohanty, Effects of adsorbent dose, its particle size and initial arsenic concentration on the removal of arsenic, iron and manganese from simulated ground water by Fe^{3+} impregnated activated carbon, J. Hazard. Mater., 150 (2008) 695–702.

[3] A.B. Jusoh, W.H. Cheng, W.M. Low, A. Nora'Aini, M.J.M.M. Noor, Study on the removal of iron and manganese in groundwater by granular activated carbon, Desalination, 182 (2005) 347–353.

[4] A.G. Tekerlekopoulou, D.V. Vayenas, Simultaneous biological removal of ammonia, iron and manganese from potable water using a trickling filter, Biochem. Eng. J., 39 (2008) 215–220.

[5] V.A. Pacini, I.A. María, G. Sanguinetti, Removal of iron and manganese using biological roughing up flow filtration technology, Water Res., 39 (2005) 4463–4475.

[6] L.I. Dong, J. Zhang, L.X. Chen, H. Yang, M.C. Gao, Application of biological removal of iron and manganese from groundwater treatment plant, China Water Wastewater, 20 (2004) 85–88.

[7] T.H. Stephens, Microbial manganese oxidation in the Lower Mississippi River: methods and evidence, Geomicrobiol. J., 22 (2005) 117–125.

[8] J.M. Cerrato, A.M. Dietrich, W.R. Knocke, C.W. Mckinney, A. Pruden, Manganese-oxidizing and -reducing microorganisms isolated from biofilms in chlorinated drinking water systems, Water Res., 44 (2010) 3935–3945.

[9] H. Yang, D. Li, J. Zhang, R. Hao, B. Li, Design of biological filter for iron and manganese removal from water, J. Environ. Sci. Health Part A Toxic/Hazard.Subst. Environ. Eng., 39 (2004) 1447–1454.

[10] X.U. Jing-Cheng, L.F. Huang, X.F. Huang, J.W. Fan, J. Zhang, Parameters of manganese sand biological filtration process in recycling of wastewater from steel and iron production, Industrial Water & Wastewater, 2008.

[11] S.Y. Qin, F. Ma, P. Huang, J.X. Yang, Fe (II) and Mn (II) removal from drilled well water: A case study from a biological treatment unit in Harbin, Desalination, 245 (2009) 183–193.

[12] L.I. Dong, J. Zhang, Y.P. Zhang, H.T. Wang, Study on optimization of biological filter bed thickness for removal of iron and manganese, China Water Wastewater, 23 (2007) 94–97.

[13] A.G. Tekerlekopoulou, I.A. Vasiliadou, D.V. Vayenas, Biological manganese removal from potable water using trickling filters, Biochem. Eng. J., 38 (2008) 292–301.