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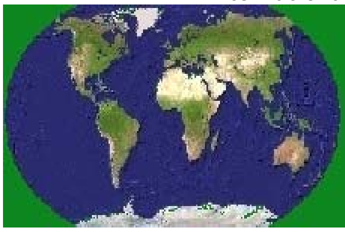

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Removal of Manganese by Microfiltration in a Water Treatment Plant

Sachin Madhavrao Kanawade

Abstract
Bench experiments and pilot studies were conducted to investigate the alternatives for removal of manganese when microfiltration was used instead of media filtration. Comparative studies were conducted with a variety of oxidants, including: chlorine, chlorine dioxide, permanganate and hydrogen peroxide. Bench-scale jar testing indicated that chlorine dioxide was the most effective oxidant, resulting in adequate removal of the manganese with less than twenty minutes of reaction time. Pilot-scale microfiltration experiments indicated that 99% of the manganese was removed when 0.5 mg/L of chlorine dioxide was dosed using a twenty-minute reaction time. As the result of these studies, a full-scale microfiltration plant expansion (10-MGD) is being designed to replace the conventional water treatment in Monroe. This plant will be first application of microfiltration membranes for municipal water treatment.

Keywords: Manganese, microfiltration, water treatment plant, removal.

1. Introduction
The existing water treatment plant is a 6.4 million gallons per day (MGD) (24,000 m^3/day) conventional surface water treatment plant with sand filters that are currently rated at a capacity of 2 gallons per minute per square feet of media (gpm/sq. ft.) (4.9 meters/hour). The majority of the existing filters have been in service for over 30 years without major modification or replacement of media. In order to continue to provide quality drinking water to Monroe and to meet the Interim Enhanced Surface Water Treatment Rule (IESWTR), the existing filters will require some rehabilitation and improvements. Due to the increased demands for safe drinking water, a possible alternative to upgrading the filters in the existing surface water plant is to install a membrane process
The primary objective of the membrane process is to provide a reliable barrier for removal of microbial pathogens. The removal of Giardia and Cryptosporidium by Microfiltration in challenge studies with viable organisms has been well-documented elsewhere [1,2]. Turbidity in the Alcovy River varies seasonally from to 8 to 200 NTUs. A secondary objective is to provide for the continued removal of manganese. In the southeastern United States, manganese is a common element in the soil and solubilizes under reducing conditions in the ambient water supplies. Removal of manganese in conventional water treatment plants often occurs without planning as the media filters often function as "naturally occurring green sand filters" by processes that have become more familiar[3]. If a microfilter membrane is installed to replace the sand filter then manganese removal by oxide coated media (i.e., green sand effect) will no longer occur.
Designers and operators need to be aware that another manganese removal mechanism needs to be in place after the filters are no longer in the process. Removal of manganese from surface water has been previously explored by Jimbo *et al.* [4] They determined that in excess of 90% of dissolved manganese can be removed using pre-oxidation and membrane filtration. Manganese has been documented as a membrane foulant[5]. In light of these studies, it is imperative to ensure that no permanent fouling of the microfiltration membranes is evident. Careful analysis of membrane operating data is required to ensure that membrane treatment is appropriate for the application.

1.1 Chemistry of Manganese Removal
The manganese is a common element in the soil where it exists primarily as manganese dioxide, which is very insoluble in water. Under anaerobic conditions manganese is reduced from an oxidation state of IV to II and becomes soluble[6]. Very similar reactions can occur

with iron at the same time. If manganese can be re-oxidized to an oxidation state of IV, the manganese will become insoluble again. While manganese can be oxidized by chlorine, this reaction may proceed slowly and may not occur until after filtration or in the distribution system. Manganese can cause staining of clothes and plumbing fixtures, and incrustation of water mains, which can result in black water and debris at the customer's tap[7]. Manganese is more difficult to oxidize than iron[8], hence if the treatment process oxidizes and removes manganese; it also oxidizes and removes iron. The raw water manganese in Monroe was in excess of the Environmental Protection Agency's recommended level of 0.05 mg/L, and is expected to increase with increased reliance on an impounded water reservoir in the summer time due to thermal stratification of the reservoir and organic loads in the ambient water.

1.2 Manganese Removal Utilizing Adsorption and Oxidation

Removal of manganese by adsorption to a commercially available zeolite mineral known as greensand was developed in the last decades. It is now apparent that removal of manganese in conventional water treatment plants by adsorption followed by oxidation can occur without planning as the granular media filters function as "naturally occurring green sand filters." Knocke, Occiano and Hungate [3] quantified the presence of the manganese in the coating by extracting the coating from samples of filter media from thirteen water treatment plants in Virginia and North Carolina. They used a 0.5 % nitric acid solution, a strong reducing agent (hydroxylamine sulfate) and a reaction time of two hours.

The amount of manganese extracted ranged from nil to 60 milligrams per gram of media (mg/gm) (6%). By comparison, an extraction of virgin greensand by the same method yielded 4.3 mg/gm (0.4 %). A similar extraction was conducted on a sample of media from a water treatment plant in South Carolina[9] **(Figure 1).** Nitric acid was used, but sodium thiosulfate was substituted as the reducing agent. Before the extraction, sand was brown and after the extraction, the sand was restored to the original off-white color **(Figure 2).** The black objects in the photographs are pieces of anthracite, which is the second component in dual media filters. Knocke *et al*.[3] determined that the capacity of the media to remove manganese without the presence of chlorine was a function of the amount of manganese in the coating and the pH. If chlorine residual is present, the rate constants for adsorption of manganese increased. In addition, the presence of chlorine extended the capacity of the media for adsorption of manganese without showing any indications of exhaustion of the media capacity.

Figure 1 - Filter Media before Extraction of Manganese

Figure 2 - Filter Media after Extraction of Manganese

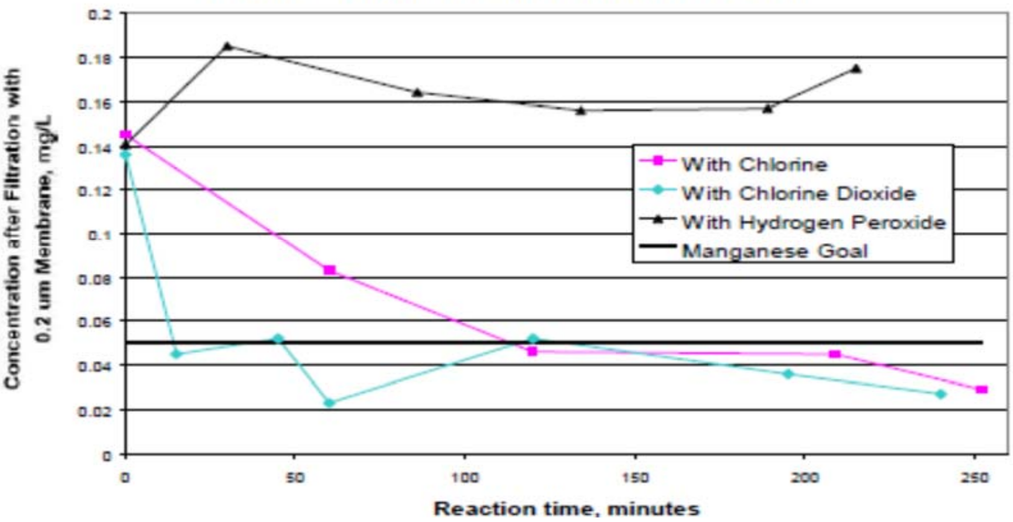
1.3 Study Objectives

The existing water treatment system successfully removes the manganese present to below this concentration due to the manganese dioxide coating on the filtration media. If microfiltration is installed to replace the granular media filters then manganese removal by oxide coated media (i.e., green sand effect) will no longer occur. In Monroe, manganese oxidation prior to microfiltration is needed since the granular media filters will no longer be in the process. In order to evaluate the efficacy of various oxidants for manganese removal using microfiltration processes, a series of batch tests was conducted. Once the most efficient oxidants were determined, a membrane pilot study was performed to obtain design data for the water treatment plant expansion.

2 Experimental Methodology

In the batch tests, the time required for removal of manganese by pre-oxidation and microfiltration was compared for a variety of oxidants, including chlorine, chlorine dioxide, permanganate and hydrogen peroxide **(Figure 3).** These experiments indicated that chlorine dioxide was the preferred oxidant for the oxidation of manganese.

Figure 3 - Manganese Removal in Batch Tests

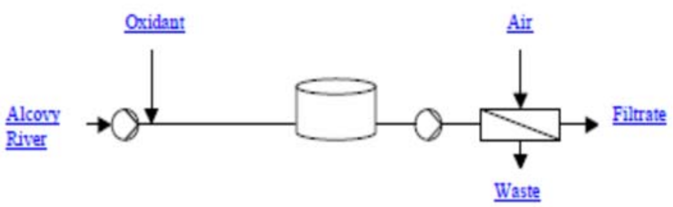


3 Membrane Pilot Study

A membrane pilot study was initiated to determine design parameters for the plant expansion. Raw water from the water treatment plant intake was pumped from the chemical treatment building to the membrane pilot unit. A chemical dosing point was located immediately downstream of the pump. The estimated length of pipe from the chemical treatment building to the membrane unit was four hundred feet. A 500-gallon polyethylene tank was installed prior to the membrane system inlet to provide additional contact time for the oxidation reaction to be completed. The tank was configured with an overflow to permit constant flow from the chemical treatment building.

Backwashes were performed every 22 minutes using compressed air and feed water to remove particulate matter from the membrane surface. During normal operation of the unit, some membrane fouling was observed that was not mitigated by the backwash sequence. Occasional clean in place procedures were implemented to remove foulants from the membrane surface, restoring the system transmembrane pressure to the state before fouling. Citric acid and sodium hypochlorite were used as the clean-in-place chemicals. The membrane system was configured with an automatic data logging device, which monitored the system pressures, flows, temperature and feed and filtrate turbidity. Other water quality parameters were monitored using manually collected samples and laboratory analysis. **Figure 4** illustrates the process utilized for the membrane pilot study. Chlorine dioxide, sodium hypochlorite and potassium permanganate were used as the oxidants during the membrane pilot study.

Figure 4 - Microfiltration Pilot Unit



4 Results & Discussion

Figure 5 shows the changes in transmembrane pressure versus time. Due to the changes in water quality observed during the study, no inferences should be drawn upon the fouling effects of each oxidant upon the membrane system.

Figure 5 - Membrane Operating Parameters

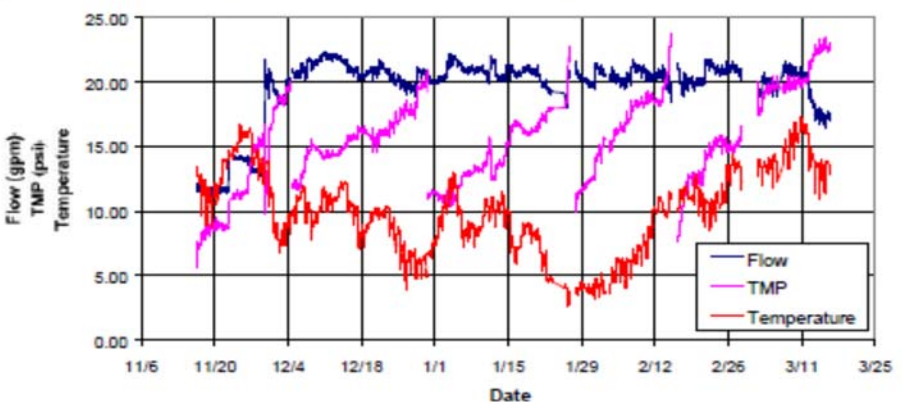


Table 2 illustrates the effectiveness of the clean-in-place regime utilized during the study. The initial membrane resistance was measured as 3.23x1012 m-1. New membranes typically take several weeks of use to adequately wet-out all of the membrane pores. The clean-in place (CIP) performed on December 3rd did not result in adequate recovery of the membrane resistance. Citric acid was added to the CIP regime to remove scales and precipitates of multivalent cations. The subsequent three CIPs resulted in a continued decrease in the membrane resistance to values expected by the manufacturer. This indicates that there is no indication of permanent fouling of the microfiltration membranes.

Table 2: Chemical Clean-in-Place Results

Run	Date CIP Performed	Chemicals Used	Recovery Resistance (m^{-1})
1	03-Dec	1000 mg/L NaOCl	4.28×10^{12}
2	30-Dec	2% citric acid; 1000 mg/L NaOCl	3.7×10^{12}
3	27-Jan	2% citric acid; 1000 mg/L NaOCl	3.04×10^{12}

Table 3
Average Manganese and Iron Removal in Pilot Study

Pre-oxidant	Sample	Manganese, mg/L	Iron (II and III), mg/L
none	Feed	0.166	0.872
	Filtrate	0.129	0.004
Chlorine	Feed	0.140	0.650
	Filtrate	0.100	0.01
Chlorine Dioxide	Feed	0.094	0.84
	Filtrate	0.001	0.00
Permanganate	Feed	0.099	0.90
	Filtrate	0.030	0.02
WTP Average	Finished water	0.027	0.00

5 Conclusions

Jar studies performed at the existing water treatment plant indicated that chlorine dioxide was the best oxidant for manganese removal. Chlorine dioxide provided removal of manganese to less than 0.05 mg/L with less than twenty minutes of reaction time. Based upon the jar tests, a membrane pilot study was performed using a reaction time of twenty minutes. During the membrane pilot study, the application of 0.5 mg/L of chorine dioxide prior to the microfiltration system resulted in the removal of 99% of the manganese. Since microfiltration membranes must be cleaned using an acid as part of the normal operating cycle, any manganese dioxide layer that may assist in the removal of manganese will be removed from the membrane. As a result, the adsorption mechanism can not be relied upon for removal of manganese. Design of membrane facilities for the removal of soluble manganese must include an effective oxidation step to ensure adequate removal. In the pilot study, the microfiltration membrane retained the precipitated manganese colloid.

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